Aggregation and Fragmentation Dynamics of Inertial Particles in Fluid Flows

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Contents

1.	Intro	oductio	n	7
2.	Dyn	amics c	of Particles in Flows	17
	2.1.	Multip	bhase Flows - Suspensions of Particles	17
		2.1.1.	Basic concepts	18
		2.1.2.	Approximations to Describe Particle-Laden Flows	23
	2.2.	Mixed	-Fluid Multiphase Flows: Tracer Particles	25
		2.2.1.	One-Way Coupled Eulerian Mixed-Fluid	25
		2.2.2.	One-Way Coupled Lagrangian Mixed-Fluid	28
		2.2.3.	Two-Way Particle-Flow Coupling for Mixed-Fluids	32
	2.3.	Separa	ted-Fluid Multiphase Flows: Inertial Particles	34
		2.3.1.	Approaches to Calculate the Surface Force and Torque on a Particle	35
		2.3.2.	Theoretical Results: Maxey-Riley and Auton-Hunt-Prud'homme .	37
		2.3.3.	One-Way Particle-Flow Coupling: Dissipative Particle Dynamics .	45
	2.4	2.3.4.	Two-Way Particle-Flow Coupling and Particle-Particle Interactions	50
	2.4.	Conclu	isions	64
3.	Мос	leling C	Coagulation and Breakup of Spherical Droplets	66
	3.1.	Introd	uction	66
	3.2.	Numer	rical Model	68
		3.2.1.	Equations of Motion	68
		3.2.2.	Coagulation	69
		3.2.3.	Breakup	69
	3.3.	Simula	ation Results	72
		3.3.1.	Fluid Flow	72
		3.3.2.	Model parameters and mean sizes	74
		3.3.3.	Approach to a Steady State	75
		3.3.4.	Droplet Size Distributions in the Steady State	76
	~ .	3.3.5.	Influence of Droplet and Flow Properties	78
	3.4.	Rate E	Equation Approach for Coagulation and Fragmentation	86
		3.4.1.	Equations for the Moments of the Droplet Size Distribution	86
		3.4.2.	Relative Growth Rates	87
	05	3.4.3. D'	Estimating a Scaling Relationship	89
	3.5.	Discus	sion	91
4.	Lagi	rangian	Modeling of Fractal-Like Aggregates	95
	4.1.	Introd	uction	95
	4.2.	Model	ing Approach	97
		4.2.1.	Lagrangian Modeling for Fractal-Like Aggregates	97
		4.2.2.	Fragmentation Model	101

		4.2.3.	Fluid Flow	105
		4.2.4.	Implementation	106
	4.3.	Simula	tion Results	108
		4.3.1.	Measured Quantities	109
		4.3.2.	Approach to a Steady State	110
		4.3.3.	Influence of Aggregate Strength	112
		4.3.4.	Influence of the Volume Fraction	113
		4.3.5.	Influence of the Fractal Dimension	115
	4.4.	Discus	sion	117
		4.4.1.	Limitations of the Model	117
		4.4.2.	Summary	120
5.	Rate	e Equat	ions for Fractal-Like Aggregates	122
	5.1.	Introd	uction	122
	5.2.	Fracta	Aggregates and Critical Shear	124
	5.3.	Momen	nts of the Size Distribution	125
	5.4.	Relativ	ve Growth Rate	127
	5.5.	Collisio	on and Fragmentation Kernels	128
	5.6.	Steady	State Solutions	132
	5.7.	Discus	sion	136
6.	Disc	rete Ele	ement Modeling of Fragmentation	138
6.	Disc 6.1.	Introd	ement Modeling of Fragmentation	138 138
6.	Disc 6.1. 6.2.	Introdu Discret	ement Modeling of Fragmentation action	138 138 140
6.	Disc 6.1. 6.2.	Introdu Discret 6.2.1.	ement Modeling of Fragmentation action	138 138 140 141
6.	Disc 6.1. 6.2.	Erete Ele Introdu Discret 6.2.1. 6.2.2.	Ement Modeling of Fragmentation action	138 138 140 141 144
6.	Disc 6.1. 6.2.	Erete El Introdu Discret 6.2.1. 6.2.2. 6.2.3.	ement Modeling of Fragmentation action	138 138 140 141 144 145
6.	Disc 6.1. 6.2.	Crete El Introdu Discret 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula	ement Modeling of Fragmentation action action ce Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation tion	138 138 140 141 144 145 146
6.	Disc 6.1. 6.2.	Crete El Introdu Discrete 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula	ement Modeling of Fragmentation action be Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation tion Results	138 138 140 141 144 145 146 148
6.	Disc 6.1. 6.2. 6.3.	Crete El Introdu Discret 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.2.2	ement Modeling of Fragmentation action be Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation Critical Behavior Superspiritical Packavier	138 138 140 141 144 145 146 148 148
6.	Disc 6.1. 6.2. 6.3.	Crete El Introdu Discret 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss	ement Modeling of Fragmentation action be Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation Critical Behavior Supercritical Behavior Sion	138 138 140 141 144 145 146 148 148 153 154
6. 7.	 Disc 6.1. 6.2. 6.3. 6.4. Con 	crete El Introdu Discret 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss	ement Modeling of Fragmentation action action Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation Critical Behavior Supercritical Behavior Sion	138 140 141 144 145 146 148 148 153 154 157
6. 7.	 Disc 6.1. 6.2. 6.3. 6.4. Con 	crete El Introdu Discret 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss clusions	ement Modeling of Fragmentation action be Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation Critical Behavior Supercritical Behavior Sion and Outlook	138 138 140 141 144 145 146 148 153 154 157
6. 7. A.	 Disc 6.1. 6.2. 6.3. 6.4. Con Inco A 1 	crete Ele Introdu Discrete 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss clusions	ement Modeling of Fragmentation action be Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation tion Results Critical Behavior Supercritical Behavior sion and Outlook	 138 138 140 141 144 145 146 148 153 154 157 165 167
6. 7. A.	 Disc 6.1. 6.2. 6.3. 6.4. Con Inco A.1. A 2 	crete Ele Introdu Discrete 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss clusions mpressi Reprodu Calcula	ement Modeling of Fragmentation action be Element Modeling of Fractal-Like Aggregates Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Implementation tion Results Critical Behavior Supercritical Behavior sion and Outlook ble, Synthetic Turbulence using a Spectral Approach hucing an Energy Spectrum ating Mean Variance and Spatial Correlations of the Flow	 138 138 140 141 144 145 146 148 153 154 157 165 167 171
6. 7. A.	 Disc 6.1. 6.2. 6.3. 6.4. Con Inco A.1. A.2. 	crete Ele Introdu Discrete 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss clusions mpressi Reprod Calcula	ement Modeling of Fragmentation action action Generating Initial Aggregates Particle-Particle Interactions Fluid Forces Fluid Forces Implementation tion Results Critical Behavior Supercritical Behavior sion and Outlook ble, Synthetic Turbulence using a Spectral Approach hucing an Energy Spectrum ating Mean, Variance and Spatial Correlations of the Flow	 138 138 140 141 144 145 146 148 153 154 157 165 167 171
б. 7. А. В.	 Disc 6.1. 6.2. 6.3. 6.4. Con Inco A.1. A.2. Para 	crete Ele Introdu Discrete 6.2.1. 6.2.2. 6.2.3. 6.2.4. Simula 6.3.1. 6.3.2. Discuss clusions mpressi Reprod Calcula	ament Modeling of Fragmentation action	 138 138 140 141 144 145 146 148 153 154 157 165 167 171 175

Zusammenfassung

Die Dynamik von Inertialpartikeln in Strömungen ist von großem Interesse in vielen Gebieten, von Atmosphärenphysik bis hin zu chemischer Verfahrenstechnik oder marinen Systemen. In dieser Arbeit wird numerisch die Dynamik von Inertialpartikeln untersucht, die durch Kollisionen aggregieren und unter bestimmten Bedingungen wieder fragmentieren. Unser Ziel ist vor allem ein besseres Verständnis natürlicher Phänomene wie zum Beispiel die Bildung von Tropfen in Wolken und mariner Aggregate.

Das in dieser Arbeit vorgestellte partikelbasierte Aggregations- und Fragmentierungsmodell bildet eine Brücke zwischen Molekulardynamik-Simulationen und den oft verwendeten Ratengleichungen. Molekulardynamik-Simulationen können zwar sehr detailliert sein, erfordern aber im allgemeinen einen hohen Rechenaufwand während Ratengleichungen für große Systeme anwendbar sind, aber viele Approximationen benötigen.

Unser Schwerpunkt ist die Untersuchung des Langzeitverhaltens von Größenverteilungen von Partikeln. Als erstes beschreiben wir ein Modell für die Aggregation und Fragmentierung von kugelförmigen Tropfen in einer turbulenten Strömung. Während frühere Arbeiten meistens die Rolle von Aggregationswahrscheinlichkeiten betonen, zeigen unsere Resultate, dass Fragmentierung der wichtigste Prozess für das Langzeitverhalten ist.

Ferner analysieren wir Systeme mit komplexerer Aggregatsruktur, wo die Struktur mit einer fraktalen Dimension approximiert werden kann. Dies ist zum Beispiel der Fall für marine Aggregate. Wir zeigen, dass die Verteilung der Fragmente nach dem Zerbrechen entscheidend für die Form der Größenverteilung der Aggregate ist. Allerdings ändern sich sowohl die mittlere Größe als auch die Zeit zum Erreichen des Langzeitzustandes mit den Systemparametern, wie zum Beispiel der Aggregatstärke oder dem Turbulenzlevel in der Strömung. Der wichtigste Parameter ist in diesem Fall die fraktale Dimension.

Schließlich zeigen wir wie sich unser Ansatz in die etablierten Modelle aus der Literatur einordnet. Die geeignete Konstruktion von Aggregations- und Fragmentationsraten führt zu einer korrespondierenden Ratengleichung, was wiederum Ausdrücke für die mittlere Aggregatgröße liefert. Desweiteren benutzen wir ein Diskrete-Elemente Modell um die Fragmentierung eines Aggregates in einer Scherströmung zu untersuchen. Wir finden ein Potenzgesetz zwischen kritischer Scherung und Aggregategröße, wobei der Exponent von der fraktalen Dimension abhängt. Ein vergleichbarer Zusammenhang bildet die Basis unseres partikelbasierten Aggregations- und Fragmentierungsmodells.

Abstract

The dynamics of inertial particles in fluid flows is a subject of great interest in many disciplines, from atmospheric science to chemical engineering or marine systems. In this thesis we study numerically the dynamics of inertial particles aggregating upon collision and fragmenting under certain conditions. Our motivation lies primarily in understanding natural phenomena such as the formation of cloud droplets and marine aggregates.

The individual, inertial-particle based approach to aggregation and fragmentation presented in this thesis bridges the gap between molecular dynamics simulations of aggregation-fragmentation systems and the usual rate equation based approaches. Molecular dynamics simulations can be very detailed but are computationally too expensive in most cases while rate equation approaches can be applied to large systems but rely on many approximations.

Our main focus is the study of the size distributions of particles which evolve in the long-term limit. First, we describe a model for the aggregation-fragmentation dynamics of spherical droplets in a turbulent flow. While most previous studies emphasize the role of aggregation probabilities, our results show that in situations where a steady state is of interest, fragmentation will be the most relevant process.

In addition, we show how to model processes where aggregates with a more complex structure appear. We discuss the problem of aggregation and fragmentation in systems where the aggregate structure can be approximated in terms of a fractal dimension. This is for example the case for marine aggregates. The distribution of the fragments after breaking is found to be the main influence on the resulting shape of the steady state size distribution. However, the mean aggregate size as well as the time to reach the steady state depend on the system parameters, such as aggregate strength or turbulence level in the flow. The most important parameter in this case is the fractal dimension.

Finally, we integrate our approach with the established models from the literature. By constructing the corresponding collision and fragmentation rates we show how our inertial particle based approach can be connected to a rate-equation based model, leading to expressions for the steady state aggregate size. Additionally, we use a discrete element model to simulate the fragmentation of an individual aggregate in a shear flow. We find a power-law relationship for the critical shear as a function of aggregate size where the exponent depends on the fractal dimension, similar to the critical shear equation forming the basis of our individual, inertial-particle based model.

1. Introduction

"Well," said Pooh, "we keep looking for Home and not finding it, so I thought that if we looked for this Pit, we'd be sure not to find it, which would be a Good Thing, because then we might find something that we weren't looking for, which might be just what we were looking for, really."

(A.A. Milne, "Tigger is unbounced")

For almost 200 years there has been a great interest in studying the dynamics of particles in fluid flows. The earliest works e.g. by Poisson, dating back at least as far as the first half of the nineteenth century, appeared even before the development of what are now called the Navier-Stokes equations, the basic equations of fluid dynamics. However, this does not mean that everything has been said on this topic. On the contrary, for example for the last 10 years the database of Web of Science (http://isiknowledge.com) lists around 15,000 publications on the topic. The reason for this continued interest in the field is that systems where particles are moving within a fluid are ubiquitous both in nature and in technical applications. Examples include systems from biology, chemistry, physics, oceanography, geo- and astrophysics and engineering, see Tab. 1.1.

While much progress has been made in the last 200 years in understanding these systems, there are still many fundamental questions that have not yet been answered. Open questions exist on all levels of the problem, ranging from the interaction of a single, isolated particle with the surrounding fluid to the description of many-particle systems of arbitrary complexity.

From a theoretical point of view two major directions have emerged. On the one hand the study of point-like 'tracer' particles that directly follow the motion of the surrounding fluid and on the other hand finite-size 'inertial' particles whose motion can deviate from that of the surrounding fluid.

The study of tracer particles, which is nothing but fluid dynamics in a Lagrangian framework, has given many new insights into the behavior of fluid flows. For laminar flows, even in cases where the dynamics of the Eulerian field is extremely simple, the motion of individual tracers or fluid elements can be very complex. It was pointed out by Aref (1984) for non-stationary two-dimensional flows that the advection of tracer particles typically leads to chaotic motion, a phenomenon now called chaotic advection. The motion of the tracer particles can be viewed as a non-integrable Hamiltonian system,

where it is known that a chaotic saddle appears. The stretching and folding of the tracer trajectory along the manifolds of this chaotic saddle leads to a greatly enhanced mixing and stirring in such systems (Chaiken et al., 1986; Aref and Balachandar, 1986; Ottino, 1989).

For large Reynolds number flows, combining methods from statistical physics with the study of the dynamics of tracer particles has led to a new perception of fluid turbulence (Toschi and Bodenschatz, 2009). In particular, using this particle-based viewpoint some progress has been made in understanding one of the major challenges of turbulence theory, the phenomena of intermittency and anomalous scaling in turbulent flows. Statistical invariants have been identified in the evolution of groups of particles, where a symmetry breaking leads to the observed non-universal scaling laws (Celani and Vergassola, 2001; Falkovich et al., 2001). In addition, models have been developed for the statistical geometry of turbulence and the Lagrangian evolution of material lines, vorticity and strain (Pumir et al., 2000; Guala et al., 2005; Lüthi et al., 2005). Additionally, much effort has gone into establishing the connection between the Lagrangian and Eulerian statistics of a turbulent flow, i.e. between the velocity and acceleration statistics along a tracer trajectory and those of the surrounding fluid. Numerically, acceleration statistics have been measured for example by Biferale et al. (2004); Biferale and Toschi (2005) and a comparison between experimental results and models is found in Mordant et al. (2004). Recently, the study of Lagrangian structure functions, i.e. temporal correlations of the velocity has given new insights into intermittency and universality in turbulence (Arneodo et al., 2008; Biferale et al., 2008).

Type of Particle-Fluid System	Application
solid particles in a gas	aerosols, snow, hail, protoplanetary discs, clean
	rooms, solid rockets
liquid droplets in a gas	water droplets in air, rain, liquid sprays, plasma
	sprays
solid particles in a liquid	plankton, bacteria, marine aggregates and sedi-
	ment, colloids, powders, solidification, liquid fil-
	ters
gas bubbles in a liquid	bubble columns, molten lava, boilers, naval ves-
	sels

 Table 1.1.:
 Examples for some of the most common types of systems where particles suspended in a liquid appear.

Inertial particles in fluid flows have lately been subject of increasing interest in several disciplines from dynamical systems (Benczik et al., 2006; Vilela and Motter, 2007; Zahnow and Feudel, 2008) to atmospheric science (Shaw, 2003; Jaczewski and Malinowski,

2005; Falkovich and Pumir, 2007) and turbulence (Wilkinson and Mehlig, 2005; Bec et al., 2005; Calzavarini et al., 2008). In particular tools from dynamical systems theory, such as the concepts of attractors, dimensions and Lyapunov exponents have been found to be very useful in tackling these problems. The dynamics of these particles is dissipative, which leads to a behavior that is very different from tracer particles. The acceleration of the inertial particles leads to a detachment from the fluid trajectories and to a preferential accumulation in certain regions in space, i.e. on attractors. This 'unmixing' phenomenon, where an initially uniformly distributed ensemble of particles approaches a spatially inhomogeneous distribution (Maxey, 1987; Wilkinson et al., 2007) has significant consequences for many problems, for example leading to locally enhanced collision rates. In addition, the detachment of the particle trajectories from the fluid trajectories can lead to the appearance of so-called 'caustics' (Falkovich et al., 2002; Wilkinson and Mehlig, 2005) where the particle attractor in phase space is folded and particles with very different velocities occupy the same region of space. While such phenomena appear naturally in the Lagrangian, particle-based viewpoint it has so far proven impossible to formulate a corresponding Eulerian theory, precisely because in this case the particle velocities no longer meet the definition of a velocity 'field'.

In addition, much effort both from theoretical and experimental side has gone into determining the connection between the statistics of the particle motion, along the trajectory of an inertial particle and the underlying turbulent velocity field of the fluid. Numerically, acceleration statistics have for example been obtained by Bec et al. (2006a); Cencini et al. (2006) or Ayyalasomayajula et al. (2008) and a comparison with experimental data can be found in Ayyalasomayajula et al. (2006). Generally, the results depend significantly on the particle size, or more precisely on the particle Stokes number (see Chapter 2.2.3 of this work for a definition) and on the particles density ratio with respect to the surrounding fluid, i.e. whether particles are heavier or lighter than the surrounding fluid.

In most of these works a dilute regime is assumed, where particle collisions can be neglected. Some authors keep track of particle collisions numerically without considering the outcome of a collision, for example to calculate collision rates (Wang et al., 2000; Bec et al., 2005). However, in many interesting applications, for example the growth of cloud droplets (Pruppacher and Klett, 1997) interactions of inertial particles due to collisions can be very important. It is well known that as a result of collisions between particles, *aggregates* can be formed that consist of a large number of smallest (primary) particles. In the literature one finds both the terms 'aggregates' and 'coagulates' or 'aggregation' and 'coagulation' but the difference between the two is not always clear. In this work we reserve aggregates for solid particles that stick to form a cluster of connected, but still individual particles¹ whereas fluid particles, e.g. water droplets coagulate, meaning they merge upon collision to form one new larger particle. For simplicity, when describing something that applies to both solid and liquid particles we use the term aggregate.

In many areas of science the formation of such aggregates and their *fragmentation* due to forces from the surrounding fluid plays a very important role, e.g. in sedimentation of particles in oceans and lakes (Winterwerp, 1998), chemical engineering systems such as solid-liquid separation (Spicer and Pratsinis, 1996a; Bäbler et al., 2008), aggregation of marine aggregates (Thomas et al., 1999) and flocculation of cells (Han et al., 2003).

Previously, this has mainly been studied using a rate equation based approach, in the framework of which one treats the problem of particle motion as a field equation. The Smoluchowski equation (Smoluchowski, 1917) is then used to model aggregation and fragmentation of these particle concentration fields, instead of individual particles. Such an approach exhibits a number of problems. For example, as already mentioned above the particle velocity may take on several values even at the same location of inertial particles when the dynamical attractor of the particles folds in the full velocity-position phase space. Thus, a field approach for inertial particles cannot be well founded and relies on many assumptions and parameterizations. In addition, determining correct collision and fragmentation rates for such a model approach usually involves many approximation steps and often a certain amount of guesswork. Much effort has gone into deriving collision rates for inertial particles (Bec et al., 2005; Ayala et al., 2008b,a) but so far only approximative solutions exist. Both the preferential concentration of inertial particles and the appearance of caustics lead to significant changes in the collision rates that have not been fully understood. Similarly, determining the fragmentation rates requires knowledge of the microscopic properties of the aggregates, such as the bond structure and its response to applied shear forces. However, in many cases this information is not available and one has to resort to approximations or even if the information is available no sufficiently simple equation for the rate can be found.

Due to the increasing availability of computational resources there has been much progress in developing models capable of directly simulating the dynamics as well as the aggregation and fragmentation behavior of inertial particles in a fluid. Some examples include applications of the lattice Boltzmann method for inertial particles (Ten Cate et al., 2004; Feng et al., 2007) and semi-resolved (Lomholt and Maxey, 2003) and resolved surface methods for multiphase flows using molecular dynamics models for each particle, e.g. with a finite element algorithm in an arbitrary Lagrange Eulerian framework (see e.g. Maury, 1999). Such approaches offer a promising alternative for the future as they can be

¹Sometimes, in particular in a biological context, the term 'floc' or 'flocculation' is also used for this.

very detailed and do not rely on many assumptions or approximations. However, because of the greatly increased numerical effort they are so far restricted to a few hundred or thousand particles (Higashitani et al., 2001; Zeidan et al., 2007). Currently, they are therefore not a feasible alternative to the established rate equation approaches, but can still serve as a basis of establishing more detailed models for example for the fragmentation of an individual aggregate.

In Zahnow et al. (2008, 2009) we therefore proposed and discussed a model for aggregation and fragmentation based on inertial particle dynamics, using a point-force approximation for the equation of motion of the particles (Maxey and Riley, 1983) to help bridge the gap between the full hydrodynamic simulations and the usual rate equation based approaches. The idea of this model was to treat each aggregate as an individual, inertial particle advected in the surrounding fluid flow but neglecting the internal structure of the aggregate. Instead, only the number of primary particle per aggregate and some measure of the overall aggregate size is kept. The complexity and numerical effort of this approach lies between that of the detailed hydrodynamical models and the much simpler rate equation approach. With this approach we were able to simulate aggregation and fragmentation processes in systems consisting of $10^5 - 10^7$ primary particles on a standard desktop PC. Such an approach has the advantage that the dynamics of the inertial particles are taken directly into account and therefore no approximations for example for the collision rates are required. Instead, particle collisions are calculated directly within the model leading to a much more detailed description. For fragmentation, results from detailed hydrodynamical simulations of aggregate fragmentation can directly be included as a property of the individual aggregates and therefore the fragmentation process is linked directly to the physical properties of the aggregates.

In our earlier work (Zahnow et al., 2008, 2009) we applied our model approach to the aggregation and fragmentation of spherical droplets in chaotic advection. This is not included in the main part of this thesis because here we focus on a slightly different question, namely turbulent flows. However, in the following we briefly summarize the main results for the sake of completeness. The full text of these works can be found in Appendix C.

We showed that such an individual, inertial particle based approach is able to capture the basic properties of aggregation and fragmentation processes. We found that the combination of aggregation and fragmentation leads to the development of a dynamic steady state, where aggregation and fragmentation balance each other. In this steady state there exists a distribution of aggregates of various sizes, each moving according to the same equations of motion but with different parameters depending on their size. The shape of this distribution was found to be different for different fragmentation mechanisms but did not depend strongly on the underlying flow field as long as sufficient mixing occurred. Additionally, when varying system parameters such as the total number of particles in the system or the binding strength of the aggregate the distribution was shifted but not changed qualitatively. In particular, rescaling the distributions for different parameters by dividing with the mean aggregate size we found that all distributions collapse onto a single 'master' curve. Similar universal size distributions have been found as solutions of the Smoluchowski equation (Spicer and Pratsinis, 1996a).

Even though to a certain extent methods from dynamical systems theory can usefully be applied, the entire problem is much more complex than that of any usual dynamical system. While particles of a single size move on specific attractors, aggregation and fragmentation lead to repeated transitions from one attractor to another one, depending on the aggregate size. The skeleton of the new dynamics is therefore a superposition of the different attractors, with transient motion in between. The structure of the individual attractors and their superposition in turn influence the aggregation probabilities due to different local concentrations of particles. Fragmentation is also affected by the particle dynamics, because shear forces can be locally different in the flow. Therefore, break-up may depend on whether an attractor for a certain particle size lies in a region with high shear or not.

In the present thesis we now discuss the application of this individual, inertial particle based model to turbulent flows. Due to the universal nature of turbulent flows, at least away from any boundaries this is a much more general setting than systems with chaotic advection. We show how the resulting steady state size distribution depends on the particle and flow properties. For many problems, such as the prediction of rain formation in wet clouds for weather forecasts, it is of significant interest to understand how the growth of the cloud droplets depends on properties such as the initial concentration of smallest droplets or the turbulence level in the cloud.

In addition, we extend the model that was initially designed for spherical particles to include processes where aggregates with a more complex structure appear. While the assumption of spherical particles was a reasonable approximation for fluid particles, such as water droplets in clouds, the aggregation of solid particles is generally more complex. We discuss the problem of aggregation and fragmentation in systems where the aggregate structure can be approximated as being *fractal-like*. By this we mean that when averaged over an ensemble of particles there exists a power-law relationship between the characteristic length and the mass of such aggregates. The exponent of the power-law is called the *fractal dimension*. Such an relationship is for example found for cohesive sediment (Kranenburg, 1994) or marine aggregates (Logan, 1999), but also in many other systems. In the context of a rate equation based approach, a complex particle structure has been incorporated in the past in terms of a density modification for the particles, e.g. by Kranenburg (1994) or Maggi et al. (2007). However, so far there are almost no attempts to treat this problem for inertial particles in a flow. We therefore discuss how such a complex structure can be approximated within the framework of our individual particle-based approach. We examine the steady state size distribution of the aggregates, as well as the time to reach the steady state and the dependence on particle and flow properties, such as the fractal dimension. For many problems the distribution of the aggregates and in particular their mean size is of great interest. An example are marine aggregates where the size of the aggregates affects their settling velocities and therefore the transport of organic carbon from the ocean surface to the bottom layers.

Since rate-equation based models are still the most common tool for the modeling of aggregation and fragmentation processes, partly because they can easily be applied to very large systems, we discuss how to translate between our approach and a rateequation based model. We show how by constructing the corresponding collision and fragmentation rates our individual, inertial particle based approach can be connected to the rate-equation approach. Additionally, we examine some aspects of such a rateequation approach, such as the (analytical) calculation of a steady state solution for the average aggregate size and emphasize the close connection between such solutions and the details of the fragmentation process.

Finally, we examine the fragmentation process in more detail, using a discrete element model to simulate the fragmentation of an individual aggregate in a shear flow. Because the steady state aggregate size distribution depends strongly on the fragmentation mechanism it is important to obtain a deeper understanding of this process. For many systems details of this are still unknown. Detailed numerical models can help bridge the gap between the understanding of the microscopic properties of the individual, primary particle in the aggregate and the macroscopic properties of the whole aggregate. This is needed for the correct formulation of less detailed models that can be applied to larger systems, such as our individual-particle based approach or rate-equation based approaches.

The thesis is structured as follows. In Chapter 2 we give a general introduction into the theoretical description and modeling of suspension of particles in fluid flows. The focus here is on different approximations for the description of the particle phase, starting with simple Eulerian and Lagrangian tracer models up to complex models for inertial particles. The focus of this section is on the discussion of the details of the point-force approximation for the motion of individual, inertial particles that forms the basis of the following work. In particular, the description of the different components of the surface force is discussed with an eye on their relevance for the overall particle motion. Additionally, some results from the literature for the dynamics of particles in a fluid flow are reviewed and discussed,

such as preferential concentration and unmixing and the appearance of caustics in the particle dynamics. Finally, we focus on the calculation of collision rates between inertial particles in a turbulent fluid flow. In particular, we numerically examine the question which terms of the equations of motion for the particles could possibly be neglected without leading to significant changes in the frequency of collisions between particles.

In Chapter 3 we present the application of our individual, inertial particle based model to the problem of coagulation and fragmentation of spherical droplets in a synthetic turbulent flow. Such synthetic turbulent flows are a fast and simple way to create a flow which captures certain characteristics of real turbulence. In our case we recreate the dissipative range of a turbulent flow with the correct energy spectrum. We discuss fragmentation due to two different mechanisms. On the one hand we introduce a sizelimited fragmentation mechanism which is inspired by the hydrodynamical instability and subsequent breaking of water drops in air settling due to gravity. On the other hand we discuss shear fragmentation, where a droplet is broken up due to hydrodynamic shear forces in the fluid. Both mechanisms lead to a steady state size distribution but the shape of the distribution is very different for the two cases. In addition, whereas for size-limiting fragmentation the mean coagulate size in the steady state only depends on the maximum stable coagulate size, for shear fragmentation the mean coagulate size changes depending on the particle and flow properties. Numerically, we find scaling relationships for the mean coagulate size in the steady state as a function of the system parameters, where the scaling exponent seems to depend on the details of the shear fragmentation model. We discuss a simplified analytical calculation that shows how in principle this dependence may arise.

Chapter 4 discusses the extention of our individual, inertial particle-based approach for aggregation and fragmentation to systems of particles with a fractal-like structure in a synthetic turbulent flow. We show how to incorporate this fractal-like structure into our model description in terms of a fractal dimension for the aggregates. A characterization with a fractal dimension leads to a modification of the radii and effective densities of the aggregates compared to a solid sphere of the same mass. Nevertheless, we still treat them as effectively spherical for the particle motion, allowing us to apply the same point-force approximation for the equations of motion (Maxey and Riley, 1983) but with modified parameters. Within this framework we discuss different splitting mechanisms for shear fragmentation of the aggregates by comparing different distributions of the fragments created after breaking. We compare large-scale fragmentation, where fragments of similar size are created, erosion, where fragments of very different size appear and uniform fragmentation as a simplified combination of the two others, where all fragment sizes appear with the same probability. This distribution of the fragments is found to be the main influence on the resulting shape of the steady state size distribution. However, the mean aggregate size as well as the time to reach the steady state depend on the system parameters, such as aggregate strength, number of particles and turbulence level in the flow. For this, the most important parameter is the fractal dimension as a measure of the aggregate structure, which significantly affects both the aggregation and the fragmentation process. With increasing fractal dimension both the relaxation time towards the steady state and the mean aggregate size in the steady state increase.

In Chapter 5 we present a rate-equation based approach for fractal-like aggregates. The main idea here is to illustrate how to connect our individual, inertial particle based approach with the usual rate-equation formulation for aggregation and fragmentation. This is for example necessary if one wants to compare results from these different approaches. While many expressions for the collision rate exist in the literature, finding an expression for the fragmentation rate that corresponds to the individual particle based approach is somewhat more involved. The key here is how to translate the concept of the critical shear, which is the individual aggregate property used to locally determine fragmentation events within the particle-based approach into a corresponding global fragmentation rate for the whole ensemble of aggregates. In this context we argue that the usual assumption of a Gaussian distribution of the shear forces which is used in many of the fragmentation rates found in the literature may lead to a significant under-estimation of the fragmentation rate. While the analytical calculations carried out in this Chapter rely on many approximations, for example simplified expressions for the fragmentation rates they serve to illustrate in principle how a steady state solution can be calculated. We show how in this case the steady state depends on the properties of the particles and the surrounding flow field as well as under which conditions the steady state solution is stable, i.e. whether the system will in fact converge to this solution. It turns out that the stability only depends on the fractal dimension and the shape of the critical shear condition. Other properties such as aggregate strength or turbulence level do not play a role. While in more realistic cases no analytic calculation will be possible and one will have to resort to numerical methods to obtain a solution, we believe that the principle remains the same and similar results would be found.

In Chapter 6 we show how to use a detailed, discrete-element simulation of an individual aggregate fragmenting in a laminar shear flow to obtain information about the connection between the microscopic particle properties such as the bond structure within the aggregate and the critical shear that is needed for the inertial particle-based model. Aggregates are build using a diffusion-limited aggregation process and then a model for the central and non-central interaction between the primary particles is described. The particles are subjected to a shear flow and the shear force is varied until the aggregate breaks. On the one hand we determine how the critical shear required for fragmentation changes with the aggregate size. On the other hand we show that for supercritical shear, i.e. shear forces above the critical point the size of the fragments is on average independent of the original aggregate size and instead only depends on the level of the shear.

Finally, Chapter 7 contains conclusions and an outlook on possible further research directions.

2. Dynamics of Particles in Flows

This chapter gives a general introduction into the modeling of suspensions of particles in fluid flows and provides a brief review of the relevant literature, focusing on the concepts relevant for this thesis. Additionally, some numerical calculations of collision rates of inertial particles in synthetic turbulence are presented.

2.1. Multiphase Flows - Suspensions of Particles

In the following chapters basic methods and results for multiphase flows are summarized, following partly the work of Loth (2010). While the selection of topics is certainly subjective and many important aspects are only mentioned very briefly or may be missing completely it should provide a brief introduction to the dynamics of particle suspensions in flows.

The term multiphase flows generally refers to the fluid dynamics of systems which contain multiple phases, but here, as in many applications means the more specific case of suspensions of individual particles in a fluid. Such a *particle-laden flow* consists of a *fluid phase* and a so-called *dispersed phase*, the collection of all the particles in the flow. If all the particles of the dispersed phase are identical, the system is called a *monodisperse suspension*, whereas systems with particles with different properties, for example different densities, are called *polydisperse suspensions*.

The most common examples of multiphase flows are solid particles in a liquid or gas, liquid particles in a gas and gas bubbles in a liquid. However, also mixtures of two immiscible liquids are often considered in a similar framework, if one of the liquids occurs in the form of isolated drops. In this context immiscible means that there is no molecular mixing at the interface between the particle and the surrounding fluid so that the particle mass does not change over time.

Generally, the full equations of motion describing such a particle-laden flow are too complicated or computationally expensive to solve fully and depending on the problem different approximations, such as mixed-fluid multiphase flow or point-force inertia particles are used. Such approximations and some results for the dynamics of these systems are discussed in the following chapters.

Typically, the particles are assumed to be much smaller than the overall domain and dispersed within the surrounding fluid, which means that the particles are not heavily concentrated. In this case the particle motion is mainly determined by the interaction of the particles with the fluid as opposed to particle-particle interactions, for example in form of collisions. The level of the coupling between the dispersed and the fluid phase depends on the concentration of the particles, the different cases are discussed in more detail in the following parts. In particular, one distinguishes between *one-way particle-flow coupling*, *two-way particle-flow coupling* and *particle-particle interactions*. For one-way particleflow coupling one assumes that particle concentrations are so low that their feedback on the surrounding flow can be neglected. In this case the equations of motion for the fluid phase can be solved independently from the equations of motion for the particles. For two-way particle-flow coupling the feedback of the particles on the surrounding flow is taken into account, for example by modifying the local density and viscosity. The equations for the different phases are now coupled. Particle-particle interactions include both interactions between particles through the fluid and direct interactions in the form of collisions. This is typically important for (locally) large particle concentrations.

2.1.1. Basic concepts

In this work the properties of the different flow phases are distinguished by subscripts. Properties of the fluid phase will be denoted by the subscript "f", for example the dynamic viscosity of the fluid will be μ_f . The properties of the particles, i.e. of the dispersed phase will be denoted by the subscript "p", for example the mass of a particle will be m_p .

The Dispersed Phase

To quantify the particle size, the diameter d_p of the particle is used. For spherical particles this is an unequivocal measure of the particle size, whereas for particles with non-spherical shapes the situation is more complicated. In many realistic cases it is not clear whether a single length scale is enough to characterize different particle sizes. However, in most cases it is assumed that non-spherical particles can be described by an equivalent particle diameter, which is the diameter of a spherical particle that would give the same behavior in a system of interests. One of the most common forms of an equivalent particle diameter is the volumetric diameter, which is the diameter of a spherical particle diameter are also often found (see table 2.1). For example, for the description of fractal-like aggregates in part 4, the combination of a characteristic length scale and a fractal dimension will be used to characterize particle sizes.

Equivalent Particle Diameter	Description
Volumetric Diameter	Diameter of a sphere with the same volume
Circular Diameter	Diameter of a circle with the same area (typi-
	cally used in 2-d projections)
Aerodynamic Diameter	Diameter of a sphere of unit density with the
	same terminal settling velocity
Mobility Diameter	Diameter of a sphere with the same (electrical)
	mobility, i.e. the same ratio of particle velocity
	to applied force
Scattering Diameter	Diameter of a spherical (often a polystyrene la-
	tex) particle with the same light-scattering sig-
	nal

 Table 2.1.: Different types of equivalent particle diameter that are commonly used to characterize the size of non-spherical particles.

Together with the particle volume V_p , the particle mass m_p defines the particle density

$$\rho_p = m_p / V_p \ . \tag{2.1}$$

If the particle is either a liquid or a gas, its dynamic viscosity μ_p and surface tension σ_p can also be important, as these determine the behavior of the fluid inside the particle and the shape of the particle in response to stresses in the surrounding fluid.

The position of a particle at time t will be denoted by the position $\mathbf{X}(t) \in \mathbb{R}^3$ of the center of gravity of the particle. The Lagrangian translational velocity and angular velocity of a particle will be written as $\mathbf{V}(t) \in \mathbb{R}^3$ and $\mathbf{\Omega}(t) \in \mathbb{R}^3$, respectively with a corresponding moment of inertia I_p .

The Fluid Phase

In the case where the particle diameter is much smaller than the mean free path of the fluid molecules, the molecules colliding with the particle usually only interact with other molecules of the fluid far from the particle surface. Then the Knudsen number Kn, which is the ratio between the mean free path of the molecules in the surrounding fluid and the particle diameter d_p is $\gg 1$, see e.g. Friedlander (2000). This means that most of the molecules hitting the particle come from the main body of the fluid and are not affected by the presence of the particle. The interaction between fluid and particle can then be determined from molecular collision theory. This is the so-called free molecular range. An example for this is the motion of micrometer-sized particles, for example small dust particles in a highly rarefied gas, where the mean free path can be of the order of 1 cm,

resulting in $Kn \approx 10^3$.

In the case where the particle diameter is much larger than the mean free path of the molecules in the fluid, i.e. $Kn \ll 1$ the molecules that collide with a particle are strongly influenced by those molecules just leaving the surface of the particle. In this case the fluid behaves as a continuum, with a no-slip boundary condition at the particle surface (for solid particles). In this so-called continuum range the motion of the surrounding fluid is then determined by the Navier-Stokes equations (Landau and Lifschitz, 1991).

The transition between the continuum range and the free molecular range takes place continuously with increasing Knudsen number, but the transition theory still poses many problems. Here, we focus on the case where the surrounding fluid can be treated as a continuum, i.e. where the Knudsen number $Kn \ll 1$. In this case the fluid phase is also called the *continuous phase*. All of the relevant applications in this work fall in this category, for example the motion of sediment particles in the ocean or raindrops in air. For raindrops in air, the mean free path at normal temperature and pressure is approximately 0.065 μ m and the size of the smallest water droplets is of the order of 10 μ m, leading to $Kn < 10^{-2}$ which is well within the continuum range.

For the velocity field of the surrounding fluid it is often convenient to distinguish between the undisturbed flow $\boldsymbol{u}(\boldsymbol{x},t) \in \mathbb{R}^3$ without the presence of the particles and the real flow $\boldsymbol{v}(\boldsymbol{x},t) \in \mathbb{R}^3$ which includes the effect of the presence of the dispersed phase. Both flows are assumed to be incompressible and isothermal with constant density ρ_f , dynamic viscosity μ_f and a characteristic length scale \check{L}_f and velocity scale \check{U}_f , which can for example be the Kolmogorov scales in a turbulent flow. The characteristic length scale and velocity scale together define a characteristic time scale $\check{T}_f = \check{L}_f/\check{U}_f$ of the flow. The size of the system, which can be for example the distance between the walls in a plane couette flow or the size of a periodic box, if periodic boundary conditions are assumed is denoted by L. It is generally assumed that $L \gg d_p$, i.e. particles are much smaller than the largest length scale in the system.

Density and Viscosity Ratios

To characterize particles much heavier or lighter than the surrounding fluid a dimensionless density parameter β is introduced as

$$\beta = \frac{3\rho_f}{\rho_f + 2\rho_p} \ . \tag{2.2}$$

The reason for this particular definition will become apparent in Sec. 2.3.2, where it turns out that this is the form in which the density ratio appears in the equations of motion for the particles. Particles much heavier than the fluid are characterized by $\dot{\beta} \to 0$, which typically includes droplets and solid particles in a gas. Such particles tend to fall in the direction of gravity.

Very buoyant particles, i.e. those much lighter than the surrounding fluid are characterized by $\beta \rightarrow 3$. The most important example of this are bubbles in a liquid, which tend to rise upwards in a flow.

Particles with $\beta = 1$ are neutrally buoyant and neither rise nor sink in a flow. This can for example be the case for solid particles in a liquid.

If the particles are either gas or liquid, the ratio of particle and fluid viscosity

$$\check{\mu} = \frac{\mu_p}{\mu_f} \tag{2.3}$$

is another important quantity, that for example influences the shear fragmentation of liquid drops (Sec. 3.2.3). Liquid drops in a gas correspond to $\check{\mu} \gg 1$, whereas gas bubbles in a liquid are in the range $\check{\mu} \ll 1$. A viscosity ratio of order unity is often the case for liquid drops suspended in an immiscible liquid.

Full Set of Equations

Next, we write down a full set of equations for a system of particles dispersed in a fluid. Since in most cases this set of equations is extremely difficult and computationally expensive to solve, in the following the aim will then be to find suitable approximations or models for this full set of equations.

The flow field \boldsymbol{v} is described by the Navier-Stokes equations

$$\rho_f \left(\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v} \cdot \nabla) \boldsymbol{v} \right) = \rho_f \boldsymbol{g} - \nabla p + \nu_f \nabla^2 \boldsymbol{v} , \qquad (2.4)$$

where g is the gravity vector and the continuity equation for an incompressible flow with constant density

$$\nabla \cdot \boldsymbol{v} = 0 , \qquad (2.5)$$

combined with suitable boundary conditions at the system boundaries. In addition to the boundary conditions at the system boundaries there exist additional boundary conditions for the flow field v at the surface of the particles. For the case of solid particles this is typically a no-slip boundary condition, for spherical particles this would read as

$$\boldsymbol{v}(\boldsymbol{x},t) = \boldsymbol{V}(t) + \boldsymbol{\Omega}(t) \times [\boldsymbol{x}(t) - \boldsymbol{X}(t)], \quad \text{for } ||\boldsymbol{x}(t) - \boldsymbol{X}(t)|| = r_p.$$
(2.6)

For uncontaminated viscous particles, i.e. drops and bubbles the boundary condition is instead often given by a no-stress condition at the surface of the particles. Generally, the evaluation of the boundary condition for viscous particles may require one to solve an additional Navier-Stokes equation for the fluid inside of the particle since the external forces may induce flows inside of the particle.

Due to the forces acting on them the particles move around in the fluid, implying moving boundaries for the equations for the fluid phase. The translational equations of motion for the dispersed phase are given by Newton's second law, where the forces acting on the particle are generally the sum of body (or volume) forces F_{body} , surface forces F_{surf} and collision forces F_{coll}

$$\frac{d\boldsymbol{X}}{dt} = \boldsymbol{V}(t) \tag{2.7}$$

$$m_p \frac{d\mathbf{V}}{dt} = \mathbf{F}_{\text{body}} + \mathbf{F}_{\text{surf}} + \mathbf{F}_{\text{coll}} .$$
 (2.8)

The body forces are typically fairly straightforward and can for example include gravitational or electromagnetic forces. In this work only gravity is considered as a body force, it is therefore $\mathbf{F}_{\text{body}} = m_p \mathbf{g}$, where \mathbf{g} is the gravity vector. The collision forces can usually be regarded as instantaneous and discontinuous and can for example be obtained from elastic or inelastic collision models (e.g. Pöschel and Schwager, 2005). The surface force is given by the integral of the fluid stress tensor

$$\sigma_{ij} = -\rho_f \delta_{ij} + \nu_f \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right)$$
(2.9)

over the surface S of the particle

$$\boldsymbol{F}_{\text{surf}} = \oint_{S} \sigma \boldsymbol{n} dS , \qquad (2.10)$$

where n is the unit vector perpendicular to the particle surface.

The rotational equations of motion for a particle are given by a torque balance, where the torques acting on a particle are generally the sum of body torques for example due to inhomogeneous density inside of the particle, surface torques from the fluid and interparticle torques, generated during collisions

$$I_p \frac{d\mathbf{\Omega}}{dt} = \mathbf{M}_{\text{body}+} \mathbf{M}_{\text{surf}} + \mathbf{M}_{\text{coll}} . \qquad (2.11)$$

For non-spherical particles one also needs to track the change of the angles defining the orientation of the particles. However, since in this work all particles are treated as spherical and therefore particle orientation does not play a role, only the equation for the angular velocity is given here. The only exception in the discrete-element model for fragmentation in Ch. 6, where non-central contact forces between particles are discussed. In this case one needs to keep track of particle orientation, or more precisely of the movement of the initial contact point between particles. We refer to the description in Ch. 6 how this can be done.

The collision torques can again be obtained from appropriate elastic or inelastic collision models, e.g. Luding (2008) while the fluid torque is obtained as the integral of the torque generated by the fluid forces over the surface of the particle

$$\boldsymbol{M}_{\text{surf}} = \oint_{S} (\boldsymbol{x} - \boldsymbol{X}(t)) \times (\sigma \boldsymbol{n}) dS . \qquad (2.12)$$

Together with the boundary conditions, both at the system boundaries and at the particle surface, equations (2.4), (2.5), (2.8) and (2.11) are a full set of equations for a suspension of dispersed particles in a continuous fluid.

2.1.2. Approximations to Describe Particle-Laden Flows

Since particle-laden flows appear in many different contexts and applications a number of approaches have been developed to describe such systems, approximating the full system with varying degrees of detail and complexity. A first, rough distinction is between the case of a *dispersed system* and a *dense system*. In the case of a dispersed system the dynamics of the particles is mainly determined by the fluiddynamical forces acting on the particles. For a dense system, particle concentrations become large enough so that the particle motion is dominated by collisions and contact between particles. If the particle concentration is large enough, the surrounding continuous phase can be neglected and a description as a granular flow becomes appropriate (Pöschel and Schwager, 2005). Here, we focus solely on dispersed systems. In this case the difference between the approaches lies mainly in the treatment of the dispersed phase. As long as the Knudsen number is large enough the fluid phase is commonly described in a standard fluid dynamics formulation in an Eulerian frame, i.e. as a velocity field $\boldsymbol{v}(\boldsymbol{x}, t)$.

There are a number of different categories to characterize the approaches to dispersed particle-laden flows. The most important is whether the particle velocity can differ noticeably from the velocity of the surrounding fluid. This is the difference between *tracers*, particles that follow exactly the dynamics of the fluid and *inertial particles* whose velocity can be significantly different from that of the surrounding fluid. Sometimes this difference is also referred to as *mixed-fluid* versus *separated-fluid* (Loth, 2010), indicating that in the first case particles and surrounding fluid are treated as one fluid, with a single partial differential equation describing the dynamics whereas in the second case each phase has different transport equations. There also exists a hybrid method, the so-called *weak-inertia* or *weakly-separated-fluid* approach, where velocity differences between the phases are assumed to be small.

The second major category is whether the dispersed phase is treated in a Lagrangian or an Eulerian frame of reference. In the Eulerian frame of reference one deals with a field equation, i.e. a velocity field for the dispersed phase, similar to the continuous phase. In the Lagrangian frame of reference one tracks the velocity V(t) along individual particle trajectories X(t), i.e. following a discrete particle.

For each of these different categories in the dispersed flow regime, one can distinguish a number of different coupling cases, that depend mainly on the concentration of the dispersed phase:

- One-way particle-flow coupling, where the particle motion is coupled to that of the fluid, but the fluid is not affected by the presence of the particles.
- Two-way particle-flow coupling, where the presence of the particles additionally affect the dynamics of the surrounding fluid.
- Particle-particle interactions, where the motion of different particles in the flow is coupled, either through interactions through the fluid or through direct particle-particle contact.

For one-way flow coupling, i.e. when the feedback of the particles on the surrounding flow is neglected the equations of motions for the particles are expressed in terms of the undisturbed flow field $\boldsymbol{u}(\boldsymbol{x},t)$ at the position of the particles. The real flow field $\boldsymbol{v}(\boldsymbol{x},t)$ is not explicitly calculated, but may appear in the derivation of the forces acting on the particle, for example in the derivation of the Maxey-Riley equations in Sec. 2.3.2.

For small particle concentrations, one-way particle-flow coupling is usually a good approximation whereas for larger particle concentrations the feedback of the particles on the fluid becomes important. Particle-particle interactions can appear both for one-way and two-way particle-flow coupling, though generally they also become more important for increasing particle concentration.

Different approximations are described in more detail in the following Sections. We start with the description of a mixed-fluid multiphase flow approximation, both in an Eulerian and a Lagrangian framework with one- and two-way particle-flow coupling in Sec. 2.2. In addition we describe some basic results for the dynamics of such systems, in particular for the Lagrangian mixed-fluid approach. Then we discuss basic models for separated and weakly-separated flows in Sec. 2.2.3, where the particle velocity is allowed to differ noticeably from the velocity of the surrounding fluid. Since such inertial particles are the focus of the remainder of this work, we discuss in detail some theoretical results for the dynamics and interactions of suspensions of inertial particles.

2.2. Mixed-Fluid Multiphase Flows: Tracer Particles

The term mixed-fluid multiphase flows refers to the description of the dynamics of tracers, where the particle motion follows completely the dynamics of the surrounding flow. In Sec. 2.2.1 an Eulerian approach to one-way coupled mixed-fluid multiphase flows is discussed briefly. The second part consists of a Lagrangian approach to one-way coupled mixed-fluid multiphase flows in Sec. 2.2.2. The model approach and some classical results are discussed. This is concluded by a short discussion in Sec. 2.2.3 of how these mixed-fluid multiphase flow approaches can be extended to two-way particle-flow coupling.

2.2.1. One-Way Coupled Eulerian Mixed-Fluid

In an Eulerian description of mixed-fluids it is assumed that the dispersed phase can also be described as a continuum, i.e. instead of individual particles one deals with a scalar concentration field $C(\boldsymbol{x},t)$. This is the so-called particle-phase continuum assumption (Drew and Passman, 1999). Since in reality particles are discrete, a number of criteria need to be met for this continuum assumption to be reasonable.

Defining a Concentration Field

Typically, the concentration field $C(\boldsymbol{x},t)$ is either a number density, a volume fraction or a mass density. A number density is the number of particles per volume, a volume fraction is the volume taken up by the particle per unit volume, whereas a mass density is the mass of particles per volume. All three are usually obtained by cell-averaging, i.e. by calculating the number, volume or mass of particles in a discrete cell of a certain volume V_c . If $N_p^{(j)}$ is the number of particles whose center lies within a certain computational cell j, $V_p^{(i)}$ is the volume of the *i*-th particle and $m_p^{(i)}$ is the mass of the *i*-th particles, then the cell-averaged densities are given by

$$C_N(\boldsymbol{x}^{(j)}, t) = \frac{N_p^{(j)}}{V_c} \qquad \text{(number density)} \qquad (2.13)$$

$$C_V(\boldsymbol{x}^{(j)}, t) = \frac{1}{V_c} \sum_{i=1}^{N_p^{(j)}} V_p^{(i)} = C_N \langle V_p \rangle \qquad \text{(volume fraction)} \qquad (2.14)$$

$$C_M(\boldsymbol{x}^{(j)}, t) = \frac{1}{V_c} \sum_{i=1}^{N_p^{(j)}} m_p^{(i)} = \rho_p C_V \qquad \text{(mass density)}. \tag{2.15}$$
(2.16)

Here $\boldsymbol{x}^{(j)}$ is the position of the center of the *j*-th computational cell and $\langle V_p \rangle$ is the average volume of a particle in the computational cell, i.e. $\langle V_p \rangle := \frac{1}{N_p^{(j)}} \sum_{i=1}^{N_p^{(j)}} V_p^{(i)}$.

For the particle-phase continuum assumption to be justified, the cell volume V_c and the number of particles per cell N_p need to meet certain criteria. First, the cell volume needs to be large enough so that at least one particle fits completely within one cell, i.e. $V_p < V_c$. Otherwise the problem is not well-posed. Second, the number of particles within a cell volume needs to be large enough, i.e. $N_p \gg 1$, so that the particle density will vary smoothly between cells and not depend strongly on the exact position of the cell. This means that the mean interparticle distance \bar{L}_{pp} is small compared to the length scale L_c of a cell. This is similar to the criterion of the Knudsen number for the continuity assumption in the surrounding fluid in Sec. 2.1.1. The simplest way to satisfy this is to take the cell volume as large as possible. However, typically one also wants to resolve spatial gradients in the concentration field which requires that the length scale of a cell is smaller than the length scale of the changes in the concentration field, i.e. $L_c \ll \frac{C}{\nabla C}$. Together, this means that in order to represent the dispersed phase as a continuum, one needs to be able to choose a cell that is neither too small nor too large, in particular the criterion $\bar{L}_{pp} \ll L_c \ll \frac{C}{\nabla C}$ needs to be satisfied.

Transport Equations

The equation of motion for the concentration field $C(\boldsymbol{x},t)$ can be derived from a conservation principle. The assumption of a one-way coupled mixed-fluid means that the dispersed phase flow follows exactly the flow of the continuum phase. The flow of the continuum phase is in this case an *undisturbed flow*, i.e. the fluid flow without the presence of the particle phase. The equations of motion for the undisturbed flow \boldsymbol{u} are the Navier-Stokes equations

$$\rho_f(\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u} \cdot \nabla)\boldsymbol{u}) = \rho_f \boldsymbol{g} - \nabla p + \nu_f \nabla^2 \boldsymbol{u} , \qquad (2.17)$$

where g is the gravity vector and the continuity equation for an incompressible flow with constant density

$$\nabla \cdot \boldsymbol{u} = 0 , \qquad (2.18)$$

together with boundary conditions at the system boundaries. We emphasize at this point that the only difference between the undisturbed flow field u and the full flow field v is that v is constrained by additional boundary conditions at the particle surface.

If the dispersed phase follows exactly the flow of the surrounding fluid this means that the advective flux j_A of the dispersed phase is given by $j_A = u(x, t) \cdot C(x, t)$. The equation of motion for C follows then directly from the continuity equation for the dispersed phase

$$\frac{\partial C}{\partial t}(\boldsymbol{x},t) = -\nabla \cdot \boldsymbol{j}_C(\boldsymbol{x},t) + S_C(\boldsymbol{x},t) , \qquad (2.19)$$

which states that the local change in concentration is given by the gradient of the total flux $\mathbf{j}_C(\mathbf{x},t)$ and a possible volumetric source $S_C(\mathbf{x},t)$. Typically, for the total flux \mathbf{j}_C one takes the sum of the advective flux \mathbf{j}_A and a diffusive flux $\mathbf{j}_D = -\Gamma_{MD}(\mathbf{x},t) \cdot \nabla C(\mathbf{x},t)$ (Fick's law), with a molecular diffusion coefficient $\Gamma_{MD}(\mathbf{x},t)$ that can in principle depend both on space and time. However, in many cases the molecular diffusion coefficient is assumed to be a constant. A space and time depended diffusion coefficient appears often when describing systems where the flow of the surrounding fluid is turbulent. The simplest approach to include the effect of turbulent fluctuations in the velocity field leads to an additional space- and time dependent diffusion term in the equations of motion (Pope, 2008). The molecular diffusion coefficient is then replaced by an effective diffusion coefficient $\Gamma_{\text{eff}}(\mathbf{x},t) = \Gamma_{MD} + \Gamma_{TD}(\mathbf{x},t)$.

However, for simple molecular diffusion with constant diffusion coefficient the equation for the concentration field C reduces to

$$\frac{\partial C}{\partial t}(\boldsymbol{x},t) = -\nabla \left(\boldsymbol{u}(\boldsymbol{x},t) \cdot C(\boldsymbol{x},t)\right) + \nabla \left(\Gamma_{MD}(\boldsymbol{x},t) \cdot \nabla C(\boldsymbol{x},t)\right) + S_C(\boldsymbol{x},t) \ . \tag{2.20}$$

If the flow \boldsymbol{u} of the surrounding fluid is assumed to be incompressible, i.e. $\nabla \boldsymbol{u} = 0$ and the source term S_C for the dispersed phase is neglected, the equation of motion becomes

$$\frac{\partial C}{\partial t}(\boldsymbol{x},t) = -\boldsymbol{u}(\boldsymbol{x},t) \cdot \nabla C(\boldsymbol{x},t) + \nabla (\Gamma_{MD}(\boldsymbol{x},t) \cdot \nabla C(\boldsymbol{x},t)) \quad (2.21)$$

$$= -u(\boldsymbol{x},t) \cdot \nabla C(\boldsymbol{x},t) + \Gamma_{MD} \cdot \nabla^2 C(\boldsymbol{x},t).$$
(2.22)

This advection-diffusion equation is probably the most widely used transport equation for the dispersed phase in a multiphase flow. Together with the equations (2.17) and (2.18) for the undisturbed fluid flow \boldsymbol{u} this defines the full set of equations for a one-way coupled Eulerian mixed-fluid multiphase flow.

2.2.2. One-Way Coupled Lagrangian Mixed-Fluid

In the case of a Langrangian approach to describe the dynamics of the dispersed phase in the mixed-fluid regime, the position X(t) of individual particles is tracked over time. Instead of an advection-diffusion equation for the particle velocity field, the motion of each individual particle is described in terms of an ordinary differential equation, or if diffusion is included in terms of a stochastic differential equation. While a Lagrangian approach offers some advantages, particularly collisions and interactions between particles can be readily included the main drawback is that for larger systems the numerical requirements can become quite large. Since one needs to solve an equation for each individual particle, instead of a single equation for the whole particle field, generally only smaller systems can be studied. On the other hand, the Lagrangian view on fluid dynamics has revealed a number of phenomena that were not apparent in the Eulerian description of fluids, both in laminar flows where the Lagrangian particle dynamics can become chaotic and in turbulent flows.

In the Lagrangian approach for the one-way coupled mixed fluid it is assumed for each individual particle that the particle velocity is given by the undisturbed velocity of the surrounding fluid at the position of the particle, possibly plus an additional diffusive term. This means that the particles are treated as fluid elements, i.e. there is no difference between the motion of an infinitesimal portion of the continuous phase and of a particle.

In the case where the diffusivity Γ_C of the dispersed phase is independent of the particle position, the Lagrangian equation of motion can be written as

$$d\boldsymbol{X}(t) = \boldsymbol{u}(\boldsymbol{X}(t), t)dt + \sqrt{2\Gamma_C}d\boldsymbol{W} , \qquad (2.23)$$

where dW is a three dimensional Wiener increment. This is the Langevin equation corresponding to the advection diffusion equation (2.21). Together with the equations (2.17) and (2.18) for the undisturbed fluid flow u this defines the full set of equations for a one-way coupled Lagrangian mixed-fluid multiphase flow.

It is important to note that this straightforward random walk model for the particle diffusion is no longer correct when the diffusivity depends on space, for example when one wants to model the influence of turbulence similar to the gradient-diffusion approximation discussed in the previous section. It was pointed out by Visser (1997) that the correct Lagrangian equations of motion, corresponding to an advection-diffusion equation with space-dependent diffusivity $\Gamma_{\text{eff}}(\boldsymbol{x}, t)$ also contains a contribution from the gradient of the diffusivity. The correct equation of motion is then given by

$$d\mathbf{X}(t) = (\mathbf{u}(\mathbf{X}(t), t) + \nabla\Gamma_{\text{eff}}(\mathbf{X}(t), t)) dt + \sqrt{2\Gamma_{\text{eff}}(\mathbf{X}'(t), t)} d\mathbf{W} , \qquad (2.24)$$

where the diffusivity is evaluated at $\mathbf{X}'(t) = \mathbf{X}(t) + \frac{1}{2}\nabla\Gamma_{\text{eff}}(\mathbf{X}(t), t)dt$. If this correction is not included, there is an artificial diffusion of particles to regions of low diffusivity, even when the initial particle distribution is uniform.

Next, we discuss some basic features of the dynamics of a suspension of particles in laminar and turbulent flows in such an Lagrangian one-way coupled mixed-fluid approximation.

Lagrangian Chaos

One of the most interesting results about Lagrangian mixed-fluid dynamics, is that even if the velocity field of the continuous phase is very simple, the motion of individual fluid elements or particles can be very complicated. If a small region of the fluid is tagged, e.g. with dye or a small group of particles is released in a section of an appropriate flow one can observe a stretching and folding of material lines. These evolving features in the flow are typically a combination of so-called "tendrils" and "whorls" (Aref, 1984), generated by different underlying structures in the flow. This deformation of fluid elements or small particle groups is often called hydrodynamical "stirring". This hydrodynamical stirring plays an important role for mixing processes. Here, mixing refers to the diffusive exchange of material between two fluids. This greatly depends in the contact surface between the two fluids. In many natural problems and applications this is achieved by making the surrounding fluid flow turbulent. However, when due to some restriction the flow can not easily be made turbulent, e.g. when very large spatial scales are involved or one wants to mix very fragile particles, stirring by Lagrangian chaos can also be used to enhance mixing by creating a greatly increased surface for diffusion.

This effect was pointed out theoretically in the work of Aref (1984), first in the case of an inviscid flow and shortly afterwards in the Stokes flow regime by Aref and Balachandar (1986) and was termed chaotic advection or Lagrangian chaos. The first experimental realization was obtained by Chaiken et al. (1986). For details on Lagrangian chaos the book by Ottino (1989) is recommended, in the following a short summary of the underlying mechanism is provided.

Lagrangian Chaos can be understood in terms of the theory of Hamiltonian systems. The phase space is identical to the spatial position of the particles. To simplify the explanation, only two dimensional systems are considered. A two dimensional, incompressible flow can always be given in terms of a stream function (Durst, 2006), which is a Hamiltonian for the system. If the flow is steady, the system possesses one degree of freedom and is integrable. In this case all bounded solutions are regular, and can be mapped to an invariant torus (Arnold-Liouville theorem). If the Hamiltonian system is disturbed, for example by introducing a time-dependence to the system it may no longer be integrable. In this case some of the Arnold-Liouville tori are destroyed, whereas others remain stable for sufficiently small disturbances. This is the result of the celebrated Kolmogorov-Arnold-Moser-theorem (KAM) and the remaining stable tori are therefore termed KAM-tori.

In place of the destroyed tori a sequence of elliptic and hyperbolic fixed points is created (Poincaré-Birkhoff theorem). Elliptic points once again lead to KAM-tori that can either remain stable or be destroyed, leading to more elliptic and hyperbolic points and so on, continuing to infinity.

The hyperbolic fixed points created in the destruction of tori are the ones responsible for the Lagrangian chaos. In their neighborhood, the stable and unstable manifolds can form homoclinic or heteroclinic points, i.e. stable and unstable manifolds of one or different hyperbolic points cross orthogonally. Since one homoclinic or heteroclinic point implies infinitely many others this results in a complicated folding of the manifolds. The chaotic saddle formed by this "homoclinic/heteroclinic tangle" leads to the exponential separation of initially close fluid elements or particles.

The phenomenon of chaotic advection plays an important role in many problems where stirring and mixing in non-turbulent flows occurs in a fluid, ranging from microfluids and material processing to stirring of fluids on geophysical or planetary scales, for example in the ocean or the earth mantle (see e.g. Zimmerman, 1986; Behringer et al., 1991). Chaotic advection in a fluid has also been found to play a role in many problems where active processes of the particles play a role, for example for plankton blooms in vortices behind islands (Sandulescu et al., 2007), chemical reactions (Károlyi et al., 1999; Tél et al., 2004) or coexistence of biological competitors (Károlyi et al., 2000). For an overview of active processes in Lagrangian chaos see for example the review by Tél et al. (2005).

Lagrangian Particles in Turbulence

Another field, where the Lagrangian study of fluid flows plays an important role is the field of fluid turbulence. This view on fluid turbulence, originally suggested by L.F. Richardson and G.I. Taylor in the 1920s and greatly developed by R.H. Kraichnan has led to considerable process in recent years. In particular for the phenomena of intermittence and anomalous scaling laws in turbulent flows, the applications of methods from nonequilibrium statistical mechanics has led to a new quantitative understanding. Two of the major problems in this context are the question of the extend of the universality of the inertial range and the question of how to obtain velocity statistics in a non-equilibrium system. Some of the observed anomalous scaling behavior and the connected breaking of the scale-invariance symmetry has been traced to the existence of statistical integrals of motion. These can be very different from the conserved quantities in equilibrium statistics and are found in the evolution of groups of particles and the geometry of the particle configuration.

An overview of the more advanced problems related to the statistical description of turbulence, such as multi-particle statistics and anomalous scaling is beyond the scope of this work. As a starting point for this the article by Falkovich et al. (2001) is recommended. In the following, only the basic results for the motion of an individual Lagrangian particles in turbulence are summarized, following the description in Pope (1994) and Falkovich et al. (2001).

For a single Lagrangian particle, moving according to the stochastic differential equation (2.23) in a turbulent flow the behavior can be understood in terms of the mean square displacement $\langle (\Delta \mathbf{X}(t))^2 \rangle_E = \langle (\mathbf{X}(t) - \mathbf{X}(0))^2 \rangle_E$, where $\langle \cdot \rangle_E$ denotes an ensemble average. Assuming the flow is statistically stationary and neglecting molecular diffusion $(\Gamma_{MD} = 0)$, the equation for the mean square displacement is

$$\frac{d}{dt} \left\langle (\Delta \mathbf{X}(t))^2 \right\rangle_E = \left\langle \left(\frac{d}{dt} \mathbf{X}(t) \right) \cdot 2 \left(\mathbf{X}(t) - \mathbf{X}(0) \right) \right\rangle_E$$
$$= 2 \int_0^t ds \left\langle \mathbf{V}(0) \cdot \mathbf{V}(s) \right\rangle_E . \tag{2.25}$$

This quantity depends strongly on the length of temporal correlations of the Lagrangian velocity. Defining the Lagrangian correlation time of the flow as

$$\tau_L := \frac{\int_0^\infty ds \, \langle \mathbf{V}(0) \cdot \mathbf{V}(s) \rangle_E}{\langle \mathbf{V}^2 \rangle_E} \,, \tag{2.26}$$

two different limit cases for the mean square displacement can be distinguished.

For times much shorter than the correlation time, i.e. $t \ll \tau_L$ it is $\langle \mathbf{V}(0) \cdot \mathbf{V}(t) \rangle_E \approx \langle \mathbf{V}^2 \rangle_E$ and therefore

$$\left\langle (\Delta \boldsymbol{X}(t))^2 \right\rangle_E \approx 2 \left\langle \boldsymbol{V}^2 \right\rangle_E t^2 = 2 \left\langle \boldsymbol{u}^2 \right\rangle_E t^2 .$$
 (2.27)

This means that on short time scales the particle motion is ballistic.

Assuming a finite correlation time τ_L of the flow, which is typically the case in turbulent flows one finds for large times, i.e. $t \gg \tau_L$ that $\int_0^t ds \langle \mathbf{V}(0) \cdot \mathbf{V}(s) \rangle_E \approx \tau_L \langle \mathbf{V}^2 \rangle_E$. This leads to

$$\left\langle (\Delta \boldsymbol{X}(t))^2 \right\rangle_E \approx 2 \left\langle \boldsymbol{V}^2 \right\rangle_E \tau_L t = 2 \left\langle \boldsymbol{u}^2 \right\rangle_E \tau_L t \;.$$
 (2.28)

Therefore, on time scales much longer than the correlation time of the flow the particle

motion is diffusive.

In general, for time scales much longer than τ_L the individual displacements ΔX are approximately independent, and the overall displacement becomes a sum of many independent, random variables, which is the regime of the central limit theorem. The displacement therefore is a Brownian motion in 3 dimensions,

$$\left\langle \Delta X_i(t) \Delta X_j(t) \right\rangle_E \approx 2 D_{ij}^{(\text{eff})} t$$
 (2.29)

with an effective diffusivity tensor $D_{ij}^{\rm (eff)}$ given by

$$D_{ij}^{(\text{eff})} = \frac{1}{2} \int_0^\infty ds \left\langle V_i(0) V_j(s) + V_i(s) V_j(0) \right\rangle_E .$$
 (2.30)

The challenge of turbulent diffusion is then to find the effective diffusivity, for a given velocity field $\boldsymbol{u}(\boldsymbol{X},t)$.

2.2.3. Two-Way Particle-Flow Coupling for Mixed-Fluids

While the results in the previous section mainly refer to the case of one-way particleflow coupling, where the feedback of the dispersed phase on the continuous phase can be neglected, the approach can easily be extended to account for feedback of the particles on the fluid. In the case of mixed-fluids, where the dispersed phase is assumed to follow closely the continuous phase the two-way particle-flow coupling is generally achieved by replacing the fluid properties of the continuous phase with effective properties (Clift et al., 2005).

The two major effects of two-way particle flow coupling are an influence of the particles on the surrounding fluid by locally modifying the density and viscosity. The local change in density can greatly influence the mean flow field. In addition, the presence of particles can lead to an effective viscosity in regions of the flow with high particle concentrations that is significantly different from the viscosity of the undisturbed fluid. In addition to the mean flow this change in effective viscosity can influence turbulent fluctuations by changing production and dissipation of turbulent kinetic energy. Both of these effects become important, if there are regions of high particle concentrations in the flow.

The effective density $\rho_m(\boldsymbol{x}^{(j)}, t)$ at the position $\boldsymbol{x}^{(j)}$ of a computational cell can most easily be given in terms of the volume fraction $C_V(\boldsymbol{x}^{(j)}, t)$ and is simply the mass of both the dispersed and continuous phase per unit volume

$$\rho_m := \rho_p C_V + \rho_f (1 - C_V) . \tag{2.31}$$

For particles much heavier than the surrounding fluid and small volume fractions, the effective density increases approximately linearly with the mass concentration

$$\rho_m \approx \rho_f + C_M, \quad \text{for } \rho_p \gg \rho_f \text{ and } C_V \ll 1.$$
(2.32)

The effective viscosity $\mu_m(\mathbf{x}^{(j)}, t)$ at the position $\mathbf{x}^{(j)}$ of a computational cell can be understood physically as the ratio of stress to rate-of-strain in the particle-fluid mixture. The effective viscosity for a suspension of solid particles was first calculated by Einstein (1906) in the creeping flow limit and negligible particle-particle interaction. His result was obtained by considering an individual particle in a straining field, with a no-slip condition at the particle surface and assuming that the effect can be summed linearly for more particles. To first order in C_v he obtained

$$\mu_m = (1 + 2.5C_V)\mu_f , \qquad (2.33)$$

which has been confirmed in experiments to be reasonable for spheres up to volume fractions of about 0.1. Non-spherical particles may have a greater increase in effective viscosity.

A similar analysis for viscous spheres, e.g. liquid drops was performed by Batchelor (1970) where he assumed a stress balance boundary condition at the particle surface. To first order in C_V he found that

$$\mu_m = \left(1 + \frac{2.5\mu_p + \mu_f}{\mu_p + \mu_f} C_V\right) \mu_f .$$
(2.34)

In the limit of infinite particle viscosity this again reduces to the result for solid particles, Eq. (2.33) whereas in the limit of negligible particle viscosity one obtains

$$\mu_m = (1 + C_V)\mu_f , \qquad (2.35)$$

i.e. the effective viscosity increases linearly with the volume fraction. This is for example the case of gas bubbles suspended in a liquid.

The full set of equations for the two-way coupled mixed-fluid approximation is therefore given by the mixed-fluid equation for the dispersed phase, either the advection-diffusion equation(2.21) in the Eulerian case or the Langevin equation (2.23) in the Lagrangian case, combined with the Navier-Stokes and continuity equations (2.17) and (2.18) for the undisturbed flow field \boldsymbol{u} from Sec. 2.2.1 where ρ_f and μ_f have been replaced by ρ_m and μ_m , respectively to approximate the real flow field \boldsymbol{v} .

In addition to the modification of density and viscosity, there can be further modifica-

tions of the continuous phase properties due to the presence of a dispersed phase, such as changes in the compressibility affecting for example the speed of sound in the suspension, but the discussion of these is beyond the scope of this work.

2.3. Separated-Fluid Multiphase Flows: Inertial Particles

In this section the equations of motion for a separated-fluid multiphase flow, i.e. a suspension of inertial particles are described. Particles are called inertial if the particle velocity can deviate from that of the surrounding fluid. This is usually a much better approximation of the dynamics of suspended particles in a flow than the simple mixed-fluid approach described in the previous section, but comes at a greater (computational) cost. Once again, one can try to formulate the equations of motion either in an Eulerian or a Lagrangian frame of reference.

In order to formulate a transport equation in an Eulerian frame of reference for a particle concentration $C(\mathbf{x}, t)$, as was done with the advection-diffusion equation (2.21) for tracers, a particle velocity field $\mathbf{v}_p(\mathbf{x}, t)$ is required. For tracers this was simply the velocity field of the surrounding fluid. Assuming the existence of such a velocity field, the equations of motion in an Eulerian frame of reference can be obtained and include both a continuity and momentum equation. The momentum equation for the dispersed phase can be derived similar to the Navier-Stokes equation (see e.g. Durst, 2006) and contains the surface forces acting on the particles, e.g. drag forces. The same forces then appear as a sink term in the governing equations for the continuous phase, leading to a simple formulation for two-way particle-flow coupling. In general, such an Eulerian approach has the advantage of allowing consistent numerics for both the dispersed and continuous phase and is generally much faster than a Lagrangian approach for large numbers of particles.

However, in the case of inertial particles there are fundamental problems associated with an Eulerian frame of reference. Due to the presence of so-called caustics (Falkovich et al., 2001; Wilkinson and Mehlig, 2005) in the dynamics of inertial particles the existence of a velocity field $v_p(x, t)$ is not guaranteed for all points and times. Instead, particles with very different velocities can come arbitrarily close, preventing the definition of a unique particle velocity at a given point and time. Such "multivalued velocity fields"¹ are the reason why Eulerian formulations can not fully capture particle inertia. Nonetheless, due to their superior efficiency for large particle concentrations they are widely in use.

In the following only a Lagrangian description of inertial particle is used, because

¹This formulation, although sometimes used in the literature, is somewhat misleading, since the definition of a field implies unique values at any given point.

inertial effects that do not appear in an Eulerian description such as caustics can greatly influence collision rates of particle. This is an important aspect for the coagulation/aggregation processes studied in Ch. 3 and 4 and should be included there. This problem does not appear in the weak-inertia approximation (see Sec. 2.3.2), where both Eulerian and Lagrangian formulations can be used.

2.3.1. Approaches to Calculate the Surface Force and Torque on a Particle

The main question in deriving an approximation for the full set of equations from Sec. 2.1.1, describing the motion of a system of particles in a continuous fluid is how to approximate the surface force and torque on the particles. To calculate the surface force F_{surf} and torque M_{surf} on a particle a number of different approaches are used, that reproduce equations (2.10) and (2.12) with varying degree of accuracy. Some results are based on theoretical arguments, while others are empirical formulations, either based on experiments or resolved surface simulations.

First, we discuss one-way particle-flow coupling approximations, where the feedback of the particles on the surrounding flow is neglected. For this, we use the so called pointforce approximation, where an expression for the surface force and torque is derived in terms of the undisturbed flow field \boldsymbol{u} at the position $\boldsymbol{X}(t)$ of a particle. This eliminates the need to calculate the detailed flow around the particle, but is only appropriate if the particle diameter d_p is smaller than the smallest characteristic length scale in the flow, i.e. $d_p \ll \tilde{L}_f$ so that local flow variations are small. If the surrounding flow is spatially discretized, the flow properties are mapped from the nearest flow node to the particle position with an appropriate interpolation scheme.

More detailed approximation methods than the point-force approximation, which usually also include two-way particle-flow coupling are for example distributed-force and -torque approximations and semi-resolved and resolved surface methods.

Distributed-force and -torque approximations are usually applied where particles are of similar length scale than the resolution of the flow. In this case the variations of the flow over the diameter of the particle needs to be included. This can be done by spatially averaging the surrounding flow properties within a region surrounding the particle center. For recent results consistent with known theoretical formulations see Loth and Dorgan (2009).

For particles larger than the resolution of the flow semi-resolved and resolved surface methods can be used to calculate the forces and torques acting on the particle. One of the most common semi-resolved surface methods is the force-coupling method by Maxey and Patel (2001) and Lomholt and Maxey (2003). In the force-coupling method a partial differential equation for the surrounding fluid is solved that contains an interface force for each individual particle, resulting in an surrounding flow field that locally represents the particle surface velocity V close to the particle.

Resolved surface methods, such as arbitrary Lagrangian-Eulerian (ALE) methods (e.g. Maury and Glowinski, 1997) and Lagrangian Multiplier (LM) methods (e.g. Glowinski et al., 1999) can be used when the resolved length scale of the surrounding flow is much smaller than the particle diameter. These methods can give very detailed results for surface forces and torques acting on the particle, but are computationally very expensive.

In this work, only the point-force approximation is discussed, since distributed-force, semi-resolved and resolved surface methods usually require a greatly increased numerical effort which is not reasonable for the large ensembles of particles studied in the following parts. However, we show how the one-way particle-flow coupling point-force approach can be expanded to include two-way particle-flow coupling. For point-force approximations with one-way particle-flow coupling the equations for the fluid phase are always the Navier-Stokes and continuity equations for the undisturbed flow field u from Sec. 2.2.1. For two-way particle-flow coupling and particle-particle coupling, as discussed in Sec. 2.3.4 and 2.3.4, the equations for the continuous phase are modified to include effects from the presence of the dispersed phase, such as modified density and viscosity as well as a momentum transfer between fluid and dispersed phase.

In the point-force approximation used in this work the integrated surface force F_{surf} and torque M_{surf} are usually separated into a sum of several independent components. The forces generally include the undisturbed fluid stresses F_S acting on the particle, quasisteady drag F_D , the added-mass force F_A , the Basset-Boussinesq history force F_H , lift forces F_L and possibly Brownian forces F_B . Similar decompositions exist for the torques, but in most cases only the torque from the quasi-steady drag M_D is considered. The point-force approximation has a number of limitations. In addition to the requirement that the particle diameter need to be smaller than the smallest characteristic length scale of the flow, the assumption of a linear decomposition of the surface force and torque into separate and independent forces is not guaranteed except in a few theoretically understood cases. Furthermore, the expressions for the different components of the surface force and torque are not theoretically known except in very few limit cases. Therefore, in many applications, particularly in higher Reynolds number flows and for non-spherical particles, empirical results and results from resolved-surface methods are used.
2.3.2. Theoretical Results: Maxey-Riley and Auton-Hunt-Prud'homme

Over the last 180 years a number of analytical expressions for the different surface forces in the equations of motion of suspended particles have been found. The first work appeared even before the development of the Navier-Stokes equations, when Poisson (1831) studied a sinusoidal potential flow around a solid sphere and discovered what is now called the added-mass term. The interest of the early researcher was mainly the motion of a pendulum in air, which was related to the construction of clocks, often for the determination of geographical longitude (see e.g. the review by Michaelides (1997)).

Twenty years later, Stokes (1851) developed a theory for the unsteady sinusoidal motion of a solid sphere in a viscous fluid, in the so-called "creeping-flow" limit. This is the limit of zero particle Reynolds number $Re_p := \Delta V d_p / \nu_f$, with ΔV the characteristic relative velocity between particles and surrounding fluid flow, where the nonlinear term of the Navier-Stokes equations can be neglected. This limit case of the Navier-Stokes equation, without the inertia term is now commonly called the Stokes equation.

Based on the work of Stokes, Basset (1888), Boussinesq (1903) and later Oseen (1927) further developed the expressions for the unsteady motion of a solid sphere in a viscous fluid. Again, under creeping-flow conditions they developed and refined the expressions for steady-state drag, added mass and what is now called the Basset-Boussinesq history force. Further improvement came from Oseens student Faxén (1922) who included contributions to the surface forces due to a nonuniform fluid velocity, that today are called Faxén corrections.

Despite the number of works on this topic, full theoretical solutions for the equations of motion have so far only been found in the case of either the creeping flow conditions, where $Re_p \rightarrow 0$ or inviscid flows, where $\mu_f = 0$. The two most important and widely used theoretical results are the Maxey-Riley equations (1983) and the Auton-Hunt-Prud'homme equations (1988). The Maxey-Riley equations describe the non-rotating motion of a solid sphere in unsteady, incompressible flows at small, but finite relative particle velocity. The Auton-Hunt-Prud'homme equations describe the relative motion of a sphere in an inviscid, rotational fluid. In both cases Brownian motion is neglected. In many applications and theoretical studies one of these equations is used, either in their original form or as a baseline equation to which additional effects or corrections to the different terms are added.

Maxey-Riley Equations

Equations for particle motion based on the creeping flow assumption are still among the most widely used representations of actual particle motion in a fluid. The most important recent contribution is the work of Maxey and Riley (1983) who published a very detailed derivation of the relevant equations of motion. In particular, they clarified some theoretical parts that were not fully apparent in the literature. They studied the motion of a rigid spherical particle in an non-uniform unsteady flow, without rotational motion. They assumed finite, but small particle velocity relative to the surrounding fluid, therefore neglecting nonlinear advection terms. Additionally, the spatial gradients in the flow were assumed to be small, so that these could be considered as small perturbations.

Formally, these restrictions can be written as

$$Re_p \ll 1$$
 (2.36)

$$(d_p^2/\nu_f)(\nabla \check{U}) \ll 1 \tag{2.37}$$

$$d_p/\check{L}_f \ll 1 , \qquad (2.38)$$

where ν_f is the kinematic viscosity of the flow, \dot{L}_f is a characteristic length scale for the variations in the flow and $\nabla \check{U}$ is a characteristic scale for the corresponding velocity gradients. With the restrictions (2.36) - (2.38) the Maxey-Riley equations can be derived from the equations of motion for the fluid (see Eq. (2.4)), assuming a no-slip condition at the particle surface (see Eq. (2.6)), non-rotating particles and assuming that the feedback from the particles on the surrounding fluid is only local. Their result includes gravitational and surface forces and reads as

$$m_{p}\frac{d\mathbf{V}}{dt} = (m_{p} - m_{f})\mathbf{g} + m_{f}\frac{D\mathbf{u}}{Dt}|_{\mathbf{X}(t)} -\frac{1}{2}m_{f}\frac{d}{dt}(\mathbf{V} - \mathbf{u}(\mathbf{X}(t), t) - \frac{1}{40}d_{p}^{2}\nabla^{2}\mathbf{u}(\mathbf{X}(t), t)) -3\pi d_{p}\nu_{f}(\mathbf{V}(t) - \mathbf{u}(\mathbf{X}(t), t) - \frac{1}{24}d_{p}^{2}\nabla^{2}\mathbf{u}(\mathbf{X}(t), t)) -\frac{3}{2}\pi d_{p}^{2}\mu_{f}\int_{0}^{t}ds\frac{1}{\sqrt{\pi\nu_{f}(t-s)}}\frac{d}{ds}[\mathbf{V}(s) - \mathbf{u}(\mathbf{X}(s), s) -\frac{1}{24}d_{p}^{2}\nabla^{2}\mathbf{u}(\mathbf{X}(s), s)].$$
(2.39)

Here

$$\frac{D\boldsymbol{u}}{Dt} = \frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u} \cdot \nabla)\boldsymbol{u}$$
(2.40)

is the substantial (Lagrangian) derivative along the trajectory of a fluid element, while

$$\frac{d\boldsymbol{u}}{dt} = \frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{V} \cdot \nabla)\boldsymbol{u}$$
(2.41)

is the derivative along the trajectory of the particle.

The first line in the Maxey-Riley equations of motion is the stress force F_S from the undisturbed fluid at the position of the particle, which together with the gravitational force also contains buoyancy forces. The second line is the added mass force F_A , the third line is the quasi-steady drag force F_D and the final lines are the Basset-Boussinesq history force F_H . The terms with $d_p^2 \nabla^2 u$ are the Faxen corrections for a nonuniform flow.

These equations were derived for particles initially starting at rest relative to the fluid, or more precisely with no initial drag, i.e. the Faxén-corrected relative velocity $\Delta \tilde{\boldsymbol{V}}(t) =$ $\boldsymbol{V}(t) - \boldsymbol{u}(\boldsymbol{X}(t), t) - \frac{1}{24}d_p^2 \nabla^2 \boldsymbol{u}(\boldsymbol{X}(t), t)$ is assumed to be zero at t = 0. A modification for non-zero initial relative velocity $\Delta \tilde{\boldsymbol{V}}(t)(0)$ was introduced by Maxey (1993), leading to an additional term $+\Delta \tilde{\boldsymbol{V}}(t)(0)/\sqrt{t}$ of the history force.

For many problems it is advantageous to rewrite the Maxey-Riley equations of motion in a slightly more compact form as

$$\frac{d\boldsymbol{V}}{dt} = \boldsymbol{G} + \frac{2}{3}\check{\boldsymbol{\beta}} \left(\frac{D\boldsymbol{u}}{Dt} + \frac{1}{2}\frac{d\boldsymbol{u}}{dt} + \frac{1}{40}d_p^2 \nabla^2 \boldsymbol{u} \right)
- \frac{1}{\tau_p} \left(\boldsymbol{V} - \boldsymbol{u} - \frac{1}{24}d_p^2 \nabla^2 \boldsymbol{u} \right)
- \sqrt{\frac{3\check{\boldsymbol{\beta}}}{\pi\tau_p}} \int_0^t ds \frac{1}{\sqrt{t-s}} \frac{d}{ds} [\boldsymbol{V} - \boldsymbol{u} - \frac{1}{24}d_p^2 \nabla^2 \boldsymbol{u}] ,$$
(2.42)

with the parameters

$$\boldsymbol{G} := \frac{m_p - m_f}{m_p + \frac{1}{2}m_f} \boldsymbol{g}$$
(2.43)

$$\tau_p := \frac{m_p + \frac{1}{2}m_f}{3\pi d_p \mu_f}$$
(2.44)

$$\check{\beta} := \frac{3}{2} \frac{m_f}{m_p + \frac{1}{2}m_f} . \tag{2.45}$$

The vector \boldsymbol{G} represents the influence of gravity, including buoyancy effects. τ_p is the particle response time, it represents the time scale on which the particle response to changes in the surrounding flow. Particles with small values of τ_p generally respond quickly to changes in the flow, whereas particles with large τ_p move almost independent from the flow. The product $\boldsymbol{W} := \tau_p \boldsymbol{G}$ is the terminal settling velocity of a particle in still fluid. The parameter $\check{\beta}$, already introduced in Sec. 2.1.1, is the mass or density ratio between the dispersed phase and the continuous phase. The limit $\check{\beta} \to 0$ is the case of particles much heavier than the surrounding fluid and the case of $\check{\beta} \to 3$ is the limit of particles much lighter than the surrounding fluid. Neutrally buoyant particles have $\check{\beta} = 1$.

Auton-Hunt-Prud'homme Equations

The Auton-Hunt-Prud'homme equations (1988) are a general expression for the force acting on a spherical particle moving relative to an inviscid, unsteady non-uniform rotational velocity field. In this derivation it is assumed that the local change in velocity over the radius of the particle is small compared to the relative velocity between the particle and the surrounding fluid. Additionally, it is assumed that the temporal changes of the relative velocity are small. These conditions can be written as

$$d_p \nabla \check{U} \ll \Delta \check{V} \tag{2.46}$$

$$\frac{\partial \Delta V}{\partial t} \ll \frac{\Delta V}{d_p} , \qquad (2.47)$$

where $\Delta \check{V}$ is a characteristic time scale for the relative velocity between particle and fluid and $\frac{\partial \Delta \check{V}}{\partial t}$ is a characteristic scale for the temporal changes of the relative velocity.

With the restrictions (2.46) - (2.47) the Auton-Hunt-Prud'homme equations can be derived from the equations of motion for the fluid (2.4), with a slip condition at the particle-fluid interface. Including gravitational and surface forces the equations of motion are given by

$$m_{p}\frac{d\boldsymbol{V}}{dt} = (m_{p} - m_{f})\boldsymbol{g} + m_{f}\frac{D\boldsymbol{u}}{Dt}|_{\boldsymbol{X}(t)}$$
$$-\frac{1}{2}m_{f}(\frac{d\boldsymbol{V}}{dt}(t) - \frac{D\boldsymbol{u}}{Dt}(\boldsymbol{X}(t), t))$$
$$+\frac{1}{2}m_{f}[\omega_{f} \times (\boldsymbol{u}(\boldsymbol{x}, t) - \boldsymbol{V}(t))], \qquad (2.48)$$

where $\omega_f = \nabla \times \boldsymbol{u}$ is the vorticity of the flow.

The terms in the Auton-Hunt-Prud'homme equations of motion are the stress F_S from the undisturbed fluid acting on the particle at position X(t), the added-mass force F_A and a lift force F_L which is a combination of rotational forces and a contribution due to an inertial force, related to the added mass effect. The form above is specific for a spherical particle. The assumption of an inviscid flow leads to vanishing total drag force on the particle, i.e. both quasi-steady drag F_D and history force F_H are zero.

In the following, the different components of the surface force in the Maxey-Riley equations and the Auton-Hunt-Prud'homme equations are compared and possible extensions discussed.

Fluid Stress Force

The fluid stress force represents the force required to accelerate the fluid at the particle position, without the presence of a particle. It is a force from the undisturbed fluid and comparing the results from Auton-Hunt-Prud'homme with the Maxey-Riley equations reveals that the force from the undisturbed flow is the same in both equations

$$oldsymbol{F}_S = m_f rac{Doldsymbol{u}}{Dt} |_{oldsymbol{X}(t)} - m_f oldsymbol{g}$$
 ,

In fact, for any incompressible flow, as long as the point force approximation is valid, the fluid stress force is given by the undisturbed continuous phase acceleration and the hydrostatic force. It requires no assumption whether the surrounding fluid is viscous and is generally the same for particles of any shape, whether solid or viscous. The importance of the fluid stress force was highlighted in the book of Batchelor (1967).

Drag Force

The drag force is generally considered to be the most important surface force acting on a particle. The derivation for the Stokes drag on a spherical, solid particle can be found in virtually every textbook on fluid dynamics (e.g. Durst, 2006). The linear quasi-steady drag force is typically proportional to the velocity difference between a particle and the surrounding fluid. The quasi-steady drag force in the Maxey-Riley equations is the Stokes drag, but corrected for non-uniform flows by being proportional to a Faxén-corrected relative velocity

$$\boldsymbol{F}_{D} = -3\pi d_{p}\nu_{f}(\boldsymbol{V}(t) - \boldsymbol{u}(\boldsymbol{X}(t), t) - \frac{1}{24}d_{p}^{2}\nabla^{2}\boldsymbol{u}(\boldsymbol{X}(t), t)) . \qquad (2.49)$$

Due to the assumption of an inviscid surrounding fluid, there is no drag force in the Auton-Hunt-Prud'homme equation.

There exists a number of empirical and theoretical corrections to the Stokes drag for a wide variety of particle and flow situations. Typically, this is written in terms of a corrective factor f_D to the Stokes drag

$$f_D := \frac{F_D}{F_D^{(\text{Stokes})}} . \tag{2.50}$$

Corrections to the Stokes drag include for example terms for finite particle Reynolds number such as the Oseen corrections (Oseen, 1910), corrections for non-spherical particles, viscous particles or particle deformations. Here only the correction to the Stokes drag for viscous particles is shown, since in Ch. 3 of this thesis we deal with the coagulation and breakup of liquid droplets. At the surface of a viscous sphere there can be a finite tangential velocity which reduces the viscous drag. In the creeping flow limit the correction for the Stokes drag is given by the Hadamard-Rybczynski expression (see e.g. the book by Clift et al., 2005)

$$f_D = \frac{2\mu_f + 3\mu_p}{3\mu_f + 3\mu_p} \ . \tag{2.51}$$

Some interesting limit cases include bubbles in a liquid, i.e. $\mu_p \ll \mu_f$ where $f_D \approx 2/3$, drops in a liquid, i.e. $\mu_f \approx \mu_p$ where $f_D \approx 5/6$ and drops in a gas. i.e. $\mu_p \gg \mu_f$, where $f_D \approx 1$. It should be noted that this result is only valid for clean fluids and particles, that means where no contaminants at the particle surface are present. In general, this is nearly impossible to obtain for small viscous particles in any experimental setup. With the presence of surface contaminants, the correction is typically $f_D \approx 1$ and the drag force is once again given by the Stokes drag.

Added Mass Force

The added mass force represents the additional force required because an inertial particle brings a certain amount of fluid into motion too. It represents a portion of the fluid that is accelerated along with the particle. It is formally included in the surface forces, because it is associated with the fluid stress acting on the particle. An added mass effect is generally found for all conditions where the fluid is disturbed by the presence of a particle and requires some rigidity of the particle surface. It generally occurs for particles of all shapes and sizes. The expression for the added mass term given by Maxey and Riley is

$$\boldsymbol{F}_{A} = -\frac{1}{2}m_{F}\frac{d}{dt}(\boldsymbol{V} - \boldsymbol{u}(\boldsymbol{X}(t), t) - \frac{1}{40}d_{p}^{2}\nabla^{2}\boldsymbol{u}(\boldsymbol{X}(t), t))$$
(2.52)

while the added mass term of Auton-Hunt-Prud'homme is

$$\boldsymbol{F}_{A} = -\frac{1}{2}m_{f}\left(\frac{d\boldsymbol{V}}{dt}(t) - \frac{D\boldsymbol{u}}{Dt}(\boldsymbol{X}(t), t)\right).$$
(2.53)

Excluding the Faxén corrections the added mass term is slightly different in the two cases, i.e. the two limits of creeping flow and inviscid fluid are not consistent. Maxey (1993) noted that the difference between the two formulations is negligible in the creeping flow limit if fluid flow gradients are small. Nonetheless, studies at small Reynolds numbers, e.g. by Coimbra and Kobayashi (2002) obtained the added mass force in the Auton-Hunt-Prud'homme formulation. It has therefore become customary to modify the Maxey-Riley

equations for spherical particles in most applications by replacing the added mass term with that of Eq. (2.53).

Similar to the drag force there exist many empirical corrections to the added mass terms, in particular for non-spherical particles. For a discussion of corrections to the added mass coefficients for different shapes the reader is referred to the book of Clift et al. (2005).

Basset-Boussinesq History Force

The Basset-Boussinesq history force represents the effects of the diffusion of vorticity around the particle. It is related to the temporal development of disturbances in the flow field around the particle surface, due to the boundary condition. It does not appear in the Auton-Hunt-Prud'homme formulation, as it depends on the fluid viscosity. The history force acts in the direction of the relative velocity and from a physical point of view represents an unsteady contribution to the drag force. The equation for the history force in the creeping flow limit was first derived by Basset (1888) and Boussinesq (1903). In the formulation of the Maxey-Riley equations it becomes

$$\boldsymbol{F}_{H} = -\frac{3}{2}\pi d_{p}^{2}\mu_{f} \int_{0}^{t} d\tau \frac{1}{\sqrt{\pi\nu_{f}(t-\tau)}} \frac{d}{d\tau} [\boldsymbol{V}(t) - \boldsymbol{u}(\boldsymbol{X}(t), t) - \frac{1}{24} d_{p}^{2} \nabla^{2} \boldsymbol{u}(\boldsymbol{X}(t), t)] . \quad (2.54)$$

This unsteady component of the drag is therefore a function of the particle acceleration along the entire particle trajectory. In many applications the history force is neglected, due to the computational problems associated with it. But in many cases it is not clear whether this is justified. An excellent discussion of this problem is found in Michaelides (1997).

Corrections to the history force for finite Reynolds number (Mei and Adrian, 1992), viscous particles (Mei et al., 1994) or non-spherical particles (Lawrence and Mei, 1995) have been proposed, but a general understanding is still missing.

Lift Force and Lift-Based History Force

In the Auton-Hunt-Prud'homme equations an additional surface force, the lift force appears that is absent from the Maxey-Riley equations. While lift forces do not appear in the creeping flow regime of the Maxey-Riley equations, in many applications at finite particle Reynolds number the lift force can be of a similar magnitude as the drag force and can therefore play a very important role (see e.g. Mazzitelli et al., 2003).

Generally, lift forces are created by vorticity in the surrounding fluid or rotation of the particle. For the quasi-steady lift force a number of different types are distinguished. The vorticity induced lift force appearing in Eq. (2.48) is the so-called Auton lift

$$oldsymbol{F}_L = rac{1}{2} m_f [\omega_f imes (oldsymbol{u}(oldsymbol{x},t) - oldsymbol{V}(t))]$$

Other general types of vorticity induced lift are the Saffman lift, which is generated by the motion of a particle in a linear shear (Saffman, 1965) and the Herron lift which is the lift in a vortex with solid-body rotation (Herron et al., 1975). Lift generated by particle rotation is usually called Robins-Magnus lift (Rubinow and Keller, 1961).

In addition to the quasi-steady lift force, there is an unsteady component to the lift force, similar to the history force for the drag. This lift-based history force has been derived for the Saffman lift conditions for solid spherical particles by Coimbra and Kobayashi (2002) and can become important when the frequency of the fluid vorticity is significantly large compared to the frequency of momentum diffusion in the fluid. However, almost no expression for the lift-based history force exist for finite Reynolds numbers or nonspherical particles.

Weak-Inertia Approximation

Of particular interest in many applications is the so-called weak inertia approximation for the motion of finite-size particles, where the particle response time τ_p becomes small. It was first introduced by Maxey (1987) as a limit-case of the Maxey-Riley equations. He showed that in the limit $\tau_p \ll \tau_f$, where τ_f is the smallest relevant time scale of the surrounding fluid the velocity of a particle can be given as a Taylor series in τ_p . In particular the history force and the Faxen corrections disappear in this limit and the equations of motion for a particle (2.42) reduce to

$$\frac{d\boldsymbol{X}}{dt} = \boldsymbol{u} + \tau_p \left(\boldsymbol{G} + (\check{\beta} - 1) \frac{D\boldsymbol{u}}{Dt} \right) + \mathcal{O}(\tau_p^2) . \qquad (2.55)$$

Practically, this means that up to first order in τ_p the particles move like tracers but in a modified velocity field $\mathbf{u}' = \mathbf{u} + \tau_p \left(\mathbf{G} + (\check{\beta} - 1)\frac{D\mathbf{u}}{Dt}\right)$. This modified velocity field \mathbf{u}' may be compressible, i.e. $\nabla \mathbf{u}' \neq 0$ even if $\nabla \mathbf{u} = 0$, leading for example to the clustering of particles in certain regions of the flow. However, problems associated with the description of inertial particles, such as caustics do not appear in this approximation. Therefore, both Eulerian and Lagrangian descriptions of the particle dynamics similar to those discussed in Ch. 2.2 for mixed-fluid conditions can be applied in the case of weak particle inertia.

2.3.3. One-Way Particle-Flow Coupling: Dissipative Particle Dynamics

In this section, the physics of one-way particle-flow coupled multiphase flows are considered and relevant results from the literature discussed. In this regime the particle concentration is assumed to be low enough, so that the fluid motion is not significantly affected by the presence of the particles. This is the case when both number and mass concentration of the particles are small, i.e. the mass and volume of particles in a certain volume is much less than the mass and volume of the surrounding fluid. This regime is particularly accessible for numerical modeling, since the equations of motion for the continuous phase can be solved independent of the equations of motion for the dispersed phase. This significantly reduces the complexity of the problem and is the reason why most numerical and theoretical studies are performed in this regime.

In the following, two recent results are discussed that capture many important aspects of the physics of suspensions of inertial particles. It is emphasized that these results were found in the one-way particle-flow coupling regime, where all interactions between the particles are neglected and only the influence of the surrounding flow on the particles is included. Other coupling regimes may lead to modified or even completely different particle dynamics.

Preferential Concentration

The effect of clustering, or preferential concentration of inertial particles was first mentioned in a work by Maxey (1987) who numerically studied the gravitational settling of aerosol particles in a simple cellular flow. He noted that particle trajectories tend to cluster in regions of low vorticity or high strain. He studied this effect in terms of the weak-inertia approximation for the particle equations of motion, introduced in Sec. 2.3.2. He noticed that the "synthetic" velocity field of the particles has negative divergence in regions of high vorticity or low strain and concluded that particles might be ejected from vortices in something like a "centrifugal mechanism". Numerical simulations, for example by Squires and Eaton (1991) and Wang and Maxey (1993) and experimental work, for example by Eaton and Fessler (1994) confirmed the clustering effect in turbulent flows. It is a rather remarkable observation, that turbulent flows which were generally believed to be very efficient at mixing can also lead to the opposite effect for suspensions of inertial particles. For an initially uniform distribution of particles, i.e. a well mixed state the occurrence of clustering means that particles and fluid can become "unmixed" over time, at least to a certain degree.

From the centrifugal mechanism suggested by Maxey it was predicted that clustering should be less apparent when the particles are overdamped or the velocity field fluctuates too fast for the particle density to follow. This is commonly characterized in terms of the Stokes number $St := \tau_p / \tau_f$ which is the ratio between the particle response time τ_p and the correlation time τ_f of the surrounding fluid:

- $St \ll 1$ particles follow the surrounding fluid closely (overdamped limit)
- St = O(1) particle motion is strongly affected by surrounding flow
- $St \gg 1$ only weak effect of the surrounding flow on the particle motion .

The strongest clustering effect is expected for St = O(1). This was indeed confirmed in experiments in a turbulent channelflow (Fessler et al., 1994) and numerical simulations (Duncan et al., 2005; Bec et al., 2006b).

A different approach to describe preferential concentrations, using methods from dynamical systems was used by Bec (2003, 2005). This follows an approach of Sommerer and Ott (1993) who studied the spatial structures of tracer particles in a random flow in terms of their fractal dimension. It can be easily seen from the equations of motion for inertial particles that due to the Stokes drag the phase space volume is contracting over time. This means that the dynamical system defined by the particle equations of motion is dissipative. Calculating the divergence of the right hand side of the equations of motion (2.42) leads to

$$V(t) = V(0) \cdot e^{-\frac{d}{St} \cdot t}$$

$$(2.56)$$

for a volume V in d-dimensional phase space. This means that generally the particles will collect on an attractor in phase space. In most realistic cases the particle dynamics is chaotic and the attractor will have a fractal structure. This structure can be characterized by its Lyapunov dimension d_L (Kaplan and Yorke, 1979).

The Lyapunov dimension is given by the Lyapunov exponents of a dynamical system, which is a measure of the exponential separation of neighboring trajectories. Chaotic dynamics is characterized by (at least) one positive Lyapunov exponent, whereas fixed points, periodic orbits and quasiperiodic behavior can be characterized by negative and/or zero Lyapunov exponents.

For two particles starting at an Euclidian distance d(0) and having a distance d(t) at time t, the largest Lyapunov exponent λ_1 is given by

$$\lambda_1 = \lim_{\substack{t \to \infty \\ d(0) \to 0}} \ln\left(\frac{d(t)}{d(0)}\right) . \tag{2.57}$$

In general, for a *d*-dimensional dynamical system there are *d* Lyapunov exponents λ_i , i = 1...d, measuring the exponential separation in *d* orthogonal directions, whereas λ_1 is the

exponent belonging to the direction of strongest separation.

Formally, the Lyapunov exponents of a differentiable dynamical system are defined as the logarithm of the limiting singular values of the Jacobian . The proof of their existence and uniqueness (almost) everywhere² is contained in the multiplicative ergodic theorem of Oseledec. Further details about Lyapunov exponents and the ergodic theory of dynamical systems in general can be found in the review by Eckmannn and Ruelle (1985). An algorithm for the computation of all Lyapunov exponents is given by Benettin et al. (1980).

Whereas a single Lyapunov exponent measures the exponential growth of distances, sums of the Lyapunov exponents measure the growth of surfaces and volumes in phase space. The idea of the Lyapunov dimension is that a phase space volume that has the same dimension as an attractor in a dynamical system will neither grow nor shrink over time. Therefore the dimension of the attractor should correspond to the number of exponents where the sum of the Lyapunov exponents crosses from positive to negative. Following the notation in Wilkinson et al. (2007) this reads as

$$d_L = k - \Delta$$
, with $\Delta = \frac{\lambda_1 + \ldots + \lambda_k}{\lambda_k}$, (2.58)

where k satisfies $\lambda_1 + \ldots + \lambda_{k-1} \geq 0$ and $\lambda_1 + \ldots + \lambda_k < 0$. The fraction Δ is called the dimension deficit. Fractal attractors in phase space are characterized by non-integer dimensions d_L , i.e. by $\Delta \neq 0$.

For inertial particles the phase space is typically 6 dimensional and even though a fractal dynamical attractor might exist in the phase space a projection onto the configuration space only leads to a fractal structure when $d_L < 3$. Otherwise, the attractor in the configuration space becomes area filling and no clustering appears. In terms of the Lyapunov exponents this means that the sum of the first three Lyapunov exponents should be negative for clustering to occur. However, if the sum is close to zero the clustering effect might be small since local stretching and folding of the clusters happens faster than the accumulation of particles.

Detailed numerical simulations by Bec (2003, 2005) showed how the fractal dimension of the spatial pattern in a suspension of inertial particles in a smooth random flow varies as a function of the Stokes number and particle-fluid density ratio β , revealing a maximum of the clustering for a Stokes number of order unity.

A local analysis of the dynamics, in terms of a linearization of the equations of motion for the inertial particles revealed that the stability of the eigendirections of the particle dynamics depends on the local structure of the surrounding flow. The eigenvalues of

²almost everywhere with respect to an ergodic, invariant probability measure on the attractor

the strain matrix $\partial u_i/\partial x_j$ of the flow distinguish the different cases. Bec found that in elliptic regions, where the strain matrix has two complex-conjugate eigenvalues for particles heavier than the surrounding fluid there is at least one unstable eigendirection, whereas for particles lighter than the surrounding fluid all eigendirections can be stable if the Stokes number is small enough. This means that particles heavier than the fluid tend to be ejected from regions of higher vorticity and therefore collect in the strain regions of the flow, whereas particles lighter than the fluid can cluster in the high vorticity regions of the flow.

However, Bec also pointed out that even though his local analysis seems to confirm the mechanism suggested by Maxey, in turbulent flows the persistence of such structures in generally not long enough to explain the observed clustering effect. Additionally, Bec numerically found clustering in flows delta-correlated in time, that are devoid of any structure. Instead, the appearance of clustering in configuration space is solely related to the dissipative character of the global particle dynamics, i.e. the existence of a chaotic, fractal attractor in phase space with a dimension lower than the spatial dimension of the system.

While the numerical calculations by Bec further quantified clustering effects, a detailed explanation was still missing. The most recent important contribution to this topic came from Wilkinson et al. (2007). They were able to explain this clustering effect, by studying the fluctuations of the particle density as a multiplicative random process of successive compression and expansion of volume elements. They showed how the mean particle density is related to the Lyapunov exponents of the system, recording the full history of the flow instead of instantaneous correlations with vortices. Additionally, they were able to calculate the first three Lyapunov exponents for inertial particle dynamics, which are the ones relevant for clustering. Their calculations revealed that in addition to the Stokes number, there is another dimensionless quantity in the system which they termed the Kubo number Ku, which is given by $Ku = \frac{\check{U}_f \tau_f}{\xi_f}$, where ξ_f is the correlation length in the fluid. The Kubo number can be considered as a measure of the flow "strength" and is of order one in fully developed turbulence and may be small in most other systems. Wilkinson et al. showed that instead of the Stokes number the relevant dimensionless quantity is the product $Ku\sqrt{St}$ and that clustering appears when this dimensionless quantity is of order one. For fully developed turbulence, where Ku = O(1) this is indeed the case for a Stokes numbers of order one, but when $Ku \ll 1$ clustering can also appear in the case of $St \gg 1$.

Formation of Caustics

In addition to preferential concentration there exists another physical effect in the dynamics of inertial particles which is very different from the behavior of tracers. This is the appearance of so-called caustics in the distribution of the particles. The name is derived from the analogy to optical caustics, but there are some differences in the two phenomena. In the context of particle dynamics in flows, the occurrence of caustics was first mentioned in a work by Falkovich et al. (2002), who studied the initiation of rain in a turbulent cloud. They pointed out that an increased collision rate, due to what they called a "sling effect" can lead to much faster growth of cloud droplets than could be explained with previous theories. However, their theory was fairly complicated and not very precise in its conclusions. The importance of this effect was then pointed out more clearly in a work by Wilkinson and Mehlig (2005) and a theory for the increasing of the collision rates was presented in Wilkinson et al. (2006).



Figure 2.1.: Sketch for the formation of a caustic showing the velocity of particles over their position along a line (adapted from Wilkinson et al. (2006)). At an initial time t_0 (blue line) the velocity is still a function of the position, but at a later time $t > t_0$ (red line) fast particles have overtaken slower ones. The manifold is folded in phase space and the velocity takes on multiple values at a single position between the two folds.

The main idea is that inertial particles, in particular when the particle response time τ_p is large, can detach from the trajectories of the fluid elements. Particles with a larger velocity can then overtake slower particle, so that at a given, arbitrarily small region in space there are particles with (possibly very) different velocities. The particle velocity has become multivalued and can no longer be treated as a velocity field. This effect can be most easily understood in terms of a one dimensional example (Figure 2.1), that has been taken from Wilkinson et al. (2006). At a starting time $t = t_0$ the particle velocity is initially single valued, but particles move towards each other. Faster ones overtake slower particles and then pass the ones moving the other way at at time $t > t_0$. The particle velocity is now multivalued.

The region of multivalued particle velocity is bounded by two fold caustics. It is obvious from the example that these caustics always appear in pairs. For three dimensional systems the occurrence of a caustic corresponds to a folding of the attractor of the particle dynamics in phase space. Again, the regions of multivalued particle velocities are bounded by a network of fold caustic lines. Along these lines, the particle density itself can diverge, analogous to the divergence of light intensity at optical caustics. But more important is that particles with very different velocities come arbitrarily close along these caustics, which leads to a greatly increased collision rate, something that has no counterpart in optical caustics.

Wilkinson and Mehlig (2005) discussed the long-term evolution of the caustics in 2dimensional flows and showed that different cases can be distinguished in terms of the two largest Lyapunov exponents. Depending on the properties of the surrounding flow, these can have different combinations of positive and/or negative values, leading to three distinct long-term states in two dimensions.

If both largest Lyapunov exponents are negative, particle paths will coalesce and the distribution of the particles will collapse onto points (Wilkinson and Mehlig, 2003). If the sum of the first two Lyapunov exponents is positive, the density fluctuations resulting from the caustic weakens for increasing time, leading to a random scattering of the particles. If the largest Lyapunov exponent is positive, but the sum of the first two exponents is negative, the concentration of the particles on the caustics increases in time leading to very sharp caustic lines. This process is limited by the stretching and folding of the caustic lines themselves. Therefore, for long times the particles aligned along the caustics persist in phase space, their effect on the spatial distribution of the particles weakens over time. However, this happens much slower than when the sum of the first two Lyapunov exponents is positive.

The appearance of caustics is the reason why for inertial particles there can be no equivalent Eulerian description of the particle dynamics. The fundamental assumption of the Eulerian approach, the existence of a well-defined particle velocity field is not met as soon as caustics appear in the particle dynamics. In this case either an approximation has to be made where caustics are neglected or a Lagrangian approach becomes necessary.

2.3.4. Two-Way Particle-Flow Coupling and Particle-Particle Interactions

The interaction between the dispersed phase and the continuous phase leads to modifications of the motion of the particles and the surrounding fluid. There are a number of aspects to this coupling between the phases that are often approximated separately. First, we discuss a two-way particle flow-coupling approximation for inertial particles that tries to account for the feedback of the particles on the fluid phase. Second, we discuss a further effect of the coupling between the phases that can be interpreted as an interaction between individual particles, mediated through the fluid. Finally, we discuss collisions between particles.

Two-Way Particle-Flow Coupling

The case of two-way particle-flow coupling for finite-size particles is very similar to the mixed-fluid case. The feedback of the dispersed phase on the continuous phase again leads to a modification of the continuous phase properties, such as density and viscosity as discussed in Sec. 2.2.3. However, for inertial particles there is an additional momentum coupling effect. Since for inertial particles the fluid exerts a force on the particles, the same force, with opposite sign is acting on the fluid at the particle position. Usually, this is done in terms of a multipole expansion of the forces. For a single particle smaller than the Kolmogorov scale, the small scale interactions that decay with $1/r^3$, where r is the distance from the particle position, are quickly dissipated by viscosity and the only effective term is the so-called Stokeslet that decays as 1/r. This term in the multipole expansion results from a delta-forcing approximation.

Following the approach presented in Mazzitelli et al. (2003), the form of this deltaforcing can easily be found by considering the momentum equation for a fluid velocity field \boldsymbol{v} in a volume V with a bounding surface S, where V contains a particle of volume V_p and surface S_p . Here \boldsymbol{v} is the full velocity field with the presence of the particles. The volume occupied by the fluid is $V_f = V - V_p$ and the momentum of the fluid is given by

$$\int_{V_f} \rho_f \frac{D\boldsymbol{v}}{Dt} dV = \int_{V_f} \rho_f \boldsymbol{g} dV - \oint_{S_p} \sigma \boldsymbol{n} dS + \oint_S \sigma \boldsymbol{n} dS , \qquad (2.59)$$

where n is a normal vector on the surface. The first term on the right hand side is the body force, the second term is the force at the particle surface (see also Sec. 2.1.1) and the last term on the right hand side is surface force at the outer boundary. Rewriting this, by using $V = V_f + V_p$ and the equations of motion for a particle (see Sec. 2.1.1)

$$m_p \frac{d\boldsymbol{V}}{dt} = \oint_{S_p} \sigma \boldsymbol{n} dS + m_p \boldsymbol{g} \tag{2.60}$$

one obtains

$$\int_{V} \rho_{f} \frac{D\boldsymbol{v}}{Dt} dV = \int_{V} (\nabla \cdot \boldsymbol{\sigma} + \rho_{f} \boldsymbol{g}) dV$$
$$-m_{p} \frac{d\boldsymbol{V}}{dt} + (m_{p} - m_{f})\boldsymbol{g} + m_{f} \frac{D\boldsymbol{v}}{Dt}.$$
(2.61)

The integral on the right hand side is the force in the undisturbed fluid, i.e. without the presence of the particles. The other terms therefore represent the momentum transfer between the particle and the fluid integrated over the volume V, the so-called interface force F_{int} . In this context a particle acts as a sink for fluid momentum.

By comparing with Sec. 2.1.1 and 2.3.2 one can see that the integrated interface force is simply the surface force plus the body force acting on the particle minus the fluid stress force from the undisturbed fluid at the position of the particle, i.e.

$$\boldsymbol{F}_{\text{int}} = -(\boldsymbol{F}_{\text{surf}} + \boldsymbol{F}_{\text{body}} - \boldsymbol{F}_{S}) . \qquad (2.62)$$

Equation (2.61) indicates that the feedback of the particles on the fluid can be understood in terms of a delta forcing. Replacing F_{int} in the integral momentum equation by

$$\boldsymbol{F}_{\text{int}} = \int_{V} \boldsymbol{f}_{\text{int}}(\boldsymbol{x}, t) dV , \qquad (2.63)$$

one obtains an expression for the local force generated by one particle

$$\boldsymbol{f}_{\text{int}}(\boldsymbol{x},t) := \left(-m_p \frac{d\boldsymbol{V}}{dt} + (m_p - m_f)\boldsymbol{g} + m_f \frac{D\boldsymbol{v}}{Dt}(\boldsymbol{x},t)\right)\delta(\boldsymbol{x} - \boldsymbol{X}(t)) , \qquad (2.64)$$

where the term $m_p \frac{d\mathbf{V}}{dt}$ represents the components of the surface force discussed in Sec. 2.3.2, such as drag, added-mass, history and lift forces.

Using Eq. (2.64) as an additional force term in the Navier-Stokes equation (2.4) for u and summing up this contribution over all particles, the feedback of the particles on the momentum of the continuous phase can be included. In combination with a concentration-dependent modification of the viscosity and density, as already discussed for the mixed-fluid approach in Sec. 2.2.3 this leads to a modification of the undisturbed flow field u, approximating the real flow field v.

It is remarked that the equations of motion for the dispersed phase discussed in Sec. 2.3.2, such as the Maxey-Riley equation contain the undisturbed velocity field \boldsymbol{u} . By including the feedback of the particles on the flow and using this perturbed velocity field in the equations of motion for the particles an error is incurred. For a grid-spacing Δx for the continuous phase, the error is of the order $d_p/\Delta x$ and therefore requires $d_p \ll \Delta x$. This error decreases with increasing number of particles, because the relative influence of each particle on a grid-point for the continuous phase becomes smaller.

Fluiddynamic Interactions

In addition to the modifications of the fluid density and viscosity and the added momentum term discussed above, the interaction between the dispersed phase and the fluid phase leads to a coupling between individual particles that is mediated through the surrounding fluid. This effect is not captured in the two-way particle-flow coupling approximation presented above and therefore needs to be addressed separately. In general, this problem of particles interacting through continuum-level forces poses many theoretical and computational difficulties. Many of these difficulties are related to the fact that the disturbance in the surrounding fluid created by the presence of a particle decays as 1/r, where r is the distance from the particle position. This long-range interaction results in nonconvergent interaction sums in systems with many particles. In this case the equations of motion between all particles in a suspension become coupled. Several procedures have been developed to approximate this friction matrix that couples the equations of motion, or alternatively its inverse, the so-called mobility matrix. For details on this, see for example the review by Brady and Bossis (1988) or for more recent results the article by Knudsen et al. (2008).

However, even with advanced methods the number of particles whose hydrodynamic interactions can be simulated directly is severely limited, usually to a few hundred which is not enough in most problems of interest. Therefore, it is often useful to model the particle-particle fluiddynamic interactions in an ensemble approximation by considering modifications to the surface forces due to a finite volume fraction of the dispersed phase.

Most of the work in this direction has been in trying to identify the effect of a finitevolume fraction C_V of the particles on the quasi-steady drag force for the dispersed phase. While some theoretical results in the limit of very small volume fractions and zero particle Reynolds number have been derived, e.g. by Richardson and Zaki (1954), generally empirical relationships are used. The expressions for the drag force at finite volume fraction are usually given in form of a correction factor f_C , that relates the drag force at a given volume fraction $F_D(C_V)$ to the undisturbed drag force F_D of a single particle, i.e.

$$f_C := \frac{F_D(C_V)}{F_D} . \tag{2.65}$$

For a given particle Reynolds number, the most commonly used formulation for the drag correction, which has been found to be consistent for small and medium volume fractions, even at larger particle Reynolds numbers is (see e.g. Maude and Whitmore, 1958)

$$f_C = (1 - C_V)^{1 - b_0} , (2.66)$$

with an empirical coefficient $b_0 \approx 4.5$. Similar empirical expressions for the drag correction, including the dependence on the particle Reynolds number can be found for example in DiFelice (1994).

The influence of the volume fraction on the other surface forces acting on a particle is even less understood than for the drag force. For the added-mass force (Zhang and Prosperetti, 1994), the history force (Ten Cate and Sundaresan, 2006) and the lift force (Prosperetti, 2007) it was found that a reasonable first-order approximation is to take the force proportional to the effective volume of the surrounding fluid, i.e. proportional to $(1 - C_V)$. The fluid stress force F_S , with proper averaging within a computational cell, is generally found to be independent of the volume fraction of the dispersed phase.

The general first-order approximation for the surface force acting on a particle in a dilute suspension of spheres, as a function of the volume fraction of the dispersed phase can therefore be written as

$$\boldsymbol{F}_{\text{surf}}(C_V) \approx \boldsymbol{F}_S + f_C \boldsymbol{F}_D + (1 - C_V)(\boldsymbol{F}_A + \boldsymbol{F}_H + \boldsymbol{F}_L) . \qquad (2.67)$$

Collision Forces

In addition to fluiddynamic interactions, particles in suspension can also interact directly, in form of collisions. Generally, to determine whether particle dynamics are influenced significantly by collisions one can consider the ratio of particle response time τ_p to the time scale of collisions τ_{coll} , which is the inverse frequency of collisions. This ratio defines a Stokes number

$$St_{\rm coll} = \tau_p / \tau_{\rm coll}$$
 (2.68)

Particle-Particle collisions can be neglected if $St_{coll} \ll 1$, whereas in dense flows, where particle dynamics are dominated by collisions one finds $St_{coll} \gg 1$. The inclusion of particle collisions can leads to new effects in the dynamics of the dispersed phase, such as the appearance of intermittent "bursts" pointed out by Medrano et al. (2008).

When particles with centers of gravity at X_1 and X_2 collide, they change their momenta in the direction of the contact vector $d = \frac{X_1 - X_2}{||X_1 - X_2||}$ in such a way that the total momentum is conserved.

Let P_{1-} and P_{2-} be the momentum of the particles before the collision and P_{1+} and P_{2+} the momentum of the particles after the collision. Then momentum conservation means that

$$P_{1-} + P_{2-} = P_{1+} + P_{2+} , \qquad (2.69)$$

or

$$P_{1+} = P_{1-} + ad$$
 and $P_{2+} = P_{2-} - ad$, (2.70)

with some constant a.

In the case of an elastic collision in addition to the total momentum of the particles the total kinetic energy of the particles is also conserved. The constant a in Eq. (2.70) then needs to be chosen in such a way to account for this conservation of total kinetic energy.

Conservation of total kinetic energy means that

$$\frac{P_{1-}^2}{m_1} + \frac{P_{2-}^2}{m_2} = \frac{P_{1+}^2}{m_1} + \frac{P_{2+}^2}{m_2} , \qquad (2.71)$$

where m_1 and m_2 are the masses of the two particles. This can be rewritten using Eq. (2.69):

$$\frac{1}{m_1} \left(\mathbf{P}_{1-}^2 - \mathbf{P}_{1+}^2 \right) = \frac{1}{m_2} \left(\mathbf{P}_{2+}^2 - \mathbf{P}_{2-}^2 \right)$$

$$\Leftrightarrow \quad \frac{1}{m_1} \left(\mathbf{P}_{1-} - \mathbf{P}_{1+} \right) \left(\mathbf{P}_{1-} + \mathbf{P}_{1+} \right) = \frac{1}{m_2} \left(\mathbf{P}_{2+} - \mathbf{P}_{2-} \right) \left(\mathbf{P}_{2+} + \mathbf{P}_{2-} \right)$$

$$\stackrel{(2.69)}{\Leftrightarrow} \qquad \qquad \frac{1}{m_1} \left(\mathbf{P}_{1-} + \mathbf{P}_{1+} \right) = \frac{1}{m_2} \left(\mathbf{P}_{2+} + \mathbf{P}_{2-} \right)$$

$$\Leftrightarrow \qquad \qquad \left(\frac{\mathbf{P}_{1-}}{m_1} - \frac{\mathbf{P}_{2-}}{m_2} \right) = -\left(\frac{\mathbf{P}_{1+}}{m_1} - \frac{\mathbf{P}_{2+}}{m_2} \right) . \tag{2.72}$$

Multiplying Eq. (2.72) by d and replacing P_{1+} and P_{2+} with the expressions in Eq. (2.70) leads to

$$a = \frac{2\left(m_1 P_{2-} d - m_2 P_{1-} d\right)}{m_1 + m_2} .$$
 (2.73)

With this, the velocity of the particles after the collision can be calculated.

In the case of an inelastic collision the total kinetic energy of the particles is not conserved during the collision, instead a part of the kinetic energy is dissipated during the deformation of the particles. The most convenient way to implement this is by using a coefficient of restitution ϵ , which is defined as the negative ratio of the velocity difference in the direction of contact after the collision and before the collision, i.e.

$$d\left(\frac{P_{1+}}{m_1} - \frac{P_{2+}}{m_2}\right) = -\epsilon d\left(\frac{P_{1-}}{m_1} - \frac{P_{2-}}{m_2}\right) .$$
 (2.74)

The value of ϵ can vary between 0 and 1, where $\epsilon = 0$ is the totally inelastic case, where all kinetic energy in the direction of contact is dissipated upon collision and $\epsilon = 1$ is the totally elastic case described above, where the total kinetic energy is conserved. In the same way as for the elastic case the value of a can be derived using Eqs. (2.74) and (2.70). This leads to

$$a = \frac{(\epsilon + 1) (m_1 P_{2-} d - m_2 P_{1-} d)}{m_1 + m_2} , \qquad (2.75)$$

for the case of an inelastic collision.

Collision Rates

In addition to quantifying the influence of collisions on the particle dynamics, there is a great interest in many fields to determine the collision frequencies or collisions rates between particles in a given system. This is in particular true where active processes of the particles take place upon collision. These processes often involve either a chemical or biological reaction (e.g. Nishikawa et al., 2001) when particles come together or particles can physically stick together upon collision, either by merging (coagulation) or by forming larger clusters composed of individual smaller particles (aggregation). Coagulation typically happens for viscous particles, e.g. drops or bubbles whereas aggregation happens for solid particles.

Often, due to limitations in computational resources one does not want to simulate the particle dynamics directly to determine for example the time evolution of the size distribution of the particles. Instead, the evolution of the particle size distribution for the whole system is written as a system of rate equations for the change in the number of particles of a given size. This approach was first introduced by Smoluchowski (1917) and has since then been extensively used in many fields of science, for example colloid science, biological systems and aerosol dynamics.

While computationally usually much more efficient than simulating each individual particle, the problem in this approach lies in the construction of appropriate kernels for the rate equations, such as collision kernels (collision rates) $Q(r_1, r_2)$. The collision kernel can be understood as the average rate at which two particle of radius r_1 and r_2 reach a distance equal to $r_1 + r_2$. The collision kernel $Q(r_1, r_2)$ between two particles of radius r_1 and r_2 is usually defined as

$$N_c(r_1, r_2) = Q(r_1, r_2) N_1 N_2 , \qquad (2.76)$$

where N_c is the number of collisions per time and N_1 and N_2 are the numbers of particles in the domain. A generalized collision relationship was developed by Sundaram and Collins (1997), who showed that the collision kernel can be written as

$$Q(r_1, r_2) = \frac{1}{2} 4\pi (r_1 + r_2)^2 g(r_1 + r_2) \int_{-\infty}^0 -w_r P(w_r | r_1 + r_2) dw_r .$$
 (2.77)

Here w_r is the radial velocity difference between particles, i.e. the difference between the particle velocities projected on their separation vector $X_1 - X_2$. $P(w_r|r)$ is the conditional probability density function of the radial velocity difference, conditioned on the distance r and g(r) is the radial distribution function. The radial distribution function is the number of particles with a distance between r and r + dr from a center particle divided by the number of particles expected if the particle distribution were uniform³.

A number of different contributions to the collision kernel have been identified theoretically in the past, such as collisions due to Brownian motion or laminar shear rate in the flow (see e.g. Friedlander, 2000, Ch. 7). For mixed-fluid conditions, i.e. the case of vanishing particle Stokes number, the classical result for the collision kernel due to turbulent shear is given by Saffman and Turner (1956) in terms of the dissipation of turbulent kinetic energy ϵ and the fluids kinematic viscosity ν_f

$$Q_{\rm ST}(r_1, r_2) = \left(\frac{8\pi}{15}\right)^{1/2} \left(\frac{\epsilon}{\nu_f}\right)^{1/2} (r_1 + r_2)^3 .$$
 (2.78)

Except for the prefactor this result is identical to the collision kernel in laminar shear.

Other major contributions to the collision kernel generally come from finite-size effects. While collision kernels due to different settling velocities W of particles without hydrodynamic interaction are easily obtained and can be written as

$$Q_{\Delta W}(r_1, r_2) = \pi (r_1 + r_2)^2 |W_1 - W_2| \quad , \tag{2.79}$$

the effect of particle inertia on the collision rates is still not well understood. A notable exception is the limit of large particle inertia where particles move ballistically leading to the result for the collision kernel of Abrahamson (1975)

$$Q_{\rm Ab}(r_1, r_2) = 5 \left(\bar{V}_1^2 + \bar{V}_2^2 \right)^{1/2} (r_1 + r_2)^2 , \qquad (2.80)$$

where \bar{V}_1 and \bar{V}_2 are the root-mean-square velocities of the two particles.

One of the main questions in recent years has been to bridge the gap between the limit cases of zero and infinite inertia, the Saffman-Turner and the Abrahamson cases. This is the regime where particle inertia effects, such as preferential concentration discussed in Sec. 2.3.3 and the appearance of caustics (see Sec. 2.3.3) lead to strong deviations of the particle dynamics from the mixed-fluid case. The effect of preferential concentration appears in the radial distribution function, whereas the appearance of caustics and

³The definition of the radial distribution function as a function of only the separation r between particles assumes a statistically homogeneous and isotropic volume.

detachment of particles from the fluid trajectories affects the distribution of the relative velocity.

In recent years, Lagrangian simulations of particle collisions in turbulent flows have been found to be a very useful tool to determine collision kernels for particles with inertia. For example the work of Ayala et al. (2008a,b) provides a detailed description of the numerical calculation of collision rates of inertial particles in a DNS simulation of a turbulent flow. Typically, simulations are performed in the case of one-way particle-flow coupling, using the so-called 'ghost collision' approach. The idea is to let particles pass through each other after they approach a distance equal to the sum of their radii. The collision event has no physical consequence, but is simply counted to estimate the rate of collisions. In cases where the mean free time between collisions is large, compared to the convergence time to a steady state the ghost collision approach alone is expected to give good approximations of the collision rates. It is therefore of particular relevance in the case of very dilute suspensions (e.g Zhou et al., 1998; Wang et al., 1998).

Extensions of this approach are discussed for example in Reade and Collins (2000) who pointed out that the influence of elastic particle collisions on the collision rate can be approximated in terms of an additive correction to the radial distribution function for the ghost collision approximation.

In the work of Wilkinson et al. (2006) the importance of caustics for the collision rates of finite size particles was pointed out. They developed a model for the collision rate between particles of the same size that bridges the gap between the Saffman-Turner and Abrahamson formulation. They showed that the collision rate varies approximately as a function of the rate of caustic formation J, where

$$J = J_0 \exp(-S/\mathcal{I}) \tag{2.81}$$

with some constant J_0 . Here, \mathcal{I} is proportional to Ku^2St and S is the action of a trajectory and a function of the Kubo number Ku (see Sec. 2.3.3 for the definition of the Kubo number). For the collision kernel they found that a good assumption over a wide range of Stokes and Kubo numbers is given by

$$Q = Q_{ST} + \exp(-S/\mathcal{I})Q_A b , \qquad (2.82)$$

where Q_{ST} is the Saffman-Turner collision kernel and $Q_A b$ the Abrahamson collision kernel.

A detailed phenomenological model for the collision kernel of polydisperse suspensions, i.e. for particles with different radii r_1 and r_2 was given by Bec et al. (2005). They found that there are three different regimes of the particle Stokes numbers, where different functional form of the collision kernel appear.

Assuming that the dimensionless equations of motion for the particles are given by

$$\frac{d\mathbf{V}}{dt} = \frac{1}{St}(\mathbf{u} - \mathbf{V}) , \qquad (2.83)$$

i.e. only Stokes drag is considered one can write an equation for the time evolution of the separation $\Delta X = X_1 - X_2$ between two particles

$$\frac{d\Delta \mathbf{X}}{dt} = \mathbf{V}_1 - \mathbf{V}_2$$

$$\frac{d(\mathbf{V}_1 - \mathbf{V}_2)}{dt} = \frac{1}{\langle St \rangle (1 - \theta^2/4)} (\Delta \mathbf{u} - (\mathbf{V}_1 - \mathbf{V}_2))$$

$$- \frac{\theta}{\langle St \rangle (1 - \theta^2/4)} (\langle \mathbf{u} \rangle_E - \langle (\mathbf{V}_1 - \mathbf{V}_2) \rangle_E) . \quad (2.84)$$

Here, $\langle St \rangle = \frac{1}{2}(St_1 + St_2)$ is the mean Stokes number of the particles, $\theta = \frac{St_1 - St_2}{\langle St \rangle}$ is the relative Stokes number difference, $\Delta u = u(X_1) - u(X_2)$ the difference in the fluid velocities and $\langle u \rangle_E$ and $\langle V \rangle_E$ are ensemble averaged fluid and particle velocities, respectively.

For small relative Stokes number differences, i.e. $|\theta| \ll 1$ the result for the collision rates depends on the characteristic length scale $l^* := L_f |\theta|$. For particle separations smaller than l^* the differences in particle motion are dominated by the Stokes number difference, i.e. the second term in Eq. (2.84). For larger separation scales, the differences are dominated by the differences in fluid motion, i.e. the first term in Eq. (2.84). For relative Stokes number differences of order one, particles see each other as an ensemble of uniformly distributed particles with independent velocities.

While much progress has been made in recent years in understanding the collision kernels for inertial particles, in particular in the regime of intermediate Stokes numbers, a full understanding is still missing.

One interesting question, in particular with regard to the next parts of this work, is whether collision rates of inertial particles can be approximated well from studying particles with an equation of motion that only contains quasi-steady drag forces. This would mean that the transient forces in the equations of motion, such as added mass and history forces can be neglected when the details of individual particle trajectories are not of interest. To this end, in this work the collision rates for particles with different approximations for the equations of motion were computed numerically in a synthetic, three-dimensional, homogeneous, isotropic turbulent flow field (see Appendix A) and the influence of the quasi-steady drag force, added mass force and history force compared. All collision rates have been computed using the ghost collision approach, i.e. any interaction between the particles has been neglected.



Figure 2.2.: Influence of drag, added mass and history term in the equations of motion (2.42) for heavy droplets, with $\tau_p = 2000r^2 \text{ s/mm}^2$ and $\beta = 5 \times 10^{-4}$ in a synthetic turbulent flow (see Appendix A) with $\epsilon = 1$, $T_f = 1$, $L_f = 1$. (a) Collision rates $Q(r_1, r_2)$ between particles with different sizes and (b) ratio q_H between the collision rate for the full equations of motion, i.e. with drag, added mass and history forces Q_{D+A+H} versus equations of motion with only drag forces Q_D .

For particles much heavier than the surrounding fluid, such as small water droplets moving in air, the computed collision rates for equations of motion with only quasisteady drag forces can be seen in Fig. 2.2(a). The result is identical to that of Bec et al. (2005). Figure 2.2(b) shows the enhancement factor q_H for the collision rates if the full Maxey-Riley equations of motion (2.42) are used. The enhancement factor is defined as the ratio between the collision rate in the case of the full equations of motion with quasi-steady drag, added mass and history force Q_{D+A+H} and the collision rates in the case of the equations of motion with only quasi-steady drag forces Q_D , i.e.

$$q_H := Q_{D+A+H} / Q_D \ . \tag{2.85}$$

It can be readily seen that the collision rates are typically larger when the full equations of motion are considered. The enhancement factor increases with increasing particle size and can become as large as 4-5 in the range of particle sizes studied here.

This can be seen in more detail by looking at cross-sections of the collision rates. To this end r_2 is fixed at 0.02 mm and the collision kernel for varying r_1 is shown (Fig. 2.3(a)). Three different equations of motion are compared. First, when only quasi-steady drag forces are included (blue squares), second with quasi-steady drag and added mass term (red triangle) and third the full equations of motion with quasi-steady drag, added mass and history integral (green circle).



Figure 2.3.: Influence of drag, added mass and history term in the equations of motion (2.42) for heavy droplets, with $\tau_p = 2000r^2 \text{ s/mm}^2$ and $\beta = 5 \times 10^{-4}$ for particles with different sizes, r_2 fixed at 0.02 mm in a synthetic turbulent flow (see Appendix A) with $\epsilon = 1$, $\tau_f = 1$, $l_f = 1$ (a) Collision rates $Q(r_1, r_2)$ and (b) ratio q_H between the collision rate for the full equations of motion with drag, added mass and history force Q_{D+A+H} versus equations of motion with only drag forces Q_D . The linear fit in (b) is done over the right tail of the curve, for $r_1 > 0.035$ mm. All values are averages, obtained from ten realizations of the carrier flow. Grey shaded area denotes plus/minus one standard deviation.

While the inclusion of the added mass force does not seem to lead to any observable changes in the collision rates compared to the case of only quasi-steady drag forces, when the history force is included the collision rate increases much faster with increasing particles size. The enhancement factor q_H is shown in Fig. 2.3(b)). There is a great increase in the collision rates between particles of the same size, and for $r_1 > r_2$ the enhancement factor increases almost linearly with r_1 .

Looking at the equations of motion (2.42) it is possible to get an idea why the history term seems to be so much more important that the added mass term in this case and why its importance increases with increasing particle size. For particles much heavier than the surrounding fluid the mass ratio parameter β is small, $\beta = 5 \times 10^{-4}$ in the case shown here. While the added mass force is proportional to β , the history force is proportional to $\beta^{1/2}$. Therefore, for small values of β one can expect a larger contribution from the history force to the total force acting on the particle.

Additionally, the quasi-steady drag force is proportional to $1/\tau_p$ and therefore proportional to d_p^{-2} , whereas the history force is proportional to $\sqrt{1/\tau_p}$ and therefore proportional to d_p^{-1} . Absolutely, both quasi-steady drag force and history force decrease with increasing particle size, but the ratio between history force and quasi-steady drag force will increase approximately proportional to d_p . This means that the contribution of the history force to the overall force acting on the particle should increase approximately linearly with d_p .

In many cases the collision rates between particles of the same size is also of special interest. Figure 2.4(a) shows the computed collision rates in this case for equations of motion with only quasi-steady drag and for the full equations of motion. For particles of the same size the increase in the collision rates due to the presence of the history integral seems to be even larger than for particles of different sizes. For all sizes, except the very smallest, the enhancement factors q_h between 2 - 4 can be found (Fig. 2.4(b)). Again, the enhancement seems to increase with increasing particle size, but in this case there appears to be a slow saturation towards larger particle sizes.

Together, our results indicate that in many of the previous works where only the Stokes drag has been used in the equations of motion the collision rates between heavy inertial particles are underestimated.

The situation is different when particles of similar density than the surrounding fluid are considered. This is for example the case for marine aggregates in the ocean. There, the particle response time τ_p is usually very small but the mass ratio β is of order one. In this case one might be tempted to forget about inertia effects altogether and treat the particles as tracers.

In this case the collision rate would be given by the classical result of Saffman and Turner, see Eq. (2.78). However, for realistic values of the particle response time $\tau_p = 2r^2 \text{ s/mm}^2$ and the mass ratio $\beta = 0.5$ our results (Fig. 2.5) still show a significant difference between the collision rates for tracers (black triangles) and inertial particles,

at least for particles $\geq 50\mu m$. Again, the difference between the various equations of motion increases with increasing particle size. In all cases the collision rate increases approximately proportional to $(r_1 + r_2)^3$, but with different prefactors.



Figure 2.4.: Influence of drag, added mass and history term in the equations of motion (2.42) for heavy droplets, with $\tau_p = 2000r^2 \text{ s/mm}^2$ and $\beta = 5 \times 10^{-4}$ for particles of the same size in a synthetic turbulent flow (see Appendix A) with $\epsilon = 1$, $T_f = 1$, $L_f = 1$ (a) Collision rates $Q(r_1, r_2)$ and (b) ratio q_H between the collision rate for the full equations of motion with drag, added mass and history force Q_{D+A+H} versus equations of motion with only drag forces Q_D . All values are averages, obtained from ten realizations of the carrier flow. Grey shaded area denotes plus/minus one standard deviation.

In this case, where β is of order one, the added mass force contributes significantly to the total force acting on the particle. It can be seen that the collision rates for particles subjected to both quasi-steady drag and added mass force (red triangles) is reduced compared to particles with only drag forces (blues squares). This may be due to that fact that particle motion is slowed down by the presence of the added mass term. However, if the history force is also included (green circles), the collision rates again become larger. As was already found for heavy particles, the presence of the history force seems to lead to increased collisions between particles. In this case, the increase of the collision rates due to the history force almost balances the decrease of the collision rates due to the presence of the added mass force.



Figure 2.5.: Collision rates: comparing the influence of drag, added mass and history term in the equations of motion (2.42) for small particles with similar density than the surrounding fluid, with $\tau_p = 2r^2 \text{ s/mm}^2$ and $\beta = 0.5$ in a synthetic turbulent flow (see Appendix A) with $\epsilon = 1, T_f = 1, L_f = 1$. All values are averages, obtained from ten realizations of the carrier flow. Grey shaded area denotes plus/minus one standard deviation.

In general, our results show that in addition to modifying individual particle trajectories changes in the particle equations of motion can lead to significant changes in the ensemble behavior and in particular the collision rates. It is therefore very likely that neglecting certain parts of the equations of motion when calculating the collision rates between particles leads to significant over- or underestimating of the collision rates. In addition to the presence of the transient forces such as added mass and history force, contributions to the equations of motion due to two-way particle-flow coupling and particle-particle coupling can likely also lead to very different collision rates.

2.4. Conclusions

In this chapter we discussed basic approaches to model particle-laden flows. We analyzed a number of different approximations of the full set of equations for a suspension of particles in a fluid flow and presented some results from the literature for the dynamics of such particle suspensions. The aim of this chapter was to provide a solid background on the most important facts when trying to model particle dynamics in a flow. In the rest of this work we will apply this knowledge to the problem of aggregation and fragmentation of particles. We will be using an inertial particle based approach, i.e. a separated-fluid approximation where the particle motion can deviate significantly from that of the surrounding fluid. In many applications, e.g. for the collision-coalescence process of droplets in warm clouds particles are sufficiently large so that inertial effects become important. The results in the last section showed that due to effects such as preferential concentration and caustics, collision rates for inertial particles deviate strongly from those of tracer particles. It is a key feature of our new approach discussed in this thesis that these effects are included directly within the model, with no need for further approximations.

To obtain reasonable statistics for the size distributions of aggregates we will need to study systems containing at least $10^4 - 10^6$ particles. Therefore, using very detailed models to calculate the surface forces on the particles, such as semi-resolved or resolved surface force models is computationally not possible. Instead, we will be working with the point-force approximation discussed in detail in this chapter. We restrict ourselves to the simplest possible description of the problem and only include the most important component of the surface force, namely the quasi-steady drag force in our description. The reason for this simplification is that the main goal of this thesis is to present a new approach for the modeling of aggregation-fragmentation systems and not a detailed analysis of a specific case. We discuss general results for aggregation-fragmentation systems that can be found within the framework of such a model and show for certain examples how the approach can be applied. We do not try to model any specific systems with all possible details or attempt a full comparison with experimental data. Therefore, the simplest possible description that captures the essential features of such aggregationfragmentation systems is suitable for the goal we have in mind here. For the same reason we will only analyze aggregation and fragmentation in the case of one-way particle-flow coupling, where the dynamics of the particles can be calculated independently from that of the surrounding fluid.

Nonetheless, the inertial-particle based model we present in the following chapters readily allows the inclusion of further terms in the equations of motion, such as added mass or history terms as well as two-way particle flow coupling and fluiddynamic interactions if they are deemed necessary for the modeling of a specific problem.

3. Modeling Coagulation and Breakup of Spherical Droplets

This chapter contains, with some minor changes, our work on individual-particle based modeling of coagulation and breakup of liquid droplets in a synthetic turbulent flow which has been published as Zahnow and Feudel, Nonlinear Processes in Geophysics, 16, 677– 690, 2009.

3.1. Introduction

In this chapter we present results from an inertial particle based model for coagulation and fragmentation of heavy drops suspended in a synthetic turbulent flow. The main new idea of this approach is to follow the motion of each individual coagulate in a Lagrangian framework. We have initially proposed and discussed such a model for particles in chaotic advection in Zahnow et al. (2008) and Zahnow et al. (2009) to help bridge the gap between full hydrodynamic simulations and rate-equation based approaches for coagulation and fragmentation.

The simulation of multiphase flows using full hydrodynamical models for each particle, e.g. with a finite element approximation in an arbitrary Lagrange Eulerian framework (see e.g. Maury, 1999) can be very detailed but is usually restricted to an extremely small number of particles (Higashitani et al., 2001; Zeidan et al., 2007). On the other hand, a rate-equation based approach, in the framework of which one treats the problem of particle motion as a field equation and then uses the Smoluchowski equation (Smoluchowski, 1917) to model coagulation and fragmentation exhibits a number of different problems. For example, the particle velocity may take on several values even at the same location of inertial particles when the dynamical attractor of the particles folds in the full velocity-position phase space (Bec et al., 2005). Due to the presence of such 'caustics' (Falkovich et al., 2002; Bec, 2003; Wilkinson and Mehlig, 2005), a velocity field cannot be well founded and the rate-equation based description relies on many assumptions and parameterizations.

Here, we follow the model approach presented in Zahnow et al. (2008, 2009) which represents a new 'intermediate-level' take on the problem. Instead of modeling each individual primary particle and the complete structure of a coagulate explicitly, primary particles are 'combined' upon collision, neglecting or parameterizing in some way the full structure of the coagulate. These coagulates are instead represented as simple, spherical particles moving in the flow. However, by still treating the coagulates as individuals, instead of a concentration or particle field a description that is much more detailed than the usual rate-equation based approaches can be obtained. In particular, relating coagulation and breakup directly to the properties of the individual coagulates involves one less approximation step than deriving the corresponding rate equations. Additionally, particle inertia can easily be included in such an individual-based approach, using for example the equations of motion discussed previously in Sec. 2.3 of this thesis.

Here, we focus on the impact of particle properties and flow properties on the steady state size distribution of the drops that develops from the balance between coagulation and fragmentation in a synthetic turbulent flow. We study two different fragmentation mechanisms. First, particles break up if their size exceeds a certain maximum allowed size. This is motivated by the hydrodynamical instability of liquid drops, for example rain drops settling due to gravity (Villermaux, 2007). Second, particles fragment if the shear forces due to the fluid flow are too strong (see e.g. Thomas et al., 1999). In contrast to our previous approach we use here a lognormal distribution for the number of fragments, and compare this with other common approaches, such as binary or ternary fragmentation.

Section 3.3 presents our results obtained from this model. We see that the distribution of particles as well as the mean average size in the steady state depends on the type of fragmentation mechanism taking place. First, when fragmentation occurs solely due to the particles exceeding a maximum stable size, the distribution is fairly uniform over all the appearing coagulate sizes. Second, for fragmentation occurring under sufficiently large shear, the distributions typically decay exponentially beyond a certain coagulate size. In the case of shear fragmentation the mean average size of the coagulates depends strongly on the particle properties and the flow properties, such as the coagulate strength or the volume fraction of the particles. We show numerically that as a good approximation the influence of each parameter on the steady state can be treated separately and determine a decomposition of the average coagulate size in the steady state with respect to the particle and flow properties. We find that the average coagulate size changes as a power law function of the particle and flow parameters, where the exponents of the power law are determined by the exponent in the stability condition for the shear fragmentation.

The fluctuations over time in the coagulate size distribution increase for larger mean average coagulate sizes. This is found to be a nonlinear effect resulting from the shear fragmentation process. A similar decomposition and power law relationship with respect to the particle and flow properties as for the average coagulate size can be found here.

For fragmentation due to particles exceeding a maximum allowed size the dependence on the particle properties is much weaker, instead the steady state depends mainly on the maximum stable coagulate size. However, the time to reach the steady state can vary greatly for different particle and flow properties. For example, flows with weak dissipation of turbulent energy allow for very large coagulates because there is almost no shear fragmentation, but since collisions also occur mainly due to shear the growth of coagulates is extremely slow.

In Sec. 3.4.3 we show how in principle a scaling relationship for the average size of coagulates in the steady state can be derived, if size distributions, collision rates and fragmentation rates are known. Using approximate expressions for these quantities we again find a power-law relationship for the average coagulate size as a function of the particle and flow properties. Our calculations illustrate the dependency of the exponents of the power-law on the stability condition for the shear fragmentation.

Section 3.5 gives a brief summary and states some conclusions.

3.2. Numerical Model

In this section we present the mathematical model that will be the framework of our study. It contains a detailed description of the dynamics of particles with inertia in a dilute suspension and a model for coagulation and breakup of spherical droplets.

3.2.1. Equations of Motion

We study suspensions of spherical inertial particles of radius r_p , transported in an incompressible flow with dynamic viscosity μ_f . We assume that the suspension is very dilute, i.e. particle-particle hydrodynamic interactions and two-way particle-flow coupling can be neglected. Additionally, we focus on a carrier flow with moderate Reynolds number and study only spatial scales below the Kolmogorov scale η where the flow is sufficiently smooth. We therefore rescale space, time and velocity by the Kolmogorov length η , time η^2/ν_f and velocity ν_f/η , where ν_f is the kinematic viscosity of the fluid.

Assuming that the Reynolds number based on the particle size as well as the difference between the particle velocity $\mathbf{V}(t)$ and the flow velocity $\mathbf{u}(\mathbf{X}, t)$ is small and the particle density ρ_p is much higher than the density ρ_f of the surrounding fluid, the motion can be approximately described by the Stokes equation. This is a simplified form of the translational equation of motion (2.42) of a spherical particle that includes only quasisteady (Stokes) drag. In dimensionless form it reads as

$$\dot{\boldsymbol{V}} = \frac{1}{St_{\eta}} \left(\boldsymbol{u}(\boldsymbol{X}, t) - \boldsymbol{V} \right) , \qquad (3.1)$$

where $\mathbf{X}(t)$ is the *d* dimensional position of the particle and $St_{\eta} = (2r_p^2 \rho_p \nu_f)/(9\eta^2 \mu_p)$ is the dimensionless Stokes number. The effect of gravity has been neglected.

3.2.2. Coagulation

Next, we describe the model for coagulation that is used in this study.

The smallest particles considered will be called primary particles. These can combine upon collision to form larger particles, called coagulates. All coagulates are assumed to consist of an integer number α of these primary particles, i.e. the primary particles can never be broken up. A coagulate consisting of α primary particles has a radius $r_{\alpha} = \alpha^{1/3}r_1$, where r_1 is the radius of the primary particles. The coagulates Stokes number depends on the radius, and therefore on α , with $St_{\eta,\alpha} = \alpha^{2/3}St_{\eta,1}$. Here $St_{\eta,1}$ is the Stokes number for the primary particles. After the coagulation of two particles the velocity of the new particle follows from momentum conservation and the position is the center of gravity of the two old particles. To ensure that no collisions are missed, we use an efficient event-driven algorithm for particle laden flows (cf. Sigurgeirsson et al. (2001) for details). Since hydrodynamic interactions between coagulates, that may affect the collision rates, are not included in such a model we approximate this by implementing a collision efficiency χ_c , which is the probability to coagulate upon collision. If particles do not coagulate upon collision they collide elastically.

3.2.3. Breakup

For the breakup of droplets two different cases will be discussed. It is known that liquid droplets can break up either due to a hydrodynamical instability when moving with a certain critical velocity relative to the surrounding fluid, for example when settling under gravity or the droplets can be broken up due to shear in the surrounding fluid. For both cases the relevant results from the literature are discussed briefly in the following and then for each case a simplified model suitable for the purpose of this work is developed.

Breakup due to Gravitational Settling

The breakup of liquid droplets settling under gravity has received considerable attention in the past, in particular in the context of atmospheric science where the distribution of raindrops falling out of a cloud is of interest, see e.g. Pruppacher and Klett (1997). In particular the measurements by Marshall and Palmer (1948) indicated that there seems to be a fairly general mechanism at work and that the distribution of drops is solely related to the rate of rainfall. Recently, the work of Villermaux (2007) showed in very detailed experiments the breakup of liquid drops when falling under gravity. In particular, Villermaux and Bossa (2009) demonstrated that the Marshall-Palmer result follows directly from the fragment size distribution of an individual breaking drop. The mechanism they found is related to a topological change in the droplet size. It requires that the pressure acting on a droplet falling with a velocity ΔV relative to the surrounding fluid overcomes the capillary pressure that is trying to preserve the droplet shape. The surrounding pressure is given by $\rho_f(\Delta V)^2$, while the capillary pressure is given by σ_p/d_p , where σ_p is the surface tension of the droplet and d_p the initial, spherical diameter of the droplet. The ratio of these pressures gives the Weber number

$$We = \frac{\rho_f (\Delta V)^2 d_p}{\sigma_p} \tag{3.2}$$

which should therefore exceed some critical value for the droplet to break up. In particular, a droplet falling under gravity in a low viscosity liquid such as air will initially be flattened into a disc. The disc will deform into a bag-shape, with a thicker rim which is followed by a break-up of the bag. Finally, if the Weber number for the particle is large enough, a hydrodynamical instability in the rim will lead to the complete breakup of the drop. In some cases the formation of a bag-shape does not appear, instead the flattened disc will immediately deform into a ligament, which then breaks due to a hydrodynamical instability if the critical Weber number is exceeded.

For a droplet falling under the influence of gravity the terminal settling velocity in turbulent conditions is given by $V_{\rm s} \approx \sqrt{\frac{\rho_p}{\rho_f}gd_p}$. Assuming that the droplet needs to exceed a critical Weber number We_c this leads to a critical droplet diameter

$$d_{p,c} = \sqrt{\frac{We_c}{\sigma}}_p g\rho_p \tag{3.3}$$

which needs to be exceeded before a droplet will break up. For raindrops falling in air Villermaux and Bossa (2009) found that the critical Weber number is approximately 6 in all situations. From these results it is concluded that breakup of particles due to gravitational settling can be modeled in a very simplified way by imposing a fixed critical size for the droplets, beyond which they will break. While this might not capture small fluctuations in the critical size due to changes in the relative velocity of the drops the essence of the problem can be captured in this way. In the context of this work, the following model for breakup due to gravitational settling (size-limiting fragmentation) is therefore used:

Size-limiting breakup: If a particle becomes larger than some maximum number of

primary particles α_{max} , it is broken up into k smaller fragments. For the *i*th fragment, where $1 \leq i < k$ we set the new number of primary particles α_i to a random number drawn from a normal distribution centered around $(\alpha_{old} - \sum_{j=1}^{i-1} \alpha_j)/(k-i+1)$ and with a standard deviation one, rounded to the nearest integer greater or equal to one. The last fragment contains the remaining primary particles. This means that typically fragments will be of very similar sizes. The number of fragments is set to $k = 2 + \xi$, where ξ is a random number from a lognormal distribution with standard deviation one, rounded towards the nearest integer. Such a distribution of fragments is a very common assumption for the fragmentation of drops, but later we will also comment on the implications of different choices for the number of fragments.

Breakup due to Fluid Shear

Breakup of liquid droplets due to shear forces in a fluid is an important mechanism in many natural problems and engineering applications such as production of emulsions or polymer blends. The mechanisms leading to breakup and the resulting fragment size distributions have been studied intensively for many years. Typically, expressions for the critical shear required to break up a droplet are measured experimentally (see e.g. the review by Tucker III and Moldenaers (2002)) or calculated theoretically (Taylor, 1934; Delichatsios, 1975) in Stokes flow conditions. A recent study combining both experimental results and adaptive-mesh simulations has been performed by Cristini et al. (2003). They found that breakup of a droplet of initial radius r_p due to fluid shear can be characterized in terms of a critical capillary number Ca. The capillary number is the ratio of the characteristic time scale τ_{σ} of the relaxation of the droplet surface due to the surface tension σ_p and the time scale τ_S of the fluid shear S_f . Taking

$$\tau_{\sigma} = \mu_f \frac{r_p}{\sigma_p} \tag{3.4}$$

$$\tau_S = 1/S_f , \qquad (3.5)$$

where μ_f is the viscosity of the fluid, the capillary number is

$$Ca = \frac{\mu_f r_p S_f}{\sigma_p} . \tag{3.6}$$

Cristini et al. (2003) found that a droplet breaks if the capillary number is smaller than some critical capillary number Ca_c . This critical capillary number is a function of the viscosity ratio $\tilde{\mu}$ between droplet and surrounding fluid. They showed that the critical capillary number diverges for $\tilde{\mu} \to 0$ and $\tilde{\mu} \to \tilde{\mu}^* \approx 3$. This means that breakup due to fluid shear only occurs if droplet and surrounding fluid have similar viscosities. For the distribution of the fragments after breakup both their experimental and numerical results showed that the droplet is initially stretched and then the middle section is pinched off creating two major 'daughter' droplets of similar size at each end. The remaining percentage of the original droplet is distributed among a series of approximately 3 different classes of smaller satellite droplets in the middle.

Following these results, a simplified model for breakup due to fluid shear has been developed for this work.

Shear breakup takes place when the hydrodynamic force acting on the particle exceeds the forces holding the coagulate together by a certain factor. The hydrodynamic force in this case is proportional to the shear force $S_f := (2S_{ij}S_{ij})^{(1/2)}$, where $S_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial X_j} + \frac{\partial u_j}{\partial X_i} \right)$ is the rate-of-strain tensor in the flow. Solving Eq. (3.6) for the shear force leads to an equation for the critical shear as a function of the droplet radius r_{α}

$$S_{f,c}(r_{\alpha}) = \frac{\gamma r_1}{r_{\alpha}} = \gamma \alpha^{-1/3} , \qquad (3.7)$$

where $\gamma := Ca_c \frac{\sigma_p}{\mu_f \cdot r_1}$ is a constant, the coagulate strength parameter that depends on the physical properties of the particle and the surrounding fluid. If the shear force, calculated across the radius of the drop exceeds the threshold value given by Eq. (3.7), the particle is broken up in the same way as for size-limiting fragmentation.

The centers of the fragments are placed at a distance equal to the sum of their radii, perpendicular to the direction of the velocity and keeping the original center of gravity. The magnitude of the velocity remains the same to ensure momentum conservation.

3.3. Simulation Results

In this chapter the results from the simulation of the model for coagulation and breakup of spherical droplets described in the previous section are discussed. First, the specific flow and the model parameterization used to obtain the results is described and then the approach to a steady state as well as the characterization of the coagulate size distribution in the steady state are discussed. Finally, the influences of the droplet and flow properties on the steady state are illustrated.

3.3.1. Fluid Flow

To be able to perform long-term simulations at reasonable computational costs we consider synthetic turbulence in the form of a incompressible, space-periodic, isotropic and homogeneous Gaussian random flow (Bec, 2005). Such flows are constructed to reproduce
certain features of turbulent flows, but can not capture all aspects of real turbulence, such as non-Gaussian tails of the velocity fluctuations and the energy cascade between scales. For further details on the construction of synthetic turbulent flows see Appendix A of this work.

The flow is written as a Fourier series

$$\boldsymbol{u}(\boldsymbol{X},t) = \sum_{\boldsymbol{k} \in \mathbb{Z}^d \setminus \{\boldsymbol{0}\}} \hat{\boldsymbol{u}}(\boldsymbol{k},t) e^{i\frac{2\pi}{L}\boldsymbol{k} \cdot \boldsymbol{x}} , \quad (3.8)$$

where $\hat{\boldsymbol{u}}(\boldsymbol{k},t) \in \mathbb{C}^d$ are the Fourier components, with the property $\hat{\boldsymbol{u}}(-\boldsymbol{k},t) =$ $\hat{\boldsymbol{u}}^*(\boldsymbol{k},t)$ because $\boldsymbol{u}(\boldsymbol{X},t)$ is real-valued. The star denotes complex conjugation. By taking for $\hat{\boldsymbol{u}}(\boldsymbol{k},t)$ the projection of a different vector $\hat{\boldsymbol{v}}(\boldsymbol{k},t) \in \mathbb{C}^d$ onto the plane perpendicular to the wave vector \boldsymbol{k} , incompressibility is ensured. The vector $\hat{\boldsymbol{v}}(\boldsymbol{k},t)$ is assumed to be an Ornstein-Uhlenbeck process. It is a solution of the complexvalued stochastic differential equation



Figure 3.1.: Snapshot of the position of 50000 particles with $St_{\eta} = 1$ transported in a synthetic turbulent flow with dissipation $\epsilon = 1$, correlation length $L_f = 1$ and correlation time $T_f = 1$. Due to dissipation the particles collect on a random attractor.

$$d\hat{\boldsymbol{v}} = -\xi(\boldsymbol{k})\hat{\boldsymbol{v}}dt + \sigma(\boldsymbol{k})\boldsymbol{dW} , \qquad (3.9)$$

with $\xi(\mathbf{k}), \sigma(\mathbf{k}) \in \mathbb{R}$, where dW is a *d* dimensional complex Wiener increment. The parameters $\xi(\mathbf{k}), \sigma(\mathbf{k})$ need to be chosen in such a way that the flow $u(\mathbf{x}, t)$ reproduces some features of a real turbulent flow, in this case the energy spectrum in the dissipative range of a turbulent flow. Here we use the exponential spectrum suggested by Kraichnan

$$E(k) = C \cdot (2\pi k L_f / \hat{L})^3 \exp(-\beta [2\pi k L_f / \hat{L}]) , \qquad (3.10)$$

with $\beta = 5.2$ (see e.g. Martinez et al., 1997) and a suitably chosen normalization constant C. The constant L_f is the length scale of coherent structures in the flow and \hat{L} is the spatial period of the flow. We choose $\xi(k) = 1/T_f$ and $\sigma(k) = \sqrt{cE(k)}$. The constant T_f is then the correlation time of the flow. The normalization constant is chosen in such a way that $2\nu k^2 E(k)$ sums to a desired value of the dissipation of turbulent kinetic energy ϵ . The flow is then characterized by the correlation time T_f , the correlation length L_f and the dissipation ϵ .

If a fluid velocity field with few Fourier modes is chosen, no interpolation of the velocity at particle position is required, since it can be calculated from direct summation of the Fourier series. This allows for a resolution of the fine structures of the particle distribution in space.

To reduce the computational effort only the case where the fluid flow depends only on two coordinates is treated here, i.e. a three dimensional flow where the velocity in the third direction is negligible compared to the other two directions is studied. Such a flow can then be represented as two dimensional. Comparisons were made with the full three dimensional case and no significant difference was found, except for a slowing down of the whole process due to a decreased number of collisions. A total of 8 spatial Fourier modes in two dimensions is taken into account, this is the lowest number for which isotropy is guaranteed. The period of the flow is $\hat{L} = 2\pi$. The correlation length of the flow is set to $L_f = 1$, the correlation time to $T_f = 1$ and the dissipation of turbulent kinetic energy to $\epsilon = 1$. This choice of parameters results in large coherent structures in the flow (compare Fig. 3.1) and a fast convergence to a steady state, due to sufficient collisions.

3.3.2. Model parameters and mean sizes

The primary particles have a radius $r_1/L_f = 5 \times 10^{-4}$ and the Stokes parameter $S_{\eta,1} = 0.05$. For a 'typical' flow situation of liquid water droplet in a moderately turbulent cloud this corresponds to primary particles in the range of $10^{-5}m$ radius. We choose $N_1 = 10^5$ primary particles as initial condition. This implies a 2-d volume fraction of particles of approximately 0.08.

The primary particles are uniformly distributed in the flow, with velocity V(0) = u(X(0), 0). Due to the limited number of primary particles, only a certain range of system parameters is available. In particular the 'standard' values for the maximum stable coagulate size and the coagulate strength parameters are chosen such that in the steady state most primary particles have formed larger coagulates, but the number of coagulates is still large enough to allow reasonable statistics. For size-limiting fragmentation the standard value is set at $\alpha_{max} = 80$ and for shear fragmentation the standard coagulate strength is $\gamma = 3.5$. The standard value for the collision efficiency is $\chi_c = 1$. Unless mentioned otherwise, these are the parameters used to obtain the following results.

To characterize the steady state for the average number of primary particles per coagulate in the case of size-limiting fragmentation one could simply use the value for $t \to \infty$. This is not very precise, because the steady state is not static as the mean of the coagulate size distribution fluctuate randomly over time. However, because the velocity field is stationary, the mean number of primary particles in a coagulate $\langle \alpha(t) \rangle$ will converge



Figure 3.2.: Average coagulate size as a function of time for (a) size-limiting breakup with $\alpha_{\text{max}} = 80$ and (b) shear breakup with $\gamma = 3.5$. The black line indicates the value of the asymptotic average coagulate size α_{∞} .

towards a constant value $\alpha_{\infty} = \lim_{t \to \infty} \frac{1}{\Delta T} \int_{t}^{t+\Delta T} ds \langle \alpha(s) \rangle$ when averaged over a time interval ΔT to remove random fluctuations. This quantity is used to characterize average coagulate sizes in steady state. Additionally, the standard deviation σ_{∞} of the size distribution is computed as a measure of the width of the distribution. To remove random fluctuations σ_{∞} is also calculated as an average over a time interval ΔT , in the same way as α_{∞} . Here, $\Delta T = 100$ is chosen for the averaging time, which was found to be a sufficiently long time interval to remove the fluctuations in the steady state.

3.3.3. Approach to a Steady State

Looking at the time evolution of the average coagulate size $\langle \alpha(t) \rangle$ it is found that the system generally converges to an asymptotic steady state. Figure 3.2 shows the average coagulate size as a function of time for the case of size-limiting breakup (Fig. 3.2(a)) and for shear breakup (Fig. 3.2(b)

Initially, coagulation leads to a fast increase in the average coagulate size, independent of the breakup mechanism. As coagulates become larger, breaking sets in and a balance between coagulation and breakup is reached. The asymptotic average coagulate size α_{∞} depends on the breakup mechanism and the particle and flow properties such as coagulate strength γ . This development of a steady state is a typical behavior in coagulationfragmentation systems. It has been observed both in experimental studies (Oles, 1992) and theoretical studies (Spicer and Pratsinis, 1996b).

Generally, fluctuations in the average coagulate size are larger in the steady state than during the transient. Additionally, the strength of the fluctuations in the steady state depends on the breakup mechanism. For size-limiting breakup, where the critical size of aggregates is fixed, fluctuations are small. For shear breakup, where the critical size depends on the flow the fluctuations in the average coagulate size are much larger, following fluctuations in the surrounding fluid.

3.3.4. Droplet Size Distributions in the Steady State

First, the size distribution in the case of size-limiting breakup for the same parameter values as in the previous part are discussed. In this case the size distribution is fairly



Figure 3.3.: Size distributions of the number of primary particles in coagulates, normalized by the total number of coagulates: (a) size-limiting fragmentation for two different values of the maximum stable coagulate size α_{max} and (b) shear fragmentation, for two different values of the coagulate strength.

broad, covering almost the complete range between the smallest size and the maximum allowed size distribution (dashed line in Fig. 3.3(a)). Comparison with other simulations for different fragmentation mechanisms shows that the shape of the size distribution depends greatly both on the number and on the size distribution of the fragments that are created during fragmentation. Here, the results are compared with the case of binary, ternary and quarternary fragmentation, i.e. the creation of two, three or four fragments instead of the lognormal distribution of the number of fragments described in the previous section (see Fig. 3.4). In particular, the width of the distribution is greatly influenced by this change in the number of fragments. Binary fragmentation leads to a single, sharp peak. Ternary fragmentation leads to two broader peaks in the distribution and for quarternary fragmentation three peaks can be seen. These peaks merge into a broad plateau if the number of fragments is not deterministic but instead can vary as is the case for the lognormal fragmentation mechanism.

In the case of shear fragmentation the situation is slightly different. The distribution is fairly broad, but with a long exponential tail towards larger size classes (Fig. 3.3(b)). The figure clearly shows the increase in the width of the distribution with increasing coagulate strength, that was also indicated by the standard deviation (compare Fig. 3.5). However, here it becomes obvious that the statistics degrades rapidly with increasing γ , as the number of coagulates available in the system is decreasing and hence the exponential tail becomes less visible. Especially in the tails of the distributions fluctuations become very large, as only a few coagulates of these sizes exist at all. For shear fragmentation different numbers of fragments, for example binary, ternary or



Figure 3.4.: Size distributions of the number of primary particles in coagulates, normalized by the total number of coagulates for size-limiting fragmentation for a critical size of $\alpha_{max} = 60$ for different distributions of the number of fragments, two (binary, solid line), three (ternary, dash-dotted line) and four (quarternary, dashed line).

quarternary fragmentation do not influence the shape of the size distribution, only the mean of the size distribution is shifted towards lower values for an increasing number of fragments.

The exponential tail of the size distribution is a feature that has also been observed for coagulation and fragmentation of marine aggregates in tidal flats (e.g. Lunau et al., 2006). A numerical comparison of different fragmentation mechanisms showed that this exponential tail is a typical feature, when the coagulates are assumed to break into fragments of very similar sizes. However, different distributions of the fragments, for example an erosion-like process where some very small and some larger fragments are created lead to different size distributions of the coagulates.

It is noted that the shape of the size distributions remains constant when the particle and flow properties, e.g. the coagulate binding strength γ or the collision efficiency χ_c are varied. The size distributions will collapse onto each other for different parameter values when rescaled with the mean coagulate size. This self-similarity property of the size distribution is a typical feature of coagulation and fragmentation processes and is discussed for example in Spicer and Pratsinis (1996a).

It is also noted that in the parameter ranges studied here, the tails of the size distributions consist of coagulates with Stokes numbers of order 1 ($\alpha \sim 100$), which is of the order of the correlation time of the flow. This strongly affects the clustering properties of these particles (see e.g. Cencini et al., 2006; Bec et al., 2007)]. However, since in our case there are typically only a very few particles in this range of Stokes numbers, this is not expected to significantly influence the properties of the steady state size distributions which we focus on.

3.3.5. Influence of Droplet and Flow Properties

In the following it is examined how the limiting values α_{∞} and σ_{∞} depend on the properties of the particles and the properties of the flow for the different fragmentation mechanisms. The large parameter space makes it difficult to interpret results from the model. We therefore consider the sensitivity of the results to each of the parameters separately. We restrict ourselves to the four most relevant parameters, namely the maximum stable coagulate size α_{max} (for size-limiting breakup), the coagulate strength γ (for shear breakup), the collision efficiency χ_c , the volume fraction of the particles, characterized by the total number N_1 of primary particles and the dissipation of turbulent kinetic energy ϵ in the flow.

Coagulate strength

First, the dependence of the average number of primary particles per coagulate α_{∞} on the maximum stable coagulate size α_{max} and the coagulate strength γ is examined. These two parameters determine the binding strength of aggregates for the different breakup cases.

Figure 3.5(a) shows the results for the case of size-limiting fragmentation. α_{∞} and σ_{∞} both increase with the maximum stable coagulate size α_{max} . Here, we find that $\alpha_{\infty} \propto \alpha_{max}$ and $\sigma_{\infty} \propto \alpha_{max}$. The proportionality constant is determined by the distribution of fragments during breaking, a fit gives a proportionality constant of approximately 0.5 for α_{∞} and 0.25 for σ_{∞} . The proportionality constant depends on the details of the fragmentation mechanism. For example, the case of binary fragmentation, i.e. splitting into two fragments shows the same scaling but proportionality constants of approximately 0.66 and 0.2, respectively.



Figure 3.5.: Variation of the binding strength of coagulates. (a) For the case of size-limiting fragmentation the average number of primary particles per coagulate (triangles) and width of the size distribution in the steady state (squares) as a function of the maximum stable coagulate size α_{max} . The fits are $0.5\alpha_{max}$ for the average (solid line) and $0.25\alpha_{max}$ for the standard deviation (dashed line). (b) For the case of shear fragmentation the average number of primary particles per coagulate (triangles) and width of the size distribution in the steady state (squares) as a function of the coagulate strength γ . The fits are $2.2\gamma^{2.6}$ for the average (solid line) and $2.0\gamma^{2.6}$ for the standard deviation (dashed line). The grey shaded area represents error bars obtained from an ensemble of 10 different simulation runs.

Figure 3.5(b) shows the results for the case of shear fragmentation. A first qualitative estimate of the shape of this $\alpha_{crit}(\gamma)$ curve can be derived from a scaling argument, which was already mentioned in Zahnow et al. (2008). Solving Eq. 3.7 for a given value of the shear in the fluid we obtain a critical coagulate size for this shear. This critical coagulate size is proportional to γ^3 . We therefore expect that α_{∞} scales the same way. This is indeed close to the result of the numerical simulations, where we determine a relationship $\alpha_{\infty}(\gamma) \propto \gamma^{2.6\pm0.1}$ (dashed line in Fig. 3.5(b)). A more detailed theoretical argument for this scaling will be given in Sec. 3.4.

However, since the shear in the flow fluctuates over space and time there is no single

critical size for coagulates. We therefore expect that the width of the size distribution will depend, among other factors, on the fluctuations of the shear in the flow. From Eq. 3.7 it follows that larger coagulates are more sensitive to fluctuations in the shear. This can be seen by considering how a change of the shear from \tilde{S} to $\tilde{S} + \Delta S$ changes the value of α_{crit} . We obtain

$$\Delta \alpha_{crit} = \gamma^3 \left((\tilde{S} + \Delta S)^{-3} - (\tilde{S})^{-3} \right) , \qquad (3.11)$$

i.e. the fluctuations in the value of α_{crit} are expected to increase proportionally to γ^3 . We therefore expect that the width of the size distribution will also increase proportional to γ^3 . This is again similar to the result of the simulations, where we find $\sigma_{\infty}(\gamma) \propto \gamma^{2.6 \pm 0.1}$ (dotted line in Fig. 3.5(b)).

Collision efficiency

Second, we examine the influence of the collision efficiency χ_c on the average number of primary particles per coagulate.



Figure 3.6.: Average number of primary particles per coagulate (triangles) and width of the size distribution (squares) in the steady state as a function of the collision efficiency χ_c , i.e. the probability to coagulate upon collision in the case of (a) size-limiting fragmentation and (b) shear fragmentation. The fits are $54.62\chi_c^{0.31}$ for the average (solid line) and $44.7\chi_c^{0.31}$ for the standard deviation (dashed line). The grey shaded area represents error bars obtained from an ensemble of 10 different simulation runs..

For size-limiting fragmentation the simulations indicate (Fig. 3.6(b)) that in this case there is almost no dependence of the average number of primary particles per coagulate in steady state on the collision efficiency. Both α_{∞} and the width of the size distribution σ_{∞} remain almost constant with varying χ_c . However, it should be noted that while the collision efficiency does not seem to have a large impact on the steady state, the transient behavior is greatly influenced by the collision efficiency. In particular, the time to reach the steady state increases greatly with decreasing χ_c , for both size-limiting and shear fragmentation.

For shear fragmentation an increase in collision efficiency increases both the average number of primary particles per coagulate α_{∞} and the width of the size distribution σ_{∞} (Fig. 3.6(b)). The numerical results suggest a dependency of the form $\alpha_{\infty}, \sigma_{\infty} \propto \chi_c^{0.31\pm0.03}$.

This increase of the average and width of the size distribution with increasing collision efficiency can be understood in terms of the balance between coagulation and fragmentation. In the steady state the size distribution, and therefore all its moments, including the average and the width of the size distribution are determined by the balance between coagulation and fragmentation. Increased coagulation due to increased collision efficiency requires a corresponding increase in the fragmentation, which in turn requires larger coagulates. Unfortunately, deriving an equation for these moments of the size distribution from this balance condition requires the apriori knowledge of the shape of the size distribution and equations for the collision and fragmentation rates. It is therefore not a trivial task. In Sec. 3.4 we show how such a calculation can be carried out if size distributions, as well as collision rates and fragmentation rates are known. This calculation will formalize the above argument and show more clearly how the scaling of the steady state with the particle and flow properties can be understood.

The reason why such a scaling does not happen in the case of size-limiting fragmentation is that this specific fragmentation rule serves as a 'brick wall' for the size distribution. For all coagulates below the critical size the fragmentation probability is zero, for all coagulates above it it is one. Therefore, a balance between coagulation and fragmentation is only possible for one specific size, independent of how coagulation is increased or decreased.

Volume fraction

The third important parameter of the system is the volume fraction of particles in the flow. This is defined as the ratio of particle volume to fluid volume. The volume fraction is a function of both the total number of primary particles in the flow N_1 and of the

radius r_1 of the primary particles. Since the impact of varying r_1 is very similar to that for varying N_1 we only focus on the variation of the total number of primary particles (Fig. 3.7).

For size-limiting fragmentation we again find almost no dependence of the steady state on the initial number of primary particles (see Fig. 3.7(a)), only the time to reach the steady state decreases with increasing number of primary particles. For shear fragmentation our results show that the average number of primary particle per coagulate in the steady state as well as the width of the coagulate size distribution increase with N_1 (see Fig. 3.7(b)). The numerical results suggest a relationship of the form $\alpha_{\infty}, \sigma_{\infty} \propto N_1^{0.3\pm0.03}$. This dependence on the number N_1 of primary particles in the system can be understood in the same way as for the collision efficiency χ_c , since both parameters increase the coagulation probability in the system. We will also illustrate this in the calculations in Sec. 3.4.



Figure 3.7.: Average number of primary particles per coagulate (triangles) and width of the size distribution (squares) in the steady state as a function of the total number of primary particles N_1 in the flow in the case of (a) size-limiting fragmentation and (b) shear fragmentation. The fits are $1.3N_1^{0.3}$ for the average (solid line) and $1.1N_1^{0.3}$ for the standard deviation (dashed line). The grey shaded area represents error bars obtained from an ensemble of 10 different simulation runs.

Dissipation of turbulent kinetic energy

Finally, we examine the impact of the flow on the simulation results. The mixing properties of the flow can be easily varied by adjusting the dissipation of turbulent kinetic energy ϵ in the flow. A higher ϵ results in higher shear in the flow and therefore influences both the coagulation and the shear fragmentation. Coagulation increases with increasing ϵ , since the collision rate due to shear increases but shear fragmentation also increases with ϵ .



Figure 3.8.: Average number of primary particles per coagulate (triangles) and width of the size distribution (squares) in the steady state as a function of the dissipation of turbulent kinetic energy ϵ in the flow in the case of (a) size-limiting fragmentation and (b) shear fragmentation. The fits are $52.92\epsilon^{-1.2}$ for the average (solid line) and $29.04\epsilon^{-1.2}$ for the standard deviation (dashed line). The grey shaded area represents error bars obtained from an ensemble of 10 different simulation runs.

For size-limiting fragmentation, again we find almost no dependence of α_{∞} and σ_{∞} on the parameter (Fig. 3.8(a)). The steady state seems to be independent of the changes in the flow. For both fragmentation rules the transient behavior is strongly influenced by the value of ϵ . For small ϵ collision rates in the flow are very low and the system takes a very long time to reach the steady state. For example, for shear fragmentation with $\epsilon = 0.1$, the system is still in a transient behavior for t = 500, while for $\epsilon = 1$ the system reaches its steady state at approximately t = 25. In the case of shear fragmentation, the average number of primary particles per coagulate in the steady state is proportional to $\epsilon^{-1.2\pm0.1}$, and the width of the size distribution also decreases approximately proportional to $\epsilon^{-1.2\pm0.1}$ (Fig. 3.8(b)). This means that fragmentation increases faster with ϵ than coagulation.



Figure 3.9.: Scaling exponents λ_i for the different parameters $(\gamma, \chi_c, N_1, \epsilon)$ as a function of the inverse exponent for the critical shear $1/\xi$ (Eq. (3.7)). The scaling exponents are linear functions of $1/\xi$. (a) Scaling exponents λ_1 for the coagulate strength γ and λ_4 for the dissipation of turbulent kinetic energy in the flow ϵ . The fits are $\lambda_1 = 0.273 + 0.763 \cdot \frac{1}{\xi}$ and $\lambda_4 = 0.091 - 0.429 \cdot \frac{1}{\xi}$. (b) Scaling exponents λ_2 for the collision efficiency χ_c and λ_3 for the number of primary particles N_1 . The fits are $\lambda_2 = 0.126 + 0.061 \cdot \frac{1}{\xi}$ and $\lambda_3 = 0.117 + 0.063 \cdot \frac{1}{\xi}$. The grey shaded area represents error bars obtained from an ensemble of 10 different simulation runs.

Discussion

In summary, we find that for size-limiting fragmentation the average number of primary particles per coagulate in the steady state and the width of the coagulate size distribution are mainly determined by the maximum stable coagulate size α_{max} . For the fragmentation mechanism used here we find $\alpha_{\infty} \approx 0.5 \alpha_{max}$ and $\sigma_{\infty} \approx 0.25 \alpha_{max}$.

For shear fragmentation the average number of primary particles per coagulate in the steady state and the width of the coagulate size distribution vary greatly with all system parameters. We obtain a relationship of the form

$$\alpha_{\infty}, \sigma_{\infty} \propto \gamma^{\lambda_1} \chi_c^{\lambda_2} N_1^{\lambda_3} \epsilon^{\lambda_4} , \qquad (3.12)$$

where the scaling exponents λ_i are given by $\lambda_1 = 2.6 \pm 0.1$, $\lambda_2 = 0.31 \pm 0.03$, $\lambda_3 = 0.3 \pm 0.03$, $\lambda_4 = -1.2 \pm 0.1$.

Further simulations confirmed that the scaling relationship for each parameter can indeed be approximately determined independently of the value of the other parameters. This means that Eq. (3.12) is expected to be valid for this model for all reasonable values of the system parameters, i.e. parameter values that lead to $1 \ll \alpha_{\infty} \ll N_1$. We note that both for increasing N_1 and χ the average coagulate size scales with an exponent of 0.3 ± 0.03 . This indicates that it is equivalent to vary the number of particles or the collision efficiency since both influence the average coagulate size in the same way.

When looking at the exponents λ_i for each parameter, one can ask the question whether these are related to the specific form of the stability condition Eq. (3.7) and in particular to the exponent appearing in this equation, as the simplified argument for the dependency of α_{∞} on γ in Sec. 3.3.5 suggests.

To examine this connection between the scaling exponents λ_i and the exponent of the fragmentation condition in Eq. (3.7), simulations with different exponents in Eq. (3.7) were performed. For example, changing the exponent for the critical shear to -1/2instead of -1/3 leads to a corresponding change in the exponents λ_i for the parameters. In this case we obtain $\lambda_1 = 1.8 \pm 0.1$, $\lambda_2 = 0.24 \pm 0.02$, $\lambda_3 = 0.23 \pm 0.02$, $\lambda_4 = -0.77 \pm 0.05$. Corresponding results were found for several other exponents for the critical shear, see Fig. 3.9. This clearly illustrates how the dependence of the steady state on the particle and flow properties is influenced by the fragmentation mechanism. We find that a fragmentation rule of $S_{f,c} = \gamma \alpha^{-\xi}$ leads to a relationship of the form of Eq. (3.12), where the scaling exponents are linear functions of $\frac{1}{\xi}$.

We also mention that in addition to the steady state discussed here the transient behavior of the system is greatly influenced by the particle and flow properties. One of the relevant quantities for this transient is the time it takes for the system to reach the steady state, which is for example very important in the formation of rain in clouds. This time decreases strongly with an increase of the coagulation rate, for example due to an increase of shear or collision efficiency. An increase in fragmentation, for example due to increased fluid shear or coagulate strength, also decreases the time to reach the steady state (see also Ch. 4 of this work).

3.4. Rate Equation Approach for Coagulation and Fragmentation

In this section we try to give some theoretical insight into the scaling of the average coagulate size in the steady state. We emphasize that it is not possible to find a closed equation for this average size of coagulates in the steady state, mainly because expressions for the collision rates and fragmentation rates are not known.

The question of the collision rates of inertial particles is a topic of ongoing research, and while some advances have been made (see e.g. Bec et al., 2005), even for the fairly simple flow situation used in this paper no equations exist that could be applied. For the fragmentation rates, the situation is slightly different. While the fragmentation rate corresponding to the fragmentation model used here is known in principle, the integral involved can not be solved in closed form.

In the following, we will make some approximations about the collision rates and fragmentation rates and try to derive an equation for the average coagulate size in the steady state. While this approach makes it clear how a scaling of the average size with particle and flow properties comes about, the result from the particle based model can not be fully recovered. This derivation also illustrates many of the difficulties associated with a rate-equation based approach for coagulation and fragmentation, which do not appear in the particle based approach described in this work.

3.4.1. Equations for the Moments of the Droplet Size Distribution

To estimate the scaling behavior of the average number of primary particles in the steady state we start by assuming a continuous probability distribution $p(\alpha, t) = \frac{C(\alpha, t)}{C(t)}$ of the coagulates¹. Here, $C(\alpha, t)d\alpha$ is the number concentration of coagulates, i.e coagulates per volume consisting of a number of primary particles in the range $[\alpha, \alpha + d\alpha]$ at time t and $C(t) = \int_0^\infty d\alpha C(\alpha, t)$ is the total number concentration of coagulates at time t.

¹In our model the distribution is in fact discrete, but the resulting sums can not be evaluated analytically.

The average of α with respect to p is then defined as

$$\langle \alpha \rangle := \int_{0}^{\infty} d\alpha \ \alpha p(\alpha, t) = \int_{0}^{\infty} d\alpha \ \alpha \frac{C(\alpha, t)}{C(t)} \ . \tag{3.13}$$

From this definition, we can derive the equation for the evolution of $\langle \alpha \rangle$ by taking the derivative with respect to t. We obtain

$$\frac{d}{dt} \langle \alpha \rangle = \int_{0}^{\infty} d\alpha \ \alpha \frac{\frac{d}{dt} C(\alpha, t)}{C(t)} - \frac{\frac{d}{dt} C(t)}{C(t)} \int_{0}^{\infty} d\alpha \ \alpha \frac{C(\alpha, t)}{C(t)} .$$
(3.14)

Defining the relative rate of change $\mu(\alpha, t)$ as

$$\mu(\alpha, t) := \frac{\frac{d}{dt}C(\alpha, t)}{C(\alpha, t)} , \qquad (3.15)$$

Eq. (3.14) reduces to

$$\frac{d}{dt} \langle \alpha \rangle = \langle \mu \alpha \rangle - \langle \alpha \rangle \langle \mu \rangle .$$
(3.16)

Using this equation the average number of primary particles can in principle be calculated for all times, if the relative growth rate $\mu(\alpha, t)$ is known. However, this quantity is exceedingly difficult to determine and to date no complete derivation of $\mu(\alpha, t)$ even for very simple cases has been found and only some approximations are known. We will see in the next part that one of the reasons for this difficulty is that $\mu(\alpha, t)$ depends on many properties of the whole system, such as the full probability distribution $p(\alpha, t)$.

We emphasize that this is one of the key advantages of our individual particle based approach, as it only requires knowledge of the properties of the individual particles and not of the whole system. In addition, our approach can be used for the numerical calculation of the relative growth rate and other global quantities if the individual particle properties are known.

3.4.2. Relative Growth Rates

The equation for the relative growth rate for coagulation $\mu_{\text{coag}}(\alpha, t)$ was developed by Smoluchowski (1917). For each value of α there is an increase in $C(\alpha, t)$ due to smaller particles coagulating so that their combined size is α and a decrease in $C(\alpha, t)$ due to particles of size α coagulating with any other particles. Formally, this can be written as

$$\mu_{\text{coag}}(\alpha, t) = \frac{1}{C(\alpha, t)} \cdot \left[\frac{1}{2} \int_{0}^{\alpha} d\alpha' \left(\chi(\alpha', \alpha - \alpha')C(\alpha', t) \cdot C(\alpha - \alpha', t)Q_{\text{coag}}(\alpha', \alpha - \alpha', t) \right) - \int_{0}^{\infty} d\alpha' \left(\chi(\alpha', \alpha)C(\alpha', t)C(\alpha, t)Q_{\text{coag}}(\alpha', \alpha) \right) \right], \qquad (3.17)$$

where $\chi(\alpha', \alpha)$ is a collision efficiency, i.e. the probability to coagulate upon collision and $Q_{\text{coag}}(\alpha', \alpha, t)$ is the collision kernel, i.e. the collision rate between particles of size α and α' at time t. Similarly, a growth rate due to fragmentation $\mu_{\text{frag}}(\alpha, t)$ can be developed, where $C(\alpha, t)$ increases due to larger particles breaking up so that the fragments are of size α and $C(\alpha, t)$ decreases due to particles of size α breaking up. This leads to

$$\mu_{\rm frag}(\alpha,t) = \frac{1}{C(\alpha,t)} \cdot \left[\int_{\alpha}^{\infty} d\alpha' \left(\vartheta(\alpha',\alpha)C(\alpha,t)Q_{\rm frag}(\alpha',t) \right) - C(\alpha,t)Q_{\rm frag}(\alpha,t) \right], \quad (3.18)$$

where $\vartheta(\alpha', \alpha)$ is the probability that a coagulate of size α' leads to a fragment of size α when it breaks. ϑ contains therefore the information about the number and size distributions of fragments, e.g. binary or ternary fragmentation. $Q_{\text{frag}}(\alpha, t)$ is the fragmentation kernel, i.e. the fragmentation rate for a particle consisting of α primary particles.

Both the coagulation and fragmentation kernels will in general not only depend on α and α' but also on system parameters, for example the coagulate strength or the turbulence level in the flow. The total relative growth rate is then given by $\mu(\alpha, t) = \mu_{\text{coag}}(\alpha, t) + \mu_{\text{frag}}(\alpha, t)$.

In recent years much effort has gone into finding approximations for the collision kernels. But in particular when particle inertia plays a role, effects such as preferential concentration and the occurrence of caustics lead to drastic modifications of the collision kernels that are still not fully understood (Bec et al., 2005).

The fragmentation kernel poses a very different problem. On the one hand it seems to be easier because it only involves individual coagulates. On the other hand it can be extremely complicated because the microscopic properties of the coagulates play a very important role and generally both the fragmentation kernel and the distribution of fragments are not well understood.

3.4.3. Estimating a Scaling Relationship

It is clear from the previous section that estimating the relative growth rate and with that the average coagulate size requires some information about collision and fragmentation kernels. We mention again that particularly for estimating these quantities and for comparing them with theoretical predictions of these quantities our individual particle based modeling approach is most useful.

The size distribution of the coagulates in the steady state (see Fig. 3.3) can be well approximated by an exponential. Therefore we take $C_N(\alpha, t) = \frac{C_N(t)}{\langle \alpha \rangle} e^{-\alpha/\langle \alpha \rangle}$.

As a first approximation for the coagulation, we assume that differences in radius between particles are small. Then the problem of the collision rates reduces to that of two particles with the same Stokes number, which is given by the mean Stokes number of the two particles. Since there is no interaction between particles through the fluid the collision kernel in our model is then approximated by (Bec et al., 2005),

$$Q_{\text{coag}}(\alpha, \alpha') = \hat{c} \epsilon^{1/2} (r_{\alpha} + r_{\alpha'})^{\theta(\langle St_{\eta} \rangle)} , \qquad (3.19)$$

where \hat{c} is a proportionality constant (see e.g. Saffman and Turner, 1956), ϵ is the average dissipation of turbulent kinetic energy in the flow and the exponent θ is a function of the mean Stokes number $\langle St_{\eta} \rangle$ of the two particles. r_{α} is the radius of a particle consisting of α primary particles, here this is given by $r_{\alpha} \propto \alpha^{1/3}$. No analytical expression is known for $\theta(\langle St_{\eta} \rangle)$, only two limit cases. For no inertia the exponent is 3 and for $\langle St_{\eta} \rangle \to \infty$ the exponent approaches 2. Numerical results e.g. by Bec et al. (2005) suggest that θ decreases monotonically for increasing $\langle St_{\eta} \rangle$, but no explicit equations are given.

To illustrate in principle the calculation of the average size we concentrate on the limit case of no inertia, where $\theta(\langle St_{\eta} \rangle) = 3$. This is the so-called rectilinear shear kernel (Thomas et al., 1999). In addition, we use a constant collision efficiency, i.e. $\chi(\alpha', \alpha) \equiv \chi_c$.

Using these approximations, the relative growth rate due to coagulation in Eq. (3.17) can be calculated. We obtain

$$\mu_{\text{coag}}(\alpha, t) = \hat{c}_1 \frac{\chi_c \epsilon^{1/2} C_N(t)}{\langle \alpha \rangle} \left[\frac{9 + 4\pi\sqrt{3}}{18} \alpha^2 - \langle \alpha \rangle^2 - 2\Gamma(\frac{2}{3}) \langle \alpha \rangle^{5/3} \alpha^{1/3} - \frac{2\pi\sqrt{3}}{3\Gamma(\frac{2}{3})} \langle \alpha \rangle^{4/5} \alpha^{2/3} - \langle \alpha \rangle \alpha \right].$$
(3.20)

For fragmentation, the first approximation for the probability $\vartheta(\alpha', \alpha)$ is that $\vartheta(\alpha', \alpha) = 2\delta(\alpha' - 2\alpha)$, where $\delta(x)$ is the Dirac delta function. This is the case of binary fragmentation, where both fragments are of the same size.

In our model all coagulates of the same size have the same critical shear $S_{f,c}(\alpha, \gamma)$. Here we studied the case $S_{f,c} = \gamma \alpha^{-\xi}$, where $\xi > 0$ and in particular the case of $\xi = 1/3$, see Eq. (3.7). If the fluid shear $S_f = (2S_{ij}S_{ij})^{1/2}$, where S_{ij} is the rate-of-strain tensor in the flow, at the position of the coagulates exceeds this critical shear $S_{f,c}$ they fragment. The probability for fragmentation of a given size is then only determined by the probability distribution of the shear $p(S_f, t)$, the influence of individual particle properties for the fragmentation kernel (see e.g. Ruiz and Izquierdo (1997)) is not considered. Again neglecting inertia effects and assuming a homogeneous distribution of the particles in the flow, the fragmentation kernel, i.e. the fragmentation rate is given by

$$Q_{\text{frag}}(\alpha, t) = \frac{\int_{S_{f,c}(\alpha,\gamma)}^{\infty} dS_f \ p(S_f, t) / \tau(S_f, t)}{\int_{0}^{S_{f,c}(\alpha,\gamma)} dS_f \ p(S_f, t)} , \qquad (3.21)$$

where $\tau(S_f, t)$ is the characteristic time of the shear S_f , see e.g. Bäbler et al. (2008). In our case, $Q_{\text{frag}}(\alpha, t)$ can not be determined analytically. Generally, $p(S_f, t)$ is a function of four (or nine, in three dimensions) random variables S_{ij} . Even in the simple case of independent normally distributed random variables which we have here, $p(S_f, t)$ can not be explicitly calculated.

For larger α Bäbler et al. (2008) argued that the fragmentation kernel is approximately given by a power law function. They estimated that the fragmentation kernel can be approximated by $Q_{\text{frag}}(\alpha, t) \approx \hat{d}\epsilon \frac{1}{S_{f,c}}$, with some constant \hat{d} . Similar power-law approximations for the fragmentation kernel have been found in other cases, see e.g. Ruiz and Izquierdo (1997). We will therefore continue our calculation using this expression.

We emphasize that the relationship between Q_{frag} and its arguments α and γ depends on the specific form of the stability condition for fragmentation, i.e. the specific model for $S_{f,c}$. It is through this dependence that the exponent of ξ of the stability condition Eq. (3.7) appears in the final result. For $S_{f,c} = \gamma \alpha^{-\xi}$ we obtain

$$\mu_{\rm frag}(\alpha, t) = \hat{d}\epsilon\gamma^{-1} \left[2e^{-\frac{\alpha}{\langle \alpha \rangle}} (2\alpha)^{\xi} - \alpha^{\xi} \right] \,. \tag{3.22}$$

These approximations for the relative growth rate can then be used in Eq. (3.16). To find the scaling behavior in the steady state we set $\frac{d}{dt} \langle \alpha \rangle = 0$, which leads to

$$\chi_c \epsilon^{1/2} C_{N,1} \langle \alpha \rangle \propto \epsilon \gamma^{-1} \langle \alpha \rangle^{\xi+1} , \qquad (3.23)$$

where $C_1 = C_N(t) \cdot \langle \alpha \rangle$ is the number concentration of primary particles in the system². The terms on the left side are the contribution from the coagulation Eq. (3.20) and the terms on the right hand side follow from the fragmentation Eq. (3.22). Solving Eq. (3.23) for $\langle \alpha \rangle$ leads to

$$\langle \alpha \rangle \propto \gamma^{1/\xi} \chi_c^{1/\xi} C_{N,1}^{1/\xi} \epsilon^{-\frac{1}{2\xi}} . \qquad (3.24)$$

We find an equation for the scaling of the average coagulate size in the steady state as a function of the particle and flow properties. While the scaling for γ and ϵ is similar to what was found in our numerical simulations using the individual particle based model (see Eq. (3.12) and Fig. 3.9), the scaling with χ_c and C_1 , where C_1 corresponds to N_1 in the individual particle model, is not entirely correct. This can therefore not be explained fully with the approximations made here. However we do find that the scaling exponents depend on $1/\xi$, as was found in the numerical simulations.

However, the calculation in this section illustrates that the average coagulate size can indeed be expected to scale with the particle and flow parameters and also makes it clear how the dependency of the scaling exponents on the exponent ξ of the stability condition Eq. (3.7) appears. Additionally, this calculation illustrates the special role of ϵ that affects both the coagulation and the fragmentation in the system. It is rather remarkable that even though the analytical calculation of the scaling is only possible in a very simplified case, a similar, simple scaling of the average coagulate size can be found numerically for the full individual particle based model.

3.5. Discussion

In the present study we described results from a coupled model for advection, coagulation and fragmentation of individual inertial coagulates. The model represents an approach to bridge the gap between the rate-equation based theory that is commonly used to describe larger coagulation and fragmentation systems and a full simulation of a multiphase flow. Full hydrodynamic simulations of coagulation and fragmentation are computationally limited to systems with very few particles and are therefore not appropriate to describe large-scale processes such as initiation of rain in a cloud. Rate-equation based models on the other hand are capable of describing coagulation and fragmentation on such scales, but rely on many approximations and parameterizations.

Our individual particle based approach was used to gain insights into the principle behavior of coagulates under different fragmentation mechanisms and to study the dependence of the steady state of the coagulates on particle and flow properties. We used

 $^{^2\}mathrm{This}$ follows from the assumption of an exponential size distribution.

synthetic turbulence in the form of a smooth random flow to approximate the motion of particles in a turbulent flow, focusing on processes which take place below the Kolmogorov scale. Even though not all features of turbulent flows are captured, the results are expected to remain qualitatively similar in more realistic flows. In realistic turbulent flows clustering and collisions between particles may depend on non-Gaussian statistics and intermittency in the velocity field, as well as the Reynolds number and could also be affected by clustering at an inertial range, where the velocity field is not smooth. However, as long as the system is well mixed, we do not expect a strong qualitative change. The same is true for the extension to three dimensional flows, where coagulation slows down, due to less frequent collisions, thereby mostly affecting the time scale of the approach to a steady state.

The applicability of the model used here to more realistic problems is limited due to the computational restriction of the number of primary particles. However, it is well suited for small systems and principal studies of underlying mechanisms. A great advantage is that an individual particle approach can easily incorporate experimental results and results from full hydrodynamic simulations to calculate average quantities such as collision or fragmentation rates which can then be incorporated into larger rate-equation based models. In particular, the fragmentation models described here only represent very simplified approximations of the real processes, for example the size-limiting fragmentation approach captures only the appearance of two or three large daughter droplets and not the full host of small satelite droplets that is created during breakup. However, this can easily be expanded within our framework with more detailed expressions if that is required for a specific problem. The fragmentation model presented here therefore serves as the simplest possible approximation to illustrate what is possible with such an individual particle based approach. Already, with these simplest approximations we can capture many essential features of coagulation and fragmentation processes.

We numerically studied the steady state that results from a balance between coagulation and fragmentation. Mainly, we examined average quantities that characterize the steady state, such as the average number of primary particles per coagulate. We compared two different fragmentation mechanisms, size-limiting fragmentation which is motivated by the hydrodynamical instability of large drops settling under gravity and shear fragmentation, where particles break due to hydrodynamic shear forces. For both size-limiting and shear fragmentation the transient behavior of the system is strongly influenced by the particle and flow properties. In particular, enhanced collision rates, for example due to increased shear or increased collision efficiency greatly decrease the time it takes to reach the steady state.

For size-limiting fragmentation this steady state shows few fluctuations and almost no

dependence on the particle or flow parameters. The main parameter that determines the coagulate size distribution in this case is the maximum stable coagulate size. The size distribution in this case is very broad, and covers almost all the available coagulate sizes. Different size distributions can appear if the number of fragments is chosen differently, for example if fragmentation is binary. The shape of the size distribution is then related to the number of fragments that are created during fragmentation.

The size distributions for shear fragmentation have a single peak with an exponential tail. This is a typical feature of a fragmentation mechanism where coagulates break into similar sized fragments (compare Ch. 4 of this work). For shear fragmentation strong fluctuations in the average number of primary particles per coagulate due to statistical fluctuations of the carrier flow appear. In this case, both the average number of primary particles per coagulate and the standard deviation of the coagulate size distribution in the steady state change strongly with the particle and flow properties. Simulations showed that the variation of each parameter within a reasonable range is approximately independent of the values of the other parameters. For variations of the coagulate strength γ the scaling relationships for both the average and the standard deviation can be inferred from the fragmentation mechanism. Scaling relationships for variations of the volume fraction, the collision efficiency and the dissipation of turbulent energy in the fluid were derived from the simulation results. For each of these parameters we find a power-law dependence, where the exponents appear to be closely connected to the shape of the stability condition for fragmentation. We illustrated this by showing how an equation for the average coagulate size in the steady state can be derived. From this we calculated scaling relationships for the average coagulate size in the steady state using severe approximations. This calculation also clarified how our individual particle based approach can be connected with the rate-equation based theory that is commonly used to describe larger coagulation and fragmentation systems. However, this approach requires expressions for the collision and fragmentation rates as well as some knowledge of the coagulate size distribution. By contrast, our individual particle based model only requires knowledge if the individual particle properties, which turns out to be a great advantage of our approach. It can therefore be a very useful tool, both for obtaining estimates of global quantities such as collision and fragmentation rates and as a comparison for results from rate-equation based models.

Our results emphasize the great importance of the fragmentation mechanism for the final size distribution of coagulates in the steady state. As a consequence it is very desirable to design experiments to investigate the fragmentation of particles in different applications.

In general, the dependence of the average quantities as well as the size distributions

on the particle and flow properties can change quantitatively for different fragmentation mechanisms, in particular for different number and size distributions of fragments created during fragmentation. However, the qualitative picture that has emerged can be expected to remain the same.

4. Lagrangian Modeling of Fractal-Like Aggregates

This chapter contains, with some minor changes, our work on individual-particle based modeling of aggregation and fragmentation of solid particles with a fractal-like structure in a synthetic turbulent flow which has been submitted for publication to Physica D as J.C. Zahnow, J. Maerz and U. Feudel, "Particle-based modelling of aggregation and fragmentation processes: Fractal-like aggregates" (2010).

4.1. Introduction

In most systems where the particle phase consists of solid particles, as opposed to bubbles or droplets, aggregation and fragmentation of the particles leads to clusters with a complex structure. This is for example the case for marine aggregates (Thomas et al., 1999), cohesive sediment (Kranenburg, 1994), colloid suspensions (Spicer and Pratsinis, 1996a) or solid-liquid separation systems (Bäbler et al., 2008). This complex structure of particles can have a great influence on particle dynamics as well as aggregation and fragmentation processes. Both the actual motion of aggregates and the probabilities for aggregation and fragmentation are influenced by the structure of the particles.

In the context of a rate-equation based approach, a complex particle structure has been incorporated in the past in terms of a density modification for the particles, e.g. by Kranenburg (1994) or Maggi et al. (2007). However, so far there are very few attempts to treat this problem for inertial particles in a flow.

Therefore, in this Chapter the individual particle based model for aggregation and fragmentation presented in Part 3 of this work, where each aggregate is modeled as an individual finite-size particle whose motion is tracked in a Lagrangian framework. Here, the consideration of spherical particles that was used in the previous Chapter is expanded to model more realistic aggregates. While this was a reasonable approximation for coagulating droplets, the aggregation of solid particles is generally more complex. We focus specifically on the problem of aggregation and fragmentation in systems where the aggregates can be described as having a *fractal-like structure*, as is for example the case for marine aggregates (see e.g. Logan, 1999). By this we mean that on average there exists a power-law relationship between the characteristic length and the mass of such aggregates. The exponent of the power-law is called the fractal dimension. We show in this Chapter how such a complex structure can be approximated within the framework of our individual particle-based approach. Such a characterization in terms of a fractal dimension leads to a modification of the radii and effective densities of the aggregates compared to a solid sphere of the same mass. Nevertheless, we still treat them as effectively spherical for the particle motion, allowing us to apply the Maxey-Riley equations of motion (Maxey and Riley, 1983) with modified parameters.

In this work we choose a parameterization of our model for the case of a suspension of marine aggregates in the ocean. In this way we can study our modeling approach for a specific case, but we emphasize that our model is a general one that can in principle be used for a wide range of applications where aggregation and fragmentation of solid particles plays a role. The concept of a fractal-like structure has been found to be a reasonable first approximation in many different applications, ranging from colloidal systems to the flocculation of cells Logan (1999).

Using this Lagrangian framework the long-term behavior of particle size distributions that develop from a balance between aggregation and fragmentation is studied. In particular, the influence of fragmentation and aggregate structure on these size distributions is examined. Since the fractal dimension of marine aggregates can vary greatly in natural systems (Maggi et al., 2007), its effect on the steady state particle size distributions in this model is examined. It is found that while the shape of the size distributions does not depend strongly on the fractal dimension, the average particle size and relaxation time towards the steady state depend strongly on this parameter.

We show that the combination of aggregation and fragmentation of fractal-like aggregates, superimposed on inertial advection dynamics, leads to a convergence to a steady state in the particle size distribution. This steady state is unique for a given set of parameters. Mainly, we compare three different types of splitting, uniform fragmentation, erosion and large-scale fragmentation. These splitting modes differ in the size of the fragments that are created during break-up. While erosion leads to one large and one relatively small fragment, large-scale fragmentation leads to two fragments of similar size. We find that the transient dynamics as well as the size distribution in the steady state depend strongly on the splitting mode. The steady state size distribution found for large-scale fragmentation conforms best to observation reported in the literature for the break-up of marine aggregates in tidal areas (Lunau et al., 2006), indicating that this may be the primary mode of fragmentation in these cases.

Section 4.2 describes the complete model for advection, aggregation and fragmentation that is used in this work. The equations of motion for heavy spherical particles (Stokes equation) are used, but with modified parameters to take a fractal-like structure into account. Rules governing the aggregation and fragmentation are introduced. Finally, the model is applied to a simple 3-d synthetic turbulent flow field.

Section 4.3 then presents a complete analysis of the influence of all major system parameters on the resulting steady state size distributions, the average aggregate size in steady state and the relaxation time towards the steady state. Namely, these parameters are aggregate strength, fractal dimension of the aggregates and total particle mass in the system.

Section 4.4 contains a discussion of the limitations of the model and the conclusions.

4.2. Modeling Approach

In this section we will present the modeling approach used in this study, that describes the motion, aggregation and fragmentation of finite-size particles. Firstly, the equations of motion used for the advection of spherical particles heavier than the surrounding fluid are presented. Secondly, a model to account for the fractal-like structure of real aggregates is described. Thirdly, a full model to include aggregation and fragmentation in this context is introduced. Finally, a simple 3-d synthetic turbulent flow field is chosen, that will be used to study the aggregation and fragmentation model in detail.

4.2.1. Lagrangian Modeling for Fractal-Like Aggregates

For simplicity, we consider all primary (smallest, unbreakable) particles to be spherical and denser than the ambient fluid. We emphasize that the equations of motion presented here for spherical particles will in the following also be used to describe the motion of aggregates which usually can not be assumed to be spherical (Kranenburg, 1994). However, to account for properties related to the fractal-like structure of aggregates some modifications to the equations of motion (in form of modified parameters) will be introduced in the next section. While this represents only a very simplified model and the surface forces acting on particles with a complex structure are an extremely complex problem where to date no satisfying expressions exists, we believe that this is a reasonable starting point. On the one hand, if one wants to employ the model discussed here to a specific case where better expressions are known, this can easily be adapted without changing the general idea of our approach. On the other hand, it has been found in many cases (see for example Zahnow et al. (2008, 2009)) that changes in the motion of the individual particles usually do not lead to significant changes in the dynamics of the particle ensemble and in particular in the collision rates which are relevant for the overall size distribution in an aggregation-fragmentation system.

Finite-size particles usually do not follow exactly the motion of the surrounding fluid,

instead inertia effects lead to deviations of the particle motion from that of the fluid. For small Reynolds numbers the equations of motion for spherical particles of finite size are the Maxey-Riley equations (Maxey and Riley, 1983). This implies that locally the flow around the particle is laminar, even though the overall fluid flow can still be turbulent. While inertia effects can be fairly small for the primary particles in the case of marine aggregates (see Sec. 4.2.4), the influence of particle inertia increases with aggregate size and can become quite important for larger marine aggregates.

In reality every particle produces perturbations in the flow that decay inversely proportional to the distance from the particle (Happel and Brenner, 1983). In this work we keep the particle concentration C_V low enough to be in a diluted regime. For particles of radius r_p and density ρ_p moving coherently within the dissipative scale \check{L}_p of a flow the feedback from the particle motion on the flow can be neglected if $C_V r_p \check{L}_p^2 \ll 1$ (Balkovsky et al., 2001). Particle-particle interactions mediated by flow perturbations are neglected, see the discussion in Sec. 4.4.1.

Assuming that the difference between the particle velocity $\mathbf{V}(t)$ and the fluid velocity $\mathbf{u} = \mathbf{u}(\mathbf{X}(t), t)$ at the position $\mathbf{X}(t) = (X_1(t), X_2(t), X_3(t))$ of the particle is sufficiently small, the drag force is proportional to this difference. This is called Stokes drag. With these restrictions the dimensionless form of the governing equation for the path $\mathbf{X}(t)$ of such a particle under the influence of drag and gravity can then be approximated from the Maxey-Riley equations as:

$$\dot{\mathbf{V}} = \frac{1}{St} \left(\mathbf{u} - \mathbf{V} - W\mathbf{n} \right), \tag{4.1}$$

where **n** is the unit vector pointing upwards in the vertical direction (which is the X_2 -axis in this study).

The particle Stokes number St, i.e. the ratio between particle response time and flow time scale is defined as

$$St = (\rho_p 2r_p^2) / (9\mu_f \check{T}_f)$$
(4.2)

and the dimensionless settling velocity in a medium at rest W is defined as

$$W = 2r_p^2(\rho_p - \rho_f)\check{T}_f g / (9\mu_f \check{L}_f) .$$
(4.3)

Here, ρ_f and μ_f are the fluids' density and dynamic viscosity and \check{L}_f and \check{T}_f are characteristic length and time scales of the flow.

Fractal-Like Aggregates

When looking at real aggregates they are typically not solid spherical particles but instead can have a complex structure as they consist of a number of primary particles. In this model the primary particles are assumed to be solid spherical particles, following the equations of motion as described in the last subsection. All aggregates are assumed to consist of an integer number of such spherical primary particles. The description of the motion of an aggregate with a complex structure is still an unsolved problem. Therefore we only consider the influence of the structure of the aggregates on their size and effective density. Here, we assume that aggregates have a fractal-like structure, meaning that there exists a power-law relationship between the characteristic length and the mass of such aggregates. The structure of the aggregates can then be characterized by a fractal dimension $f_d < 3$ (Mandelbrot, 1983). Their size can still be defined approximately by a radius, that can be considered as the characteristic length scale of the aggregate. This radius r_{α} of an aggregate that consists of α primary particles and has a given fractal dimension f_d is derived in the following. We emphasize that the number of primary particles α in an aggregate is here also used as an index to describe a quantity, e.g. the radius or the volume, that corresponds to an aggregate consisting of α primary particles. The solid volume, i.e. the volume of an aggregate that is filled with particulate matter follows from the definition of the fractal dimension f_d (see e.g. Logan, 1999) as

$$V_{\alpha,\text{solid}} = c_{f_d} r_{\alpha}^{f_d} , \qquad (4.4)$$

where c_{f_d} is a proportionality constant that can depend on f_d . As mass conservation must be fulfilled we get

$$V_{\alpha,\text{solid}} = \alpha V_1 = \alpha \frac{4}{3} \pi r_1^3 , \qquad (4.5)$$

where r_1 and V_1 are the radius and volume of a primary particle, respectively. The proportionality constant c_{f_d} can be derived from Eqs. (4.4) and (4.5) by setting $\alpha = 1^{-1}$

$$c_{f_d} = \frac{4}{3}\pi r_1^{3-f_d} \ . \tag{4.6}$$

In combination with Eqs. (4.4) and (4.5) this leads to

$$r_{\alpha} = \alpha^{1/f_d} r_1 \tag{4.7}$$

¹Actually, one would have to relate the radius of a fractal-like aggregate to the fractal generator, i.e. the smallest structure with the same fractal properties instead of to the (spherical) primary particles. This would introduce additional shape factors and constants that are not present in the current form, but not change the qualitative results. For details on this, see e.g. Logan (1999)

for the radius of an aggregate. It is evident that due to the fractal-like structure the radius r_{α} is greater than for a completely solid particle of the same mass.

A part of a fractal-like aggregate, i.e. of the total volume encased by an aggregate

$$V_{\alpha,\text{total}} = \frac{4}{3}\pi r_{\alpha}^3 , \qquad (4.8)$$

is not filled with matter but with the surrounding fluid. The aggregate density therefore decreases with an increasing number α of primary particles in the aggregate. From mass conservation it follows

$$\rho_{\alpha} V_{\alpha,\text{total}} = \rho_1 V_{\alpha,\text{solid}} + \rho_f (V_{\alpha,\text{total}} - V_{\alpha,\text{solid}})$$
(4.9)

Solving Eq. (4.9) for ρ_{α} and substituting $V_{\alpha,\text{total}}$ and $V_{\alpha,\text{solid}}$ leads to

$$\rho_{\alpha} = \rho_f + (\rho_1 - \rho_f) \alpha^{1 - 3/f_d} . \tag{4.10}$$

Going back to the equations of motion we now see that as far as the particle dynamics are concerned, a first approximation for the fractal-like aggregates is to treat them as spheres with an increased radius r_{α} compared to solid spheres of the same mass, but a reduced density ρ_{α} , because of the fluid encased in their spherical volume.

This leads to a modification of the particle Stokes number St and dimensionless settling velocity W for fractal-like aggregates in the equations of motion (4.1), when compared to a solid sphere:

$$St_{\alpha} = (\rho_{\alpha} 2r_{\alpha}^2)/(9\mu_f \check{T}_f) \tag{4.11}$$

$$W_{\alpha} = 2r_{\alpha}^2(\rho_{\alpha} - \rho_f)\check{T}_f g/(9\mu_f \check{L}_f) . \qquad (4.12)$$

For fractal-like aggregates these parameters replace St and W in Eq. (4.1), leading to a motion with different parameters for aggregates with different numbers α of primary particles.

Aggregation Model

The physical, chemical or biological process that leads to aggregation of particles can vary from case to case and is not examined in detail here, as this is beyond the scope of this study. Instead, a general model is used. The only assumption is that during a collision particles can somehow stick together and form an aggregate. No detailed mechanism of sticking is considered. Whenever the distance between two particle centers becomes smaller than the sum of their radii, these two particles aggregate immediately, creating a new particle that replaces the two old particles. For two particles of radius a_{α_i} and a_{α_j} the new number of primary particles after aggregation is obviously $\alpha_{\text{new}} = \alpha_i + \alpha_j$, leading to a new radius $a_{\alpha_{\text{new}}}$, which can be derived from Eq. (4.7). The position of the new particle is the center of gravity of the old particles. The velocity of the new particle follows from momentum conservation.

4.2.2. Fragmentation Model

The physical process leading to fragmentation can vary as well. While there are detailed models for the fragmentation of e.g. water droplets (Villermaux, 2007), the mechanism of fragmentation of marine aggregates is not well understood. Even experiments give no clear indication how fragmentation of such aggregates occurs in detail. Therefore only a few theoretical approaches exist for this process (Pandya and Spielman, 1982; Hill and Ng, 1996). In this work a model for fragmentation is used that is only based on some very general properties of the aggregates involved.

In the following the fragmentation is described in two parts, that need to be clearly distinguished. Firstly, a *splitting condition*, that determines *if* a fragmentation event takes place and secondly, a *splitting rule*, that determines *how* fragmentation takes place, are defined.

Splitting Condition

The splitting condition describes if fragmentation of an aggregate takes place. Generally, only particles that are composed of more than one primary particle can fragment. The break-up of an aggregate occurs when the hydrodynamical forces $F_{\rm hyd}$ acting on the aggregate exceed the forces $F_{\rm agg}$ holding the particles in the aggregate together. The criterion for breakup can therefore be expressed as

$$F_{\rm hyd}/F_{\rm agg} > const.$$
 (4.13)

For aggregates with a fractal-like structure, consisting of a number of solid spheres, the hydrodynamical forces in the dissipative range where viscous forces dominate is proportional to the shear force integrated over the surface of the aggregate (Kobayashi et al., 1999). For a fracture at a distance $\zeta \cdot r_{\alpha}$, $\zeta \in [0, 1]$ from the equator of the aggregate this results in

$$F_{\rm hyd} \propto S_f (1-\zeta) r_{\alpha}^2$$
, (4.14)

where S_f is the shear rate in the flow.

For a porous aggregate Ruiz and Izquierdo (1997) give the force F_{agg} holding an aggregate together as proportional to the area of constituent matter in a cross-section of the aggregate. Ruiz and Izquierdo then related $V_{\alpha,\text{solid}}$ to the porosity of an aggregate. Here, we instead rewrite this relationship in terms of the fractal dimension f_d . For fractures across the equator of an aggregate the area is proportional to $V_{\alpha,\text{solid}}^{2/3} \propto r_1^2 \left(\frac{r_\alpha}{r_1}\right)^{2f_d/3}$. For a fracture at a distance $\zeta \cdot r_\alpha$, $\zeta \in [0, 1]$ the area of constituent matter in the cross-section is reduced. It equals the area of a cross-section across the equator of an aggregate with decreased radius $(1 - \zeta^2)^{1/2} r_\alpha$. In general we get

$$F_{\text{agg}} \propto (1 - \zeta^2)^{f_d/3} r_\alpha^2 \left(r_\alpha / r_1 \right)^{2f_d/3 - 2} . \tag{4.15}$$

The splitting condition (4.13) then becomes

$$S_f \cdot \frac{(1-\zeta)}{(1-\zeta^2)^{f_d/3}} \cdot \left(\frac{r_{\alpha}}{r_1}\right)^{2-2f_d/3} > \gamma,$$
 (4.16)

where the proportionality constant γ is determined by the force holding the primary particles in an aggregate together. It is therefore a measure of the aggregate strength. Solving for the shear rate S and using Eq. (4.7) leads to an expression for the critical shear $S_{f,c}$ required to break up an aggregate

$$S_{f,c} = \gamma \frac{(1-\zeta^2)^{f_d/3}}{(1-\zeta)} (r_\alpha/r_1)^{-2+2f_d/3}$$

= $\gamma \frac{(1-\zeta^2)^{f_d/3}}{(1-\zeta)} \alpha^{2/3-2/f_d}$. (4.17)

It can be seen that for a fractal dimension $f_d < 3$ the critical shear force required to break up an aggregate decreases with the aggregate size, i.e. larger aggregates are less stable than smaller ones. Additionally, the critical shear required for fragmentation is smallest for fractures across the equator of an aggregate ($\zeta = 0$) and increases with increasing distance from the equator.

Within our model, the local shear force S_f can be calculated at any point in space and time as (1/2)

$$S_f = \left(2\sum_{i,j} S_{ij} S_{ij}\right)^{(1/2)} , \qquad (4.18)$$

where $S_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial X_j} + \frac{\partial u_j}{\partial X_i} \right)$ is the rate-of-strain tensor in the flow.

Splitting Rules

The splitting rules describe how an aggregate will split, when the splitting condition is met. During fragmentation only particles whose mass is an integer multiple of the mass of a primary particle are created. This means that, even though they have become part of some larger aggregates, primary particles can never be broken up. Only the connection among each other can break. Different size distributions of the fragments are possible.

When a splitting condition is met, an aggregate consisting of α_{old} primary particles is split into 2 fragments with the number α_k of primary particles of each fragment being a random fraction of the original number α_{old} . Typically, one distinguishes between two mechanisms for fragmentation (Jarvis et al., 2005). For each mechanism fragmentation occurs on average at a different distance from the equator of the aggregate, leading to different distributions of the fragments α_k . This can be expressed in different values for ζ , the fraction of the distance from the equator of the aggregate where fragmentation is assumed to take place (see Sec. 4.2.2). Large-scale fragmentation happens when an aggregate is 'pulled apart' somewhere close to the equator, leading to fragments of similar size. This





is characterized by $\zeta = 0$. *Erosion* happens when shear forces act closer to the edge of an aggregate (Vassileva et al., 2007), implying that $0 < \zeta < 1$. In this case only few primary particles are split off from the aggregate (see Fig. 4.1).

Here we will compare three different fragmentation modes, with different distributions for the number of primary particles in the fragments. First a large-scale splitting rule, second an erosion splitting rule, and third a *uniform splitting* rule. In most realistic cases one expects different fragmentation modes to appear together, even though with slightly different probabilities. However, here we apply these fragmentation modes separately to determine their individual influence on the aggregate size distribution.

1. For both large-scale fragmentation and erosion we assume that there is a preferred distance ζr_{α} , $\zeta \in [0, 1]$ from the center of the original aggregate where fragmentation occurs. The two mechanisms are then characterized by different values of ζ . A fracture at a distance ζr_{α} leads to a fragment containing a fraction $V_{\rm fr}/V_{\alpha,\text{total}}$ of the original volume, where $V_{\rm fr} = \pi r_{\alpha}^3 (\frac{2}{3} - \zeta + \frac{\zeta^3}{3})$. Since ζ is assumed to be only the average distance of a fracture from the equator of the aggregate, for each fragmentation event we choose the number of primary particles in the fragment randomly from a Gaussian distribution, centered around $\alpha_{\rm old} \cdot V_{\rm fr}/V_{\alpha,\text{total}}$. This allows for a certain variation of the fragment size, meaning that

$$\alpha_1 = \left(\frac{4}{3}\left(\frac{2}{3} - \zeta + \frac{\zeta^3}{3}\right) + \xi\right)\alpha_{\text{old}} , \qquad (4.19)$$

where ξ is a random number from a normal distribution with mean 0 and standard deviation σ_{frag} and the right-hand side of Eq. (4.19) is rounded towards the nearest integer. We note that our results do not depend strongly on the choice of σ_{frag} , here we choose $\sigma_{\text{frag}} = 0.2$ which results in a typical variation of one primary particle between fragments coming from identical aggregates. As an additional restriction it is required that $1 \leq \alpha_1 < \alpha_{\text{old}}$, otherwise a new random number ξ is chosen.

For large-scale fragmentation, aggregates are assumed to break along the equator into two fragments of similar size, which corresponds to $\zeta = 0$. For erosion, fragmentation is assumed to occur at a distance from the center, leading to one aggregate being much smaller than the other. This corresponds to $0 < \zeta < 1$. Here, we choose $\zeta = 0.6$ for erosion, which leads to smaller fragments containing on average 10 percent of the mass of the original aggregate. Similar results for erosion were found for other values of ζ . However, if ζ becomes too large particles will no longer fragment because the critical shear required to break off a fragment increases greatly as $\zeta \to 1$.

2. In the uniform splitting rule the number of primary particles for the first fragment is chosen from a uniform distribution in the interval $I = [1, \alpha_{old}]$. The uniform splitting rule is a simplified model for the full case where both large-scale fragmentation and erosion of an aggregate can happen. However, all sizes of fragments occur here with the same probability. For all three cases the remaining aggregate consists of $\alpha_2 = \alpha_{old} - \alpha_1$ primary particles. Whenever a particle is split according to one of the splitting rules, all parts keep the velocity of the original particle. That way momentum is conserved. The first fragment remains at the position $\mathbf{X} = (X_1, X_2, X_3)$ of the original particle. The center of the other fragment is placed along a line segment in a random direction, so that for the two fragments the distance equals the sum of the radii.

For each fragment, the splitting condition is checked again and if it is met, the whole process is repeated until no fragment fulfills the fragmentation condition. This leads to a splitting cascade and aggregates can break up into more than two fragments, if the aggregate is large enough or shear forces are strong enough. This is consistent with experimental observations of marine aggregates that larger particles tend to break into more fragments (Alldredge et al., 1990). In that way ternary, quarternary and other splitting types besides binary splitting naturally appear in this model.

Here, large-scale fragmentation and erosion are treated as two separate processes to study the influence of fragmentation at certain distances ζ on the aggregate size distributions in the steady state. In reality, aggregates will break with certain probabilities at certain distances ζ from the center but there will be no two separate processes. Therefore, depending on the probability distribution for fragmentation at a certain distance one can expect different combinations of the steady state size distributions found in this work. The uniform fragmentation rule represents one such possible combination, where fragmentation at all distances ζ appears with the same probability.

4.2.3. Fluid Flow

As a fluid velocity field we consider synthetic turbulence in the form of a space-periodic, isotropic and homogeneous Gaussian random flow (Bec, 2005), since it allows us to perform long-term simulations at reasonable computational costs. We use a smooth, incompressible flow since we focus on effects typically taking place on scales smaller than the Kolmogorov scale of a turbulent flow.

The flow is written as a Fourier series

$$\boldsymbol{u}(\boldsymbol{X},t) = \sum_{\boldsymbol{k} \in \mathbb{Z}^d \setminus \{\boldsymbol{0}\}} \hat{\boldsymbol{u}}(\boldsymbol{k},t) e^{i\frac{2\pi}{L}\boldsymbol{k} \cdot \boldsymbol{x}} , \qquad (4.20)$$

where $\hat{\boldsymbol{u}}(\boldsymbol{k},t) \in \mathbb{C}^d$ are the Fourier components, with the property $\hat{\boldsymbol{u}}(-\boldsymbol{k},t) = \hat{\boldsymbol{u}}^*(\boldsymbol{k},t)$ because $\boldsymbol{u}(\boldsymbol{X},t)$ is real-valued. The star denotes complex conjugation. By taking for $\hat{\boldsymbol{u}}(\boldsymbol{k},t)$ the projection of a different vector $\hat{\boldsymbol{v}}(\boldsymbol{k},t) \in \mathbb{C}^d$ onto the plane perpendicular to the wave vector \boldsymbol{k} , incompressibility is ensured. The vector $\hat{\boldsymbol{v}}(\boldsymbol{k},t)$ is assumed to be an Ornstein-Uhlenbeck process. It is a solution of the complex-valued stochastic differential equation

$$d\hat{\boldsymbol{v}} = -\xi(\boldsymbol{k})\hat{\boldsymbol{v}}dt + \sigma(\boldsymbol{k})\boldsymbol{dW} , \qquad (4.21)$$

with $\xi(\mathbf{k}), \sigma(\mathbf{k}) \in \mathbb{R}$, where dW is a *d* dimensional complex Wiener increment. The parameters $\xi(\mathbf{k}), \sigma(\mathbf{k})$ need to be chosen in such a way that the flow $u(\mathbf{x}, t)$ reproduces some features of a real turbulent flow, in this case we choose the energy spectrum in the dissipative range of a turbulent flow. Here we use the exponential spectrum suggested by Kraichnan

$$E(k) = C \cdot (2\pi k \check{L}_f / \hat{L})^3 \exp(-\beta [2\pi k \check{L}_f / \hat{L}]) , \qquad (4.22)$$

with $\beta = 5.2$ (Martinez et al., 1997) and a suitably chosen normalization constant C. The constant \check{L}_f is the length scale of coherent structures in the flow and \hat{L} is the spatial period of the flow. We choose $\xi(k) = 1/\check{T}_f$ and $\sigma(k) = \sqrt{E(k)/\check{T}_f}$. The constant \check{T}_f is then the correlation time of the flow. The normalization constant is chosen so that a desired value of the mean shear rate $\langle S \rangle$ is obtained. The flow is then characterized by the correlation time \check{T}_f , the correlation length \check{L}_f and the mean shear rate $\langle S \rangle$.

If a fluid velocity field with few Fourier modes is chosen, no interpolation of the velocity at particle position is required, since it can be calculated from direct summation of the Fourier series. This allows for a resolution of the fine structures of the particle distribution in space.

4.2.4. Implementation

Next, we will describe some specifics of the numerical implementation and the system parameters used in this work.

For the simulations in this work we choose particle properties similar to those of marine aggregates in coastal waters. The primary particles considered in this model have a radius $r_1 = 4\mu$ m, density $\rho_1 = 2.5 \times 10^3$ kg/m³ and mass $m_1 = \rho_1 \frac{4}{3}\pi r_1^3$. The relevant characteristic length scales for marine aggregates in coastal areas of the ocean are typically the Kolmogorov scales. Shear rates in coastal areas can be of order $1s^{-1}$, leading to Kolmogorov length and time scales of $\check{L}_f = 1$ mm and $\check{T}_f = 1$ s, respectively (Kranenburg, 1994). Using these scales to make the equations of motion of the particles dimensionless leads to a Stokes parameter of $St_1 = 10^{-5}$ and a dimensionless settling velocity of $W_1 =$ 0.1 for the primary particles.

The aggregate strength parameter γ is fixed at $\gamma = 8$, unless otherwise mentioned.

The number of aggregates N(t) changes over time due to aggregation and fragmentation leading to a distribution of aggregates of different radii in the flow. However, the total mass $M = \sum_{i=1}^{N(t)} \alpha_i m_1$ remains constant during one simulation. As initial condition we take $N_1 = 10^5$ primary particles and no larger particles. Furthermore particles are uniformly distributed over one periodic cell of size \hat{L}^3 of the configuration space, with velocities matching that of the fluid. This choice fixes the total mass of the system to be $M = 10^5 m_1$. For the flow we choose a periodic cube with $\hat{L} = 4\check{L}_f$, so that we obtain a volume fraction of about 0.4×10^{-3} .

The fractal dimension f_d of marine aggregates varies between approximately 1.5 for very open, fragile aggregates like marine snow in the open ocean and approximately 2.5 for stronger, compact aggregates. The average is typically around 1.9 - 2.0 (see e.g. Winterwerp, 1998), therefore in the following we choose $f_d = 2.0$ unless otherwise mentioned.

As a first approximation the aggregation and fragmentation processes are assumed to have no effect other than to change the size of the particles and the effective density, and therefore do not directly influence the motion of the particles. Hence all three aspects, motion, aggregation and fragmentation that define the whole system can be modeled separately. Aggregation is checked constantly during the integration, whereas fragmentation is applied after every time step of the system.

1. All particles move in the flow for one time step dt using the equations of motion described in Sec. 4.2.1. We emphasize at this point that each aggregate size is characterized by different values of St_{α} and W_{α} , so that the motion of aggregates of different size is governed by the same equations but with different parameters.

The length of the time step dt needs to be chosen small enough so that the simulation result becomes independent of this values, here we found dt = 0.01s to be sufficiently small.

Because of the spatial periodicity of the flow, all particle dynamics will be folded back onto one \hat{L}^3 cell in the flow, using periodic boundary conditions. Usually particles do not stay in one cell of the flow, i.e. they are not suspended in the flow. Instead particles will generally fall downwards through the flow, if they are heavier than the fluid (Maxey and Corrsin, 1986). This means that folding the dynamics of the particles back onto a single cell is only a convenient way to visualize an infinitely extended system and does not completely mirror what one would see in a comparable experiment. However, if particles are initially distributed homogeneously over the whole configuration space, the total particle mass in each periodic cell remains the same over time. Therefore even if aggregation and fragmentation are included, it is sufficient to restrict our studies to one unit cell with periodic boundaries.

- 2. Particles aggregate upon collision, i.e. if their distance becomes equal to the sum of their radii. To ensure that no collisions are missed, we use an efficient event-driven algorithm for particle laden flows (cf. Sigurgeirsson et al. (2001) for details). Computationally, the aggregation process is the most costly component of the simulation. In particular, the naive approach to check which particles are colliding involves looping over all pairs of particles and therefore scales as $O(N^2)$, where N is the number of particles. Therefore, here a linked-list algorithm (Hockney and Eastwood, 1981), sometimes also called link-cell algorithm is used to compute the distance between particles. The configuration space is divided into grid cells of size ϵ , where each grid cell stores information on which particles it contains. The looping over particle pairs to calculate their distance is then done only over particles in a given grid cell and the neighboring cells. If the grid cell size ϵ is small enough (but larger than the largest appearing particle size) the link-cell algorithm scales as O(N) and is thus much faster than the naive approach.
- 3. After each time step dt particles can fragment if the shear at their position exceeds a critical value. If that is the case, new fragments are created according to the rules described in Sec. 4.2.2.

We note that at first glance it looks like aggregation and fragmentation are treated very different, in particular aggregation seems to be independent of the aggregate strength γ in this model. However, this is not the case. Initially all particles that come into contact aggregate, i.e. here the aggregation probability upon collision is equal to one. But when looking at aggregation and fragmentation together over one time step dt it is in fact smaller than one because some aggregates that just formed during this time step will break up again. These are the aggregates where the aggregate strength γ is not strong enough to hold the aggregate together. This means after one time step only some of the particles that came into contact will actually have aggregated and this number will depend on the aggregate strength γ . This means that both aggregation and fragmentation probabilities depend on the same aggregate property, which one would expect in reality.

4.3. Simulation Results

In the following section we will present simulation results using the model described above, to determine the influence of the different splitting rules on the resulting particle size distributions. As the parameters used in the model system can vary greatly in natural
systems, we examine the sensitivity of the system with regard to the following parameters: aggregate strength γ , fractal dimension f_d and total number of primary particle N.

4.3.1. Measured Quantities

From previous works it is known that the balance of aggregation and fragmentation typically leads to a steady state of the particle size distribution (Zahnow and Feudel, 2009). This follows from the fact that normally aggregation dominates for small sizes, whereas fragmentation is the dominant process for large sizes. In addition to studying this size distribution of the particles in the steady state, we introduce different measures to characterize first the approach to the steady state and then the steady state itself. To follow the convergence of the system towards a steady state, we use two different quantities. The first quantity that we measure during the simulations is the average number of primary particles per aggregate, defined as

$$\langle \alpha(t) \rangle = \sum_{\alpha} \alpha N_{\alpha}(t) / N(t) .$$
 (4.23)

In the context of our model $2 \langle \alpha(t) \rangle$ corresponds to the 'mean equivalent circular diameter' that is often used as a measure in experiments with marine aggregates (Lunau et al., 2006). We will use this quantity as a first estimate whether the particle size distribution has converged to a steady state and to follow the evolution of the particle size distribution towards the steady state.

A second quantity of the aggregation and fragmentation process that may be experimentally measured is the time it takes to reach the steady state. Especially in technical applications this can be an important quantity, where processes need to be timed appropriately to allow for a smooth work flow. Here we introduce a measure for this relaxation time in our model and show how different system parameters influence this time to reach the steady state.

We define the relaxation time τ_{∞} as

$$\tau_{\infty} = \int_{0}^{\infty} dt \left(\left| 1 - \overline{\langle \alpha(t) \rangle} / \alpha_{\infty} \right| \right)$$
 (4.24)

 $\overline{\langle \alpha(t) \rangle}$ is a running (time)-average of the average number of primary particles in an aggregate. It is used to remove fluctuations due to the periodic changes in the flow. This definition of the relaxation time is analogous to the definition of the correlation time for stochastic processes as the integral over the autocorrelation function. For example, for an exponential relaxation process $\propto e^{-t/t_R}$ Eq. (4.24) leads to the expected result of $\tau_{\infty} = t_R$. However, Eq. (4.24) stays an appropriate measure for more irregular relaxation processes.

As a simple measure to characterize the steady state of the particle size distribution we use the average number of primary particles in an aggregate in the steady state that is defined as

$$\alpha_{\infty} = \lim_{t \to \infty} \left\langle \alpha(t) \right\rangle \ . \tag{4.25}$$

4.3.2. Approach to a Steady State

First, we use the average number of primary particles per aggregate to follow the convergence of the systems towards a steady state for all three splitting rules (Fig. 4.3).



Figure 4.2.: The histogram shows the percentage of fragmentation events for the number of fragments created in that event, for the same simulation run as shown in Fig. 4.3. Large-scale fragmentation leads typically to the smallest number of fragments, while erosion typically generates most fragments.

Our initial condition is always a uniform distribution of primary particles.

Initially, aggregation leads to a fast increase in the average number of primary particles per aggregate similar for all splitting rules. Then fragmentation sets in and a balance between aggregation and fragmentation is reached, with a different steady state average particle size for the different splitting rules. α_{∞} fluctuates over time, due to the statistical fluctuations in the flow. Large-scale splitting leads to the highest average number of primany particles per aggregate, erosion to the lowest and uniform fragmentation is in between. This can be intuitively understood, since for erosion typically more

fragments are created than for large-scale fragmentation (see Fig. 4.2). When a particle gets eroded, one of the fragments is usually close to the same size as the original aggregate. This leads to a high probability that this fragment will break again and therefore in many cases fragmentation will not be binary, but many fragments will be created. For large-scale fragmentation, aggregates will typically break only once, since both fragments are much smaller than the original aggregate.

In general, it is less likely in the case of large-scale fragmentation that a large number of fragments is created. This leads in the mean to a larger average aggregate size than



Figure 4.3.: Average number of primary particles as a function of time for aggregate strength $\gamma = 9$ and aggregate fractal dimension $f_d = 2.0$ for (a) large-scale fragmentation (b) erosion and (c) uniform fragmentation.

for erosion.

The different splitting rules lead to very different distributions (cf. Fig. 4.6). Largescale fragmentation creates a distribution with a single peak at intermediate radii and no particles of the smallest sizes. The right hand side of the aggregate size distribution for large-scale fragmentation follows approximately an exponential decay.

The size distribution found for large-scale fragmentation corresponds well to those observed for marine aggregates (Lunau et al., 2006) where exponential size distributions have also been reported. This may indicate that large-scale fragmentation is indeed the primary mode of break-up for many marine aggregates, as proposed in some works (see e.g. Thomas et al., 1999) and that erosion plays a very small role there.

By contrast, the size distribution for erosion has two different regimes, with a sharp maximum at the smallest aggregate size and a slower decaying tail at larger aggregate sizes.

Uniform splitting, where both larger and smaller fragments are created leads to a plateau in the size distribution at smaller aggregate sizes and an exponential decay to-

wards larger aggregate sizes.

Many of the system parameters that appear in our model can vary much in natural systems, in particular the aggregate strength, the number of primary particles involved and the fractal dimension of aggregates. In the following we therefore examine the sensitivity of our results to these parameters.

4.3.3. Influence of Aggregate Strength

To determine the influence of the forces holding the aggregates together on the resulting steady state size distribution, α_{∞} is computed for different values of the aggregate strength γ . α_{∞} increases with γ for all fragmentation rules. The increase is fastest for large-scale fragmentation and slowest for erosion. (see Fig. 4.4(a)). The relaxation time



Figure 4.4.: Influence of aggregate strength γ . (a) Average number of primary particles per aggregate in steady state α_{∞} and (b) relaxation time τ_{∞} for the approach to the steady state for different values of the aggregate strength γ . Error bars are obtained from an ensemble of 5 different realizations of the carrier flow.

 τ_{∞} as a function of γ is shown in Fig. 4.4(b). For all three fragmentation rules, the relaxation time is independent of the value of γ .

The relaxation time is defined *relative* to the average number of primary particles in

steady state α_{∞} and therefore the actual value of α_{∞} does not influence the relaxation time.

When looking at the particle size distribution in steady state (Fig. 4.6(a)-(c)), the difference between the three fragmentation rules is again clearly visible. However, this difference does not seem to depend on the value of the aggregate strength γ , as the distributions for each fragmentation rule remain qualitatively the same for different γ , but getting wider with increasing aggregate strength.



4.3.4. Influence of the Volume Fraction

Figure 4.5.: Influence of the total number of primary particles N_1 . (a) Average number of primary particles per aggregate in steady state α_{∞} and (b) relaxation time τ_{∞} for the approach to the steady state for different values of the total number of primary particles N_1 . Error bars are obtained from an ensemble of 5 different realizations of the carrier flow.

To determine the influence of the volume fraction, i.e. the total number of primary particles N_1 in the system, on the resulting size distribution, α_{∞} is computed for different values of N_1 . α_{∞} increases with increasing number of primary particles, due to the increased number of collisions. Again, the average number of primary particles per aggregate in the steady state α_{∞} is largest for large-scale fragmentation and smallest for



Figure 4.6.: Histogram of the particle size distribution. Number of particles versus the normalized radius r/r_1 for (a) different values of γ for large-scale splitting (b) different values of γ for erosion splitting, (c) different values of γ for uniform splitting, (d) different values of the total number of primary particles N_1 for large-scale splitting, (e) different values of N_1 for erosion splitting and (f) different values of N_1 for uniform splitting.

erosion. The increase in α_{∞} with N_1 is almost linear for all three fragmentation rules.

The relaxation time (see Fig. 4.5(b)) decreases for increasing N_1 for all three fragmentation rules. This again shows that the relaxation time does not depend strongly on the absolute value of α_{∞} . Instead, this indicates that the relaxation time is mainly determined by the collision rate between the particles. While a change in fragmentation rate, for example due to increased γ dues not affect the relaxation time, an increased collision rate, due to an increased number of primary particles in the system seems to decrease the relaxation time significantly.

Again, the size distributions (Fig. 4.6(d)-(f)) remain clearly different for the different fragmentation rules, independent of the total number of primary particles N_1 in the system.

4.3.5. Influence of the Fractal Dimension

The last important system parameter that typically varies a lot in natural systems is the fractal dimension f_d of the aggregates (Winterwerp, 1998). To determine the influence of the fractal dimension of the aggregates, α_{∞} is computed for different values of f_d .

For all three fragmentation rules we find a drastic increase in the average number of primary particles per aggregate in the steady state (see Fig. 4.7(a)) with increasing f_d .

In the case of varying the fractal dimension this increase in α_{∞} is much more drastic than for the other parameters studied in the previous sections. α_{∞} increases by approximately a factor of 100 between $f_d = 1.5$ and $f_d = 2.3$. Initially, one might assume that the increase of the average number of primary particles per aggregate in the steady state is only due to the aggregates becoming more compact as the fractal dimension is increased and does not really reflect a change in the aggregate size. However, plotting the average radius of the aggregates in the steady state as a function of f_d (inset in Fig. 4.7(a)) shows that there is also a significant increase in the aggregate size with increasing f_d .

This increase when varying the fractal dimension can be understood by looking at the stability condition for the aggregates (cf. Eq. (4.17)). The stability curve defined by Eq. (4.17) becomes almost horizontal for larger aggregate sizes. The larger f_d the greater becomes the range of aggregate sizes where increasing the size has almost no effect on the stability (with the limit of $f_d = 3$ where stability is independent of the size). Increasing the aggregate strength γ also leads to larger aggregates being stable at a given value of shear force. However, the increase in the range of stable aggregate sizes is much lower.

We note that the relaxation time increases weakly with f_d for all splitting rules (see Fig. 4.7(b)). Increasing the fractal dimension leads to more compact aggregates, i.e. less overall volume occupied by aggregates and therefore smaller collision probabilities. This

in turn increases the relaxation time. This effect appears strongest for erosion, where the relaxation time becomes very short for small values of f_d whereas for large-scale fragmentation and uniform fragmentation there seems to be a saturation of the relaxation time for values of $f_d < 2$.

Once again, the shapes of the particle size distributions retain their characteristic differences for the different fragmentation rules. Aside from the increasing fluctuations in the distributions for increasing f_d , due to the decreasing number of aggregates in the system, the qualitative shape of the distribution remains characteristic for the fragmentation rule, independent of the value of f_d .



Figure 4.7.: Influence of the fractal dimension f_d of the aggregates. (a) Average number of primary particles per aggregate in steady state α_{∞} , the inset shows the relative average size of aggregates in the steady state $r_{\infty}/r_1 = \alpha_{\infty}^{1/f_d}$ of the aggregates as a function of f_d and (b) relaxation time τ_{∞} for the approach to the steady state for different values of the fractal dimension f_d of the aggregates. Error bars are obtained from an ensemble of 5 different realizations of the carrier flow.

The range of f_d that is observed in natural systems reaches even further than $f_d = 2.3$, up to approximately 2.6. However, this is not shown here, because due to the drastic increase in the average number of primary particles per aggregate only very few aggregates would remain for such high values of f_d (if the other parameters remain fixed). Hence, no meaningful statistics or size distributions could be obtained.

This is a computational limitation of the current model, due to the finite number of primary particles that can be studied. While it occurs for every parameter, e.g. increasing the aggregate strength γ too far has the same effect, it is most pronounced for the fractal dimension. Small increases of the fractal dimension lead to proportionally much larger increases in the mean aggregate size, therefore reducing the total number of aggregates in the system very quickly.

4.4. Discussion

To conclude this work, in the final section we provide a brief discussion of the limitations of the particle-based aggregation and fragmentation model proposed here and finish with a brief summary of our results.

4.4.1. Limitations of the Model

In this work we showed the application of our particle-based model to the problem of aggregation and fragmentation of marine aggregates. We emphasize that the particlebased model introduced here is a very general model that can be applied to a wide range of other problems, for example in chemical engineering and has the potential to be a useful addition to the usual modeling approaches for aggregation and fragmentation. However, there are a number of limitations that should be pointed out. Some of these are due to physical aspects of the problem that are not yet fully understood and can therefore not be captured, others are mainly due to computational limitations.

The main physical aspect of the problem that is still not fully understood are the details of the fragmentation mechanism. In particular for marine aggregates, but also for many other systems where aggregates with a fractal-like structure appear, there still exists no satisfying microscopic theory for the fragmentation process. The fragmentation model used in this work can therefore only be considered as a very simplified view on the problem and most likely does not capture many aspects of the real situation. However, it serves as a useful basis to consider the qualitative impact of different fragmentation mechanisms on the overall size distribution of the aggregates.

Additionally, the equations of motion for fractal-like aggregates can be expected to be very different from the simple equations used here. The inclusion of the increased radius and effective density which was done in this work represents a very simple modification of the relevant forces, such as the drag forces acting on the particle and is unlikely to



capture the full complexity of the problem. However, it is a useful approximation to consider the impact of a complex structure on the steady state size distribution.

Figure 4.8.: Histogram of the particle size distribution. Number of particles versus the radius a for different values of the fractal dimension f_d of the aggregates for (a) largescale splitting (b) erosion splitting and (c) uniform splitting.

Furthermore, the description of marine aggregates as having a fractal-like structure is only a first approximation. Measurements have shown that when averaged over many different individual aggregates there exists a power-law relationship between the aggregate size and mass. This is certainly not true for each individual aggregate, but has been found to be a reasonable model in many cases (Kranenburg, 1994). In addition, the concept of a fractal dimension is usually only valid over a certain size range of aggregates. Aggregates that consist of only one or two primany particles generally do not have the same structure as a larger aggregate. In the context of this work it is possible to consider as primary particles the smallest fraction of an aggregate that still has the same power-law relationship as the large aggregates, i.e. as the fractal generator of the aggregate (see also the related footnote in Sec. 4.2.1).

In addition, there are a number of aspects that are theoretically understood quite well, but can not be included in such a model due to computational limitations. First among these is the two- and threeway coupling between the particles and the surrounding fluid. Two-way coupling,

i.e the feedback of the particles on the fluid can in principle be included but requires solving the equations of motion for the fluid together with the equations for the particles and therefore leads to a drastic increase in computational effort. In addition, as the particle radius r is assumed to be small, the feedback from the particle motion on the flow is usually not significant (Michaelides, 1997) and can therefore be neglected. The inclusion of three-way particle-flow coupling, i.e. the interaction of particles through the fluid is usually a bigger challenge. In particular at low Reynolds numbers this interaction affects the particle motion even at low particle concentrations (Brady and Bossis, 1988). While there exist a number of interaction models, that are able to compute this three-way coupling (see e.g. (Knudsen et al., 2008)) they are typically limited to systems with only a few particles because of the computational costs involved. While the modification of individual particle trajectories can be significant, the main effect for the collective aggregation and fragmentation dynamics of an ensemble of particle is typically a reduction of the collision rates. This can be approximated in a simple way by introducing a collision efficiency, i.e. a probability for aggregation upon collision. However, the introduction of such a collision efficiency does not qualitatively affect the results shown here. Additionally, the fluiddynamic interaction of permeable particles such as particles with a fractal-like structure discussed here are not understood very well. It is likely that flows through an aggregate can lead to very different interactions between fractal-like particles compared to solid spherical particles (see e.g. (Stolzenbach and Elimelech, 1994; Li and Logan, 2001)), in particular such permeability effects may actually decrease the influence of hydrodynamic interactions between the aggregates compared to the case of completely solid particles (Bäbler et al., 2006).

A further limitation for the applicability of the model is the number of primary particles that can be computationally considered. The present computational capacities do not allow to apply this approach to large systems, e.g. models that are used to study aggregation, fragmentation and aggregate transport on spatial scales up to several hundred kilometers. For large systems a rate-equation approach is therefore much better suited. However, for many small systems and also for the principle study of the processes involved, this is not a severe limitation.

Furthermore, as the model was tested with a simple 3-d synthetic turbulent flow, it is an open question how representative the results are to draw general conclusions about the evolving steady state size distribution. In particular intermittency effects and clustering of particles on the inertial scale of a real turbulent flow may significantly affect aggregation and fragmentation probabilities. In order to achieve more general statements it will therefore be necessary to study the model and the resulting size distribution for various, more realistic flows, for example using DNS simulations of real turbulent flows. Nevertheless, the influence of different system parameters and fragmentation mechanisms has been tested and gives a detailed insight for this specific flow.

4.4.2. Summary

In the present study we described in detail a coupled model for advection, aggregation and fragmentation of individual inertial particles with a fractal-like structure. We showed how typical properties of aggregation and fragmentation processes can be incorporated. In particular, we introduced an approximate way, using modified aggregate sizes and effective densities, to account for the fractal-like structure that is common for aggregates in many natural systems. The model represents an alternative approach to the rateequation based theory that is usually used to describe aggregation and fragmentation processes and was used to gain insights into principle behavior of fractal-like aggregates under different fragmentation mechanisms. The model was parameterized for the case of a suspension of marine aggregates in the ocean, but can in principle be used in a wide range of applications such as cohesive sediment dynamics, the flocculation of biological cells or solid-liquid separation systems in chemical engineering (Kranenburg, 1994; Han et al., 2003; Bäbler et al., 2008).

We observed the development of a balance between aggregation and fragmentation, leading to a steady state. It was found that with increasing aggregate strength the mean aggregate size in steady state increases, whereas the relaxation time stays constant. With increasing fractal dimension the relaxation time towards steady state and the mean aggregate size in steady state increase. By contrast, an increase in particle volume fraction decreases the relaxation time due to higher collision probabilities and increases the steady state mean size. In general, increased aggregation rates or decreased fragmentation rates lead to an increased mean aggregate size in steady state. The relaxation time decreases for increasing aggregation rates, but does not change with decreasing fragmentation rates.

In the context of our model different types of fragment size distributions can easily be tested and compared with each other. We compared numerical results for three commonly used distributions of fragment sizes, large-scale fragmentation where fragments typically have similar sizes, erosion, where one fragment is typically very small and uniform fragmentation, where all fragment sizes appear with the same probability. Large-scale fragmentation and erosion were treated as two separate processes to study the influence of fragmentation at a distance ζ from the center of the aggregate on the size distributions in the steady state. In reality, aggregates will break with certain probabilities at a distance ζ from the center but there will not be two separate processes. Therefore, depending on the probability distribution for fragmentation at a certain distance, one can expect different combinations of the steady state size distributions found in this work.

One such combination, where fragmentation at all distances ζ appears with the same probability was given by the uniform fragmentation rule. Uniform fragmentation leads to a distribution with a broad plateau for small aggregate sizes and an exponential tail towards larger aggregate sizes. A distribution with two different regimes evolves for erosion-like fragmentation. Large-scale fragmentation leads to an exponential tail of the particle size distribution. Similar shapes of the size distribution of the aggregates for largescale fragmentation have also been found in a number of theoretical and experimental studies (Spicer and Pratsinis, 1996a; Mietta et al., 2008), indicating that our model is able to reproduce the major features of such aggregation-fragmentation processes. Such an exponential tail has also been measured in field studies of marine aggregates (Lunau et al., 2006). This may indicate that large-scale fragmentation could be the primary mode of break-up for such aggregates, as has previously been discussed by e.g. Thomas et al. (Thomas et al., 1999).

In all cases the steady state particle size distribution follows a specific shape for each fragmentation rule. This indicates that the fragmentation process is most relevant for the shape of the distribution. The ratio of aggregation and fragmentation probabilities, mainly influenced by the aggregate strength, total particle volume fraction and fractal dimension, determines the mean aggregate size in steady state and the relaxation time. Out of these three parameters the fractal dimension has the strongest effect since it influences both aggregation and fragmentation probabilities.

The influence of large-scale fragmentation versus erosion for marine aggregates has recently been studied numerically and compared to experimental results in a work by Verney et al. (Verney et al., 2010). They used a rate-equation based Smoluchowski equation approach and obtained results comparable to those of our model. Thus, as both model approaches lead to similar results the insight into fragmentation and fragment distributions provided by the perspective of our model can provide a useful addition to the understanding of aggregation and fragmentation processes. Additionally, in the particlebased model presented here particle inertia can be fully considered, while the correct incorporation of particle inertia into a rate-equation based theory is still an unsolved problem. Hence, future model studies using this approach can lead to a better understanding of particle inertia effects in aggregation and fragmentation processes. Therefore, the model suggested here has the capability to be a powerful tool to investigate the validity of different approximative strategies in the formulation of a rate-equation based theory.

5. Rate Equations for Fractal-Like Aggregates

This chapter illustrates how to bridge the gap between the new, individual-particle based approach to aggregation and fragmentation that was introduced in this work and the usual rate-equation based approach typically found in the literature. The contents of this chapter have not yet been published elsewhere, but are written in the form of a publication in preparation.

5.1. Introduction

In this chapter the rate-equation based description of aggregation and fragmentation processes for fractal-like aggregates is discussed. While the Lagrangian approach using individual, inertial particles discussed in the last chapter offers many advantages, rateequations are much more suitable for large systems, where the computational effort of a particle-based approach is prohibitive. In this context the individual-particle based approach can be used as a guideline for comparison and a powerful tool to determine the validity of different approximations in the rate-equation approach. However, to be able to do this one needs to be able to compare these two approaches. The individual-particle based approach directly calculates collisions and fragmentation events locally, for each particle whereas in the rate-equation approach these are applied globally as a probability per time for a whole population of particles. While much effort has gone into determining suitable collision rates, in particular for inertial particles (see e.g. Ch. 2.3.4 of this work) the understanding of fragmentation rates is still much less complete. In this chapter we will therefore attempt to emphasize the fundamental connection between fragmentation rates and the underlying properties of the individual aggregates, namely the critical shear required to break an aggregates.

In the following a formulation for the aggregation and fragmentation of solid particles that form fractal-like clusters is discussed for an infinite, homogeneous system. There have been a number of efforts to incorporate such a fractal-like structure in a rate-equation approach, for example by Kranenburg (1994) or Maggi et al. (2007). Similar to most previous works inertial effects on the collision rates of the particles are ignored because even though some advances have been made (see e.g. Bec et al., 2005), even for fairly simple flow situations no equations exist that could be applied. For the fragmentation rate, the situation is slightly different. Since it is a single aggregate property inertial effects probably do not play a significant role. Still, the connection between the fragmentation rate in a turbulent flow and the properties of the individual aggregates is not very clear in most cases. While many approximations exist, they are usually based on the assumption of a Gaussian distribution of the shear forces responsible for breakup (see e.g. Bäbler et al., 2008). Here it is argued that even if the distribution of the individual velocity increments is Gaussian, as is for example the case in the synthetic turbulent flow used in the numerical model in the previous chapter, the full shear distribution is very different from a Gaussian. In particular, the probability to find a shear close to zero is very small. From this, an expression for the fragmentation rate is derived that leads to a great increase in the fragmentation rate compared to most models found in the literature.

Usually, once expressions for the collision and fragmentation rate are obtained the corresponding rate equation for aggregation and fragmentation is then solved numerically, for example with a size-class based model (Spicer and Pratsinis, 1996a) or a distribution based model (Maerz and Wirtz, 2009). In this way, spatial inhomogeneities can also be taken into account, by solving the model for different regions that are coupled via advective transport.

Here, an analytical solution of the aggregation - fragmentation rate equation is discussed. This is mainly for illustrational purposes, in principle a numerical model based on this description offers much more flexibility and offers a much more detailed description. However, we believe it is still instructive to see how far an analytical approach can be carried. For the special case of an exponential size distribution, which has for example been observed for the size distribution of marine aggregates in coastal areas, an explicit solution for the time evolution of the average aggregate mass can be found. We emphasize that in order to obtain correct expressions for the fragmentation rates it is necessary to take a reasonable approximation for the shear distribution. The solutions for the classical fragmentation rate obtained from a Gaussian distribution of the shear and for a Gamma distribution of the shear, as an approximation for the full shear distribution are compared. While the Gaussian distribution is commonly used in the literature for simplicity reasons, we argue that it is a poor approximation and for example leads to a very strong underestimation of fragmentation rates.

We find analytically that in most cases a non-trivial steady state solution exists. However, this solution is only stable under certain conditions. More precisely, the non-trivial steady state solution is only stable if aggregation dominates for small aggregate masses, whereas fragmentation must dominate for large aggregate masses. From this, a condition for the critical shear required to break up an aggregate can be derived. While this result is certainly limited to the special case for which the analytical solution is obtained, we believe similar results will hold even for the most general cases where only numerical solutions are possible.

The chapter is structured as follows. In Sec. 5.2 the description of a fractal-like aggregate and the general shape of the equation for the critical shear for fragmentation is discussed. In Sec. 5.3 the basic equation for the moments of the aggregate size distribution is derived. Section 5.4 discusses the equation for the relative growth rate and in Sec. 5.5 the collision and fragmentation kernels required for solving the equation for the relative growth rate are presented. In addition to the classical power-law expression for fragmentation this includes a new expression for the fragmentation rate derived from a model for the full distribution of the shear forces in a turbulent fluid. Finally, using the expression for the relative growth rate the equation for the moments of the size distribution can be solved, leading to an analytical solution for the average aggregate mass in the steady state and its dependence on particle and flow properties in Sec. 5.6. Section 5.7 contains a short summary and discussion of the results.

5.2. Fractal Aggregates and Critical Shear

When looking at real aggregates they are typically not spherical particles but instead can have a fractal-like structure as they consist of a number of primary particles. The structure of the aggregates can be characterized by a fractal dimension $f_d < 3$ Mandelbrot (1983). Their size can still be defined approximately by a radius, that can be considered as the characteristic length scale of the aggregate. This radius r_{α} of an aggregate consisting of α primary particles with a given fractal dimension f_d is derived in the following.

The relationship between mass and characteristic length r of an aggregate follows from the definition of the fractal dimension f_d (see e.g. Logan (1999)) as

$$\alpha = c_{fd} r^{f_d} , \qquad (5.1)$$

where c_{f_d} is a proportionality constant that can depend on f_d . As mass conservation must be fulfilled we get

$$m = \alpha m_1 = \alpha \rho_1 \frac{4}{3} \pi r_1^3 , \qquad (5.2)$$

where r_1 , m_1 and ρ_1 are the radius, mass and density of a primary particle, respectively. The proportionality constant c_{f_d} can be derived from Eqs. (5.1) and (5.2) by setting $m = m_1$

$$c_{f_d} = \rho_1 \frac{4}{3} \pi r_1^{3-f_d} \ . \tag{5.3}$$

In combination with Eqs. (5.1) and (5.2) this leads to

$$r_{\alpha} = \alpha^{1/f_d} r_1 \tag{5.4}$$

for the radius of an aggregate. It is evident that due to the fractal-like structure the radius r_{α} is greater than for a completely solid particle of the same mass.

The break-up of an aggregate occurs when the hydrodynamical forces F_{hyd} acting on the aggregate exceed the forces F_{agg} holding the particles in the aggregate together. The criterion for breakup can therefore be expressed as

$$F_{\rm hyd}/F_{\rm agg} > const.$$
 (5.5)

For aggregates with a fractal-like structure, consisting of a number of solid primary particle Generally, the critical shear required to break up an aggregate takes the form

$$S_{f,c} = \gamma \alpha^{-\theta(f_d)} , \qquad (5.6)$$

where $\theta(f_d)$ is a function with positive values for fractal dimensions $f_d < 3$. It can be seen that the critical shear force required to break up an aggregate decreases with the aggregate size, i.e. larger aggregates are less stable than smaller ones. Specific models for $\theta(f_d)$ can be derived in a number of ways and depend among other things on details of the aggregate structure (Sonntag and Russel, 1987b; Potanin, 1993; Kobayashi et al., 1999; Higashitani et al., 2001; Harada et al., 2006).

5.3. Moments of the Size Distribution

In systems where particles can both aggregate to form larger clusters and fragment again, there will typically be a distribution of aggregates of various sizes. It has been found in the past (see e.g. Spicer and Pratsinis, 1996a) that in many cases aggregation rates will dominate for small sizes, whereas fragmentation becomes the dominant process for large sizes. In this case the combination of these two processes leads to a (dynamic) steady state, where the overall size distribution remains approximately constant in time as long as external parameters such as shear rate in the flow remain unchanged. However, individual aggregates still grow and fragment, resulting in a constant flux of particles between various sizes. Generally, the existence of such a steady state will depend on the functional form of the aggregation and fragmentation kernels. Various forms have been compared for example by Vigil and Ziff (1989). Here we will analyze for one specific example the occurrence of a steady state by looking at the time evolution of the moments of the size distribution. Together, these determine the shape of the size distribution and therefore their change over time shows whether the system converges to a steady state solution.

For the description of this we start by assuming a continuous probability distribution¹ $p(\alpha, t) = \frac{C_N(\alpha, t)}{C_N(t)}$ of the aggregates. In the following $C_N(\alpha, t)d\alpha$ is called the number concentration of the aggregates and is the number of aggregates per volume consisting of a number of primary particles in the range $[\alpha, \alpha + d\alpha]$ at time t. $C_N(t) := \int_0^\infty d\alpha C_N(\alpha, t)$ is the total number of aggregates per volume at time t.

Typically, aggregation and fragmentation processes are described in terms of integral quantities of the size distribution of the aggregates, such as the average mass of aggregates $\langle \alpha \rangle$. Here, we are interested in obtaining an equation for the moments of the size distribution which can then be used to characterize the steady state. The average of α with respect to p is defined as

$$\langle \alpha \rangle := \int_{0}^{\infty} d\alpha \ \alpha p(\alpha, t) = \int_{0}^{\infty} d\alpha \ \alpha \frac{C_N(\alpha, t)}{C_N(t)} \ .$$
 (5.7)

Using this definition an equation for the evolution of the moments $\langle \alpha^n \rangle$, $n \in \mathbb{N}_{>0}$ of the size distribution can be derived. Taking the derivative with respect to t one obtains

$$\frac{d}{dt} \langle \alpha^n \rangle = \int_0^\infty d\alpha \ \alpha^n \frac{d}{dt} \frac{C_N(\alpha, t)}{C_N(t)} - \frac{\frac{d}{dt} C_N(t)}{C_N(t)} \int_0^\infty d\alpha \ \alpha^n \frac{C_N(\alpha, t)}{C_N(t)} .$$
(5.8)

The relative rate of change $\mu(\alpha, t)$ of the number of particles per volume of a certain size can be defined as

$$\mu(\alpha, t) := \frac{\frac{d}{dt}C_N(\alpha, t)}{C_N(\alpha, t)} .$$
(5.9)

¹In the previous sections α was always a discrete variable, but here we assume α to be continuous, which implies that the system contains primary particles within a continuous range of masses, instead of only one type of primary particles. The reason for this is mainly that for a discrete variable in the following all integrals turn into sums which can not be analytically evaluated. However, we believe that for large enough systems the results will qualitatively remain the same. Normally, one would write this more general problem, where primary particles of different mass can occur in terms of the aggregate mass m, instead of the number of primary particles per aggregate α . However, we keep using α in this section for consistency reasons with the rest of this work. But one should keep in mind this slight difference, that α is now assumed to be continuous.

Equation (5.8) can then be rewritten as

$$\frac{d}{dt} \langle \alpha^n \rangle = \langle \mu \alpha^n \rangle - \langle \alpha^n \rangle \langle \mu \rangle . \qquad (5.10)$$

With this equation the moments of the size distribution of the aggregates can in principle be calculated for all times, if the relative growth rate $\mu(\alpha, t)$ is known. However, this quantity is exceedingly difficult to determine and to date no complete derivation of $\mu(\alpha, t)$ even for very simple cases has been found and only some approximations are known. In the following, we will derive a solution for $\mu(\alpha, t)$, using approximate expressions for collision and fragmentation rates as well as for the shape of the probability distribution $p(\alpha, t)$.

5.4. Relative Growth Rate

The equation for the relative growth rate for aggregation $\mu_{agg}(\alpha, t)$ was first written down by Smoluchowski (1917). For a given value of α the number density of particles per volume $C_N(\alpha, t)$ increases when smaller particles aggregate in such a way that their combined size is α and decreases when particles consisting of α primary particles aggregate with any other particles. Formally, this can be written as

$$\mu_{\text{agg}}(\alpha, t) = \frac{1}{C_N(\alpha, t)} \cdot \left[\frac{1}{2} \int_0^{\alpha} d\alpha' \left(\chi(\alpha', \alpha - \alpha') C_N(\alpha', t) \cdot C_N(\alpha - \alpha', t) Q_{coll}(\alpha', \alpha - \alpha', t) \right) - \int_0^{\infty} d\alpha' \left(\chi(\alpha', \alpha) C_N(\alpha', t) C_N(\alpha, t) Q_{coll}(\alpha', \alpha) \right) \right].$$
(5.11)

Here, $\chi(\alpha', \alpha)$ is called the collision efficiency, i.e. the probability to coagulate upon collision and $Q_{coll}(\alpha', \alpha, t)$ is the collision kernel, i.e. the collision rate between particles of size α and α' at time t. In the same way a relative growth rate due to fragmentation $\mu_{\text{frag}}(\alpha, t)$ can be developed. The number of particles per volume $C_N(\alpha, t)$ increases due to larger particles breaking up so that the fragments are of size α and decreases due to particles of size α breaking up. This results in

$$\mu_{\rm frag}(\alpha,t) = \frac{1}{C_N(\alpha,t)} \cdot \left[\int_{\alpha}^{\infty} d\alpha' \left(\vartheta(\alpha',\alpha) C_N(\alpha,t) Q_{frag}(\alpha',t) \right) - C_N(\alpha,t) Q_{frag}(\alpha,t) \right],$$
(5.12)

where $\vartheta(\alpha', \alpha)$ is the probability that an aggregate of mass α' leads to a fragment of size α when it breaks. ϑ contains therefore the information about the number and size

distributions of fragments, for example binary or ternary fragmentation. $Q_{frag}(\alpha, t)$ is the fragmentation kernel, i.e. the fragmentation rate for a particle of mass α . The total relative growth rate is then given by $\mu(\alpha, t) = \mu_{agg}(\alpha, t) + \mu_{frag}(\alpha, t)$.

5.5. Collision and Fragmentation Kernels

The evaluation of the relative growth rate requires the aggregation and fragmentation kernel for a given system. Both the aggregation and fragmentation kernels will in general not only depend on α and α' but also on system parameters, for example the aggregate strength or the turbulence level in the flow.

In recent years much effort has gone into finding approximations for the aggregation kernels. Generally, the formulation of the aggregation rate is different for each system. Here, we are interested in particles aggregating in a turbulent flow, where aggregation happens due to collisions of particles. These collisions occur due to a number of physical mechanisms which are treated separately in theoretical considerations. They include collisions due to Brownian motion, differential settling, fluid shear and inertia effects. The collision kernels for Brownian motion, differential settling and fluid shear can be described fairly well theoretically whereas when particle inertia plays a role, effects such as preferential concentration and the occurrence of caustics in the particle dynamics lead to drastic modifications of the collision kernels that are still not fully understood (Bec et al., 2005). Therefore, we focus on the aggregation dynamics of tracer particles, i.e. particles that follow closely the dynamics of the surrounding fluid where inertia effects and settling can be neglected. In addition, collisions due to Brownian motion are neglected in the following, since this is usually only relevant for particles < 1 μm in diameter.

The collision kernel for non-interacting particles without inertia in turbulent shear was developed by Saffman and Turner (1956) and reads as

$$Q_{coll}(\alpha, \alpha') = \hat{c} \left(\frac{\epsilon}{\nu_f}\right)^{1/2} (r_\alpha + r_{\alpha'})^3 , \qquad (5.13)$$

where ϵ is the average dissipation of turbulent kinetic energy in the flow, ν_f is the fluids kinematic viscosity and r_{α} is the radius of a particle of size α . Here the radius is given by $r_{\alpha} \propto \alpha^{1/f_d}$. For solid particles the constant is given by Saffman and Turner as $\hat{c} = \sqrt{8\pi/15}$. This is the so-called rectilinear shear kernel (Thomas et al., 1999). In addition, we use a constant collision efficiency, i.e. $\chi(\alpha', \alpha) \equiv \chi_c$.

The fragmentation kernel poses a very different problem. On the one hand determining an expression seems easier because it only involves individual aggregates. On the other hand it can be extremely complicated because the microscopic properties of the aggregates play a very important role and generally both the fragmentation kernel and the distribution of fragments are not well understood.

For fragmentation, the first approximation for the probability $\vartheta(\alpha', \alpha)$ is that $\vartheta(\alpha', \alpha) = 2\delta(\alpha' - 2\alpha)$, where $\delta(x)$ is the Dirac delta function. This is the case of binary fragmentation, where both fragments are of the same size. In many aggregation systems, such as cohesive sediment dynamics, solid-liquid separation systems or marine aggregates this is a common assumption.

In our approach we assume that all aggregates of the same size have the same critical shear $S_{f,c}(\alpha)$. Here we study the case $S_{f,c} \propto \alpha^{-\theta(f_d)}$, where $\theta(f_d) > 0$, see Eq. (5.6). If the fluid shear S_f at the position of an aggregate exceeds this critical shear $S_{f,c}$ aggregates fragment. The probability for fragmentation of a given size is then only determined by the probability distribution of the shear $p(S_f)$, the influence of individual particle properties for the fragmentation kernel (see e.g. Ruiz and Izquierdo (1997)) is not considered. Assuming a homogeneous distribution of the particles in the flow, the fragmentation kernel is given by

$$Q_{frag}(\alpha) = \frac{\int_{S_{f,c}(\alpha)}^{\infty} dS_f \ p(S_f) / \tau(S_f)}{\int_{0}^{S_{f,c}(\alpha)} dS_f \ p(S_f)} , \qquad (5.14)$$

where $\tau(S_f)$ is the time scale on which the shear S_f persits around an aggregate, see e.g. Bäbler et al. (2008).

In this case the problem of evaluating $Q_{frag}(\alpha)$ lies in finding expressions for the probability distribution of the shear flow $p(S_f)$. The characteristic time of the shear S_f is taken to be proportional to the local Kolmogorov time scale and is given by $\tau(S_f) = (\hat{d}S_f)^{-1}$, with a constant \hat{d} . For the simple case of a Gaussian distribution of the shear, i.e.

$$p(S_f) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{S_f^2}{2\sigma^2}}$$
(5.15)

with $\sigma^2 = \frac{\epsilon}{15\nu_f}$ (see e.g. Saffman and Turner (1956)) the integral (5.14) can be solved analytically, yielding

$$Q_{frag}(\alpha) = 2\hat{d}\sqrt{\frac{2}{\pi}}\sigma \frac{e^{-\frac{S_{f,c}^{2}(\alpha)}{2\sigma^{2}}}}{\operatorname{erf}\left(\frac{S_{f,c}(\alpha)}{\sqrt{2}\sigma}\right)} .$$
(5.16)

For small aggregates the critical shear $S_{f,c}$ becomes large and the argument of the error function becomes much larger than one, meaning that the error function approaches one.

In the limit of small aggregates the fragmentation kernel therefore becomes

$$Q_{frag}(\alpha) = 2\hat{d}\sqrt{\frac{2}{\pi}}\sigma e^{-\frac{S_{f,c}^2(\alpha)}{2\sigma^2}}, \qquad (5.17)$$

which is the form of the fragmentation kernel proposed by Flesch et al. (1999). In the other limit of large aggregates the critical shear $S_{f,c}$ becomes small and Eq. (5.16) can be approximated by

$$Q_{frag}(\alpha) \approx \hat{d} \frac{4}{\sqrt{\pi}} \sigma^2 S_{f,c}(\alpha)^{-1} = \hat{d} \frac{4}{15\sqrt{\pi}} \frac{\epsilon}{\nu_f} S_{f,c}(\alpha)^{-1} .$$
 (5.18)

This is the familiar power-law formulation for the fragmentation rate employed by many authors, such as Pandya and Spielman (1982); Spicer and Pratsinis (1996a); Winterwerp (1998).



Figure 5.1.: Estimating the distribution of the shear forces in a flow with Gaussian velocity increments. The shear distribution is sampled numerically (green curve) from Gaussian distributions with standard deviation $\sigma = \left(\frac{\epsilon}{15\nu_f}\right)^{1/2} = 8$ (blue curve) and a Gamma function is fitted to this (red curve).

However, the assumption of a Gaussian distribution of the shear that this result is based on is generally not valid. While the probability distribution of velocity increments can have an approximately Gaussian distribution, at least on the largest scales in a turbulent flow Kailasnath et al. (1992); Sreenivasan (1999), the shear itself is a function of all the different velocity increments. In general, the shear force S_f in a flow is given by

$$S_f = \left(2\sum_{i,j} S_{ij}S_{ij}\right)^{(1/2)}$$
, (5.19)

where $S_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial X_j} + \frac{\partial u_j}{\partial X_i} \right)$ is the rate-ofstrain tensor in the flow. While the distribution of the velocity increments can indeed be Gaussian, which is for example

also the case for the synthetic turbulent flow used throughout this work, the distribution of S_f will be very different.

However, no exact analytical expression can be given for the distribution of the shear. Therefore, here the distribution of S_f is sampled numerically by assuming a Gaussian probability distribution of the velocity increments and then a suitable function is fitted to approximate the shear distribution (see Fig. 5.1). In principle, one could numerically calculate fragmentation rates directly from the real distribution. This can also be done for the case of a real turbulent flow where the velocity increments are not Gaussian. If the velocity field is known with sufficient detail, e.g. from measurements or DNS simulations one can numerically calculate an expression for the fragmentation rate for this field using the procedure explained below. However, since we are interested in an analytical solution here we need to derive an approximate expression for the shear distribution.

For the case of Gaussian velocity increments it was found that a Gamma function gives a reasonable fit to the overall shear distribution. To determine the dependence of prefactor of the Gamma distribution on the flow parameters, the fit was repeated for different values of ϵ/ν_f . We emphasize that there is indeed a strong difference between the assumption of a Gaussian distribution of the shear and the full shear distribution. In particular, the Gaussian distribution leads to the maximum of the shear distribution being at $S_f = 0$, whereas in the real shear distribution the probability to find a shear around $S_f = 0$ is very small. It can therefore be expected that a Gaussian distribution for the shear significantly underestimates the fragmentation probability. Assuming that

$$p(S_f) = \frac{\left(3\sqrt{\tilde{\epsilon}}\right)^5}{\Gamma(5)} S_f^4 e^{-3\sqrt{\tilde{\epsilon}}S_f} , \qquad (5.20)$$

where $\tilde{\epsilon} := \epsilon/\nu_f$, the equation for the fragmentation kernel (5.14) can be solved, leading to

$$Q_{frag} = \frac{\hat{d}}{3} \frac{40\tilde{\epsilon}^4 + 120\tilde{\epsilon}^{7/2}S_{f,c} + 180\tilde{\epsilon}^3 S_{f,c}^2 + 180\tilde{\epsilon}^{5/2} S_{f,c}^3 + 135\tilde{\epsilon}^2 S_{f,c}^4 + 81\tilde{\epsilon}^{3/2} S_{f,c}^5}{8e^{\frac{3}{\sqrt{3}}S_{f,c}}\tilde{\epsilon}^{7/2} - 8\tilde{\epsilon}^{7/2} - 24\tilde{\epsilon}^3 S_{f,c} - 36\tilde{\epsilon}^{5/2} S_{f,c}^2 - 36\tilde{\epsilon}^2 S_{f,c}^3 - 27\tilde{\epsilon}^{3/2} S_{f,c}^4} \ .$$
(5.21)

In the limit of large aggregates the critical shear $S_{f,c}$ becomes small and to leading order the fragmentation rate can be approximated as

$$Q_{frag}(\alpha) = \hat{d} \frac{40}{48.6} \left(\frac{\epsilon}{\nu_f}\right)^3 S_{f,c}(\alpha)^{-5} .$$
 (5.22)

It is noted that this fragmentation rate does indeed increase much faster with increasing aggregate size as the expression calculated from the assumption of a Gaussian distribution of shear (5.18). This is due to the much higher probability to find larger shear values when using the full shear distribution (5.20).

We emphasize that the relationship between Q_{frag} and its argument α depends on the

specific form of the stability condition for fragmentation, i.e. the specific model for $S_{f,c}$. It is through this dependence that the properties of the individual aggregates appear in the final result.

5.6. Steady State Solutions

In addition to the collision and fragmentation kernels, solving Eq. (5.11) for the relative growth rate requires some information about the shape of the number concentration of the aggregates. Here, we assume that an exponential distribution is an approximate steady state solution for the total relative growth rate $\mu(\alpha, t)$. Such exponential distributions have been observed for example for the number distribution of marine aggregates in coastal areas (Lunau et al., 2006) and is also found in many numerical studies, see for example the results in the previous chapter of this work. We take $C_N(\alpha, t) = \frac{C_N(t)}{\langle \alpha \rangle} e^{-\alpha/\langle \alpha \rangle}$. With this assumption of an exponential distribution which is characterized by the single parameter $\langle \alpha \rangle$ the system of equations for the all the moments (5.10) reduces to a onedimensional dynamical system, i.e. a single equation for $\langle \alpha \rangle$. A solution of this dynamical system is in this case equivalent to a solution for the full distribution.

With the collision and fragmentation kernels discussed above and the assumption about the shape of the distribution C_N the equation for the mean size of the aggregates can now be solved. Separating the contributions of aggregation and fragmentation the equation for the mean number of primary particles of the aggregates reads as

$$\frac{d}{dt} \langle \alpha \rangle = f(\langle \alpha \rangle)_{\text{agg}} + f(\langle \alpha \rangle)_{\text{frag}} , \qquad (5.23)$$

where

$$f(\langle \alpha \rangle)_{\text{agg}} := \langle \alpha \mu_{\text{agg}} \rangle - \langle \alpha \rangle \langle \mu_{\text{agg}} \rangle$$
(5.24)

$$f(\langle \alpha \rangle)_{\text{frag}} := \langle \alpha \mu_{\text{frag}} \rangle - \langle \alpha \rangle \langle \mu_{frag} \rangle .$$
 (5.25)

The contribution from shear aggregation, using the kernel (5.13) is given by

$$f(\langle \alpha \rangle)_{\text{agg}} = c(f_d) C_{N,1} \chi_c \epsilon^{\frac{1}{2}} \langle \alpha \rangle^{\frac{3}{f_d}} \quad , \tag{5.26}$$

where $C_{N,1}$ is the initial number density of primary particles and

$$c(f_d) = \frac{1}{4} \Gamma\left(f_d^{-1}\right) \frac{\hat{c}}{f_d^2 \pi^{3/2}} \left[2\sqrt{3\pi} f_d 27^{f_d - 1} \Gamma\left(\frac{3 + f_d}{3f_d}\right) \Gamma\left(\frac{2f_d + 3}{3f_d}\right) \right]$$

$$+3\pi 4^{\frac{1+f_d}{f_d}} \Gamma\left(\frac{f_d+2}{2f_d}\right) \Gamma\left(f_d^{-1}\right) \right] .$$
(5.27)

Using the complete expressions for the fragmentation rate, both with the assumption of a Gaussian distribution of the shear and with the Gamma distribution as approximation for the full shear distribution (Eqs. (5.16) and (5.21)) the equation for the relative growth rate can no longer be solved analytically. However, using the power-law approximations (5.18) and (5.22) for the fragmentation kernels for larger aggregates the mean growth rate and the contribution to the steady state can be calculated. In the case of the Gaussian distribution of the shear the contribution from fragmentation is given by

$$f(\langle \alpha \rangle)_{\text{frag,gauss}} = -d_1(f_d) \frac{\epsilon}{\nu_f} \gamma^{-1} \langle \alpha \rangle^{\theta(f_d)+1} , \qquad (5.28)$$

where

$$d_1(f_d) = \frac{\hat{d}}{2} \cdot \frac{4}{15\sqrt{\pi}} \Gamma\left(\theta(f_d) + 2\right) .$$
 (5.29)

In the case of a Gamma distribution of the shear the contribution from fragmentation is given by

$$f(\langle \alpha \rangle)_{\text{frag,gamma}} = -d_2(f_d) \left(\frac{\epsilon}{\nu_f}\right)^3 \gamma^{-5} \langle \alpha \rangle^{5\theta(f_d)+1} , \qquad (5.30)$$

where

$$d_2(f_d) = \frac{\hat{d}}{2} \cdot \frac{40}{48.6} \Gamma \left(5 \cdot \theta(f_d) + 2 \right) .$$
 (5.31)

A steady state solution corresponds to a fixed point $\langle \alpha \rangle^*$ of the dynamical system defined by Eq. 5.23. The equation for the fixed point is $\frac{d}{dt} \langle \alpha \rangle = 0$, corresponding to values of $\langle \alpha \rangle$ where $f(\langle \alpha \rangle)_{\text{agg}} = -f(\langle \alpha \rangle)_{\text{frag}}$. The 'standard' approach to determine the (linear) stability of a fixed point in a dynamical system by considering a Taylor series expansion around the fixed point and then looking at the Eigenvalues of the Jacobian (see e.g. Argyris et al. (1994)) does not work here. All derivatives at the fixed point are either zero or infinity, meaning the dynamical system considered here can not be expanded in a Taylor series around zero. However, the stability of a fixed point $\langle \alpha \rangle^*$ can easily be determined by examining $\frac{d}{dt} \langle \alpha \rangle$ for small deviations from the fixed point. If $\frac{d}{dt} \langle \alpha \rangle > 0$ in some neighborhood around the fixed point, the fixed point is unstable whereas if $\frac{d}{dt} \langle \alpha \rangle < 0$ in some neighborhood around the fixed point, the fixed point is stable.

It can be seen that without fragmentation the only steady state solution for the mean mass is $\langle \alpha \rangle^* = 0$. It is readily seen that for any small deviation from the fixed point, $\frac{d}{dt} \langle \alpha \rangle > 0$. The fixed point is therefore unstable and the mean mass grows to infinity.

The shear collision kernel is a so-called 'gelling kernel', in the absence of fragmentation it leads to a transition, where the system looses its mass to an infinite sized particle (Vigil and Ziff, 1989). Without aggregation, for both $f(\langle \alpha \rangle)_{\text{frag,gauss}}$ and $f(\langle \alpha \rangle)_{\text{frag,gamma}}$ the only steady state solution is also $\langle \alpha \rangle^* = 0$. For any deviation from this fixed point, $\frac{d}{dt} \langle \alpha \rangle < 0$ and the fixed point is therefore stable. Both fragmentation kernels are so-called 'shattering kernels', where the system looses its mass to a system of zero-sized particles (Vigil and Ziff, 1989).

The combination of aggregation and fragmentation typically leads to two fixed points.

- 1. The 'trivial' solution $\langle \alpha \rangle^* = 0$
- 2. a) For the Gaussian fragmentation kernel

$$\langle \alpha \rangle^* = \left(\frac{c(f_d)}{d_1(f_d)} \cdot \gamma \chi_c \epsilon^{-\frac{1}{2}} \nu_f C_{N,1}\right)^{\left(\frac{f_d}{(\theta(f_d)+1)f_d-3}\right)}$$
(5.32)

is a positive steady state solution for the mean number of primary particles if $(\theta(f_d) + 1)f_d \neq 3$. Otherwise, aggregation and fragmentation grow with the same power and a non-zero steady state solution only exists if the absolute values of the prefactors in Eqs. (5.26) and (5.28) are identical. In this case every value of $\langle \alpha \rangle$ represents a steady state solution.

b) For the Gamma distribution fragmentation kernel

$$\langle \alpha \rangle^* = \left(\frac{c(f_d)}{d_2(f_d)} \cdot \gamma^5 \chi_c \epsilon^{-\frac{5}{2}} \nu_f^3 C_{N,1}\right)^{\left(\frac{f_d}{(5 \cdot \theta(f_d) + 1)f_d - 3}\right)}$$
(5.33)

is a positive steady state solution for the mean number of primary particles if $(5 \cdot \theta(f_d) + 1)f_d \neq 3$. Otherwise, aggregation and fragmentation grow with the same power and a non-zero steady state solution only exists if the prefactors in Eqs. (5.26) and (5.28) have the same absolute values. Then every value of $\langle \alpha \rangle$ represents a steady state solution.

We emphasize once more that these steady state solutions for the mean number of primary particles per aggregate were derived under the assumption that an exponential distribution of C_N is a steady state solution for the total relative growth rate $\mu(\alpha, t)$. If that is the case these solutions for the mean determine the full size distribution in the steady state.

To determine the stability of the fixed points, i.e. to see whether the system will converge to one of these solutions one can again look at the sign of $\frac{d}{dt} \langle \alpha \rangle$ in some neighborhood around the fixed points. Since the system is one dimensional, if there are two fixed points it is sufficient to determine the stability of one of these, since the other one will be of the opposite type. Looking at the solution $\langle \alpha \rangle^* = 0$ the stability can readily be determined and from this we conclude the stability of the second, non-zero fixed point.

1. For the Gaussian fragmentation kernel the second, non-zero fixed point (5.32) is stable if

$$(\theta(f_d) + 1)f_d - 3 > 0 \leftrightarrow \theta(f_d) > \frac{3}{f_d} - 1 .$$

$$(5.34)$$

In this case the system approaches a non-zero steady state solution over time. This means that the fixed point $\langle \alpha \rangle^* = 0$ is unstable.

2. For the Gamma distribution fragmentation kernel the second, non-zero fixed point (5.33) is stable if

$$(5 \cdot \theta(f_d) + 1)f_d - 3 > 0 \leftrightarrow \theta(f_d) > \frac{1}{5} \left(\frac{3}{f_d} - 1\right) .$$

$$(5.35)$$

In this case the system approaches a non-zero steady state solution over time. Again, this implies that the fixed point $\langle \alpha \rangle^* = 0$ is unstable.

If the non-zero fixed point is stable, aggregation dominates for small average aggregate sizes and fragmentation dominates for large average aggregate sizes. If the non-zero fixed point is not stable, the aggregation rate increases faster for large average aggregate sizes than the fragmentation rate, whereas fragmentation dominates for small average aggregate sizes. Depending on the initial condition, the average number of primary particles per aggregate will then either grow to infinity or approach zero over time.

As already pointed out before, the assumption of a Gaussian distribution for the shear leads to an under-estimation of the fragmentation probability and when comparing Eq. (5.34) with (5.35) we can see now that this in turn decreases the stability regime for the non-zero fixed point. Therefore, taking such an over-simplified approximation for the shear distribution may lead in some cases to the conclusion that there is no stable steady state while in fact there is.

It is a rather remarkable result that the stability of the fixed point does not depend on any of the particle or flow properties besides the fractal dimension. While the fixed point itself shifts if for example the level of turbulence in the flow is changed or the aggregate strength changes, the stability of the fixed point is independent of that. This emphasizes the need for a more detailed understanding of the underlying fragmentation mechanism for different applications, as this is what leads to a specific formulation of $\theta(f_d)$.

While the solution presented here contains many approximations, among them neglecting particle inertia for aggregation, taking only the limit of large aggregates for determining the fragmentation rates and assuming an exponential size distribution, the whole approach could in principle also be carried out numerically with much fewer simplifications for a specific application. However, we believe that in principle similar results would be found.

5.7. Discussion

In this chapter we have presented a rate-equation based description of aggregation and fragmentation processes for aggregates with a complex, i.e. fractal-like structure. We started out with an equation for the time evolution of the moments of the number distribution of the aggregates. Its solution required expressions for the relative growth rates due to aggregation and fragmentation. These in turn depend mainly on the collision and fragmentation rates. While collision rates, at least for tracers, are fairly well established there is still an ongoing debate about the formulation of suitable fragmentation rates.

We showed how to formulate this rate-equation approach in such a way as to link it to the description of fractal-like aggregates that we used in the previous Chapters for our individual-particle based model. The main question is here how to link the notion of a critical shear required to break up an aggregate to an equation for a fragmentation rate.

We showed how in principle such an expression for the fragmentation rate of aggregates in a turbulent flow can be derived, at least under certain conditions, that explicitly incorporates the concept of a critical shear required for fragmentation. The calculation of this fragmentation rate required us to make an assumption about the distribution of the shear forces in the fluid flow. In most previous work a simple Gaussian distribution was assumed for the shear forces, while here we argued that the full distribution of the shear is likely to have a very different shape. Even if each velocity increment has a Gaussian probability distribution, the acting shear force will be a combination of these increments, leading to a very different result. We approximated such a shear distribution with a Gamma distribution and showed how these two assumptions, a Gaussian shear distribution and a Gamma shear distribution lead to very different expressions for the fragmentation rate.

Using approximations of these two different fragmentation kernels for the limit of large aggregates we were able to solve the equation for the time evolution of the moments of the number distribution of the aggregates for the simple case of negligible inertia and an exponential shape of the number distribution. While in particular the assumption of negligible inertia represents a severe restriction and more realistic collision kernels should be used in practice, we see this as an instructive example how such a calculation can be carried out. For more realistic situations, for example when one wants to use collision kernels obtained from inertial-particle based models as was for example done in Ch. 2.3.4 of this work or in Ayala et al. (2008b,a) the corresponding equations can only be solved numerically. Still, the principle remains unchanged and therefore we restrict ourselves to the simplest case where an analytical solution is still possible.

In particular, with these analytical expressions one can directly calculate the longterm behavior of such an aggregation-fragmentation system. We showed that in most cases, where both aggregation and fragmentation occurs there exists a non-zero steady state solution for the moments of the number distribution. However, the stability of this solution can be linked to the expression for the critical shear and it is found that as stable steady state solution only exists if certain conditions for the critical shear are met. In particular, it is required that aggregation dominates for small aggregate sizes, whereas fragmentation must dominate for large aggregate sizes.

Comparing these results with the previous chapter we find that under the simple assumption of a Gaussian distribution of the shear, the analytical calculations predict that our simple model for the critical shear (5.6) does not lead to a stable steady state. However, the numerical simulations using the individual particle-based approach from the last chapter clearly showed the development of a stable steady state from the balance of aggregation and fragmentation. This is due to the non-Gaussian distribution of the shear in the numerical model. Using the Gamma function approximation to incorporate this, the analytical approach also predicts the existence of a stable steady state for the critical shear model (5.6).

Our analysis illustrates how to connect the individual particle-based approach to the established rate-equation formulation for aggregation and fragmentation. The particle properties, in particular the critical shear required for fragmentation need to be translated into a fragmentation rate in the correct way. Linking the fragmentation rate directly to a physically easily understandable property of the aggregate such as the critical shear provides a deeper insight into the construction of appropriate fragmentation rates.

Finally, we emphasize that deriving expressions for the fragmentation rate based on a simple Gaussian shear distribution can lead to significant deviations in the fragmentation rate. Our assumption of a non-Gaussian shear distribution is much more realistic and leads to significant corrections in the fragmentation rates and we therefore believe that expressions similar to the one derived here can provide a useful addition to the established fragmentation rates from the literature.

6. Discrete Element Modeling of Fragmentation

In this chapter a detailed computational model for the fragmentation of an individual aggregate is discussed. The aim is to make a connection between the microscopic, physical properties of the individual components of an aggregate and macroscopic aggregate properties such as the critical shear stress required for fragmentation. The contents of this chapter have not yet been published elsewhere, but are written in the form of a publication in preparation.

6.1. Introduction

The direct simulation of the deformation and fragmentation of individual aggregates has received an increasing interest in recent years (Sonntag and Russel, 1987c; Potanin, 1993; Higashitani et al., 2001; Harada et al., 2006; Becker et al., 2009). While larger systems with many aggregates are still not computationally feasible, ever-increasing computational capabilities have made very detailed modeling of the interaction of individual particles and the resulting behavior of a single aggregate possible. The most common approach is that of soft particle molecular dynamics, often also called discrete element modeling (DEM). Such an approach provides very detailed insight into the dynamics of one individual aggregate by simulating each individual 'element', in this case each primary particle of an aggregate and the interaction with its neighbors and the surrounding fluid.

One of the first relevant studies in this direction was the work of (Potanin, 1993) who examined simple two-dimensional colloidal aggregates in a shear flow and characterized the elastic modulus and yield strength of such aggregates. Detailed simulations of the breakup of three dimensional aggregates both in shear and elongational flows were performed by Higashitani et al. (2001) who found a power-law relationship between the number of fragments created during breaking and the shear/strain intensity. Recently, Eggersdorfer et al. (2010) studied the restructuring and fragmentation of soft aggregates in fluid shear and found lognormally-shaped fragment size distributions as well as generalized scaling laws for the average number of primary particles per fragment.

In this chapter we follow up on these works and discuss a similar model for the manyparticle simulation of the fragmentation of an individual aggregate in a shear flow. The main goal is to bridge the gap between the microscopic (contact) properties of the individual elements (primary particles) and the macroscopic (fragmentation) behavior of the whole aggregate. This would allow one to understand the combined behavior of the whole aggregate as a function of a few parameters, namely the aggregate size, some measure of its structure, such as a fractal dimension and the contact properties of the individual primary particles. Such a description naturally leads to fragmentation models for whole aggregates, which are a main ingredient of the particle-based aggregation-fragmentation models discussed in the previous chapters of this thesis.

There we assumed based on very simple arguments that there is a critical fluid shear stress required to fragment an aggregate. Our arguments led to a power-law relationship between this critical shear stress and the number of primary particles in the aggregate where the exponent of the power law depends on the fractal dimension of the aggregate. Furthermore, we introduced models for the distribution of the fragments after breaking, such as large-scale fragmentation or erosion (see Ch. 4). The discrete-element model introduced in this chapter can now serve as a basis to validate and further improve these fragmentation models.

In the following a simplified model by Luding (2008) for the forces acting on the primary particles is introduced. It is a simplified approach in that sense that it provides a compromise between a realistic and an easy to use model. Many of the contact details are over-simplified, but only so far as they do not seem to be relevant for the macroscopic properties of the aggregate. It can be seen as a first step to understand the connection between microscopic and macroscopic properties, but still leaves room for improvement in the future.

There are two different types of interactions between the primary particles in an aggregate that are typically treated separately. On the one hand there are normal forces, i.e. they act between the centers of two primary particles. Usually, these forces are modeled with the classical Derjaguin-Landau-Verwey-Overbeek (DLVO) theory. This theory describes the combined effect of attractive van der Waals and repulsive double-layer (electrostatic) forces, (see e.g. Israelachvili, 1985). This is considered to be a reasonable model for non-touching particles aggregating in the so-called secondary energy minimum, resulting from the balance of electrostatic repulsion and van der Waals attraction. For particles aggregating in the primary energy minimum, i.e. when the surfaces are touching, the deformation of the surfaces leads to an elasto-plastic repulsion, dissipation of energy and to adhesion effects.

In addition to the normal forces, for particles whose surfaces are touching tangential (frictional) forces play an important role. It was shown experimentally by Pantina and Furst (2005) that aggregates from colloidal particles can have tangential forces that are capable of supporting a bending moment. The classical model for static friction was introduced by Cundall and Strack (1979) and has been extended by Luding to include

rolling and torsion resistance. In a similar framework, Becker et al. (2009) showed that without tangential forces, restructuring of aggregates is predicted incorrectly.

Aggregates without tangential interactions are usually called 'soft' as they can be deformed easily whereas systems with tangential interactions are called 'rigid'. While such aggregates are solids in the spirit of the Cauchy-Born theory (see e.g. Born and Huang, 1954) as an expansion around a (rigid) reference state, they do not have the periodic 'crystal' structure usually associated with solids and are therefore often referred to as 'amorphous'. An excellent discussion of the structure and rigidity of such amorphous solids from a slightly different viewpoint, emphasizing the role of stresses in the bond structure, can be found in the review by Alexander (1998).

In this chapter, we focus on the deformation and fragmentation of rigid aggregates, i.e. particles aggregating in the primary energy minimum, where tangential forces play an essential role. This is the most common situations and most real aggregates, such as the marine aggregates discussed in Ch. 4 fall into this category. Aggregates are generated via a diffusion-limited/reaction-limited aggregation (DLA/RLA) algorithm and then placed in a simple linear shear flow. The fragmentation behavior is studied, as a function of both the strength of the shear stress and the aggregate properties, such as number of primary particles or fractal dimension of the aggregate. It is found that, depending on the aggregate properties, a critical shear stress exists beyond which the aggregate breaks. The line of critical shear stress is estimated in parameter space from a series of simulations. Close to the critical shear stress aggregates typically break into two fragments (binary fragmentation). For shear stresses significantly larger than the critical shear stress the size of the fragment is found to be independent of the initial size of the aggregates. The size of the fragments depends only on the level of shear stress.

6.2. Discrete Element Modeling of Fractal-Like Aggregates

To be able to perform computer simulations of the deformation and fragmentation of an aggregate it is first necessary to generate an initial aggregate with a certain structure. In many natural systems and industrial applications it is found that aggregates possess a fractal-like structure. For fractal objects the exact shape and structure can not be described in a simple manner, but the general shape can be expected to change predictably as the viewing scale changes. This self-similarity over different scales is an essential property of fractal objects and has been found in many objects in nature, from snowflakes to coastlines (Mandelbrot, 1983). In general, for a fractal object the number of particles

N within a circle of radius a scales as

$$N = ca^{f_d} av{6.1}$$

where c is a proportionality constant. For aggregates of finite-size this can obviously only be true for a limited range of a-values, at most ranging from the size of the primary particles r_1 to the size of the whole aggregate r_{agg} .

In most natural systems an individual aggregate will not exhibit this fractal scaling, instead one can measure the properties of an ensemble of aggregates of various sizes. One then relates the cross-sectional area A_{solid} or the solid volume V_{solid} to a measure of the characteristic length l of the aggregate. Here, l can be a number of different things, such as the longest diameter, the mean diameter or the equivalent diameter (see Ch. 2.1.1). In many cases this relationship between area or volume and characteristic length will on average exhibit a fractal scaling

$$A_{\rm solid} \propto l^{D_2}$$
 (6.2)

$$V_{\rm solid} \propto l^{D_3}$$
, (6.3)

where D_2 and D_3 are the two- and three dimensional fractal dimension, respectively. This value can be obtained from the slope of a log-log plot when the respective aggregate property (area or volume) is plotted over the characteristic length.

The book by Logan (1999) provides a table with approximate values of this fractal dimension for a number of examples, from inorganic colloids formed by Brownian motion $(D_3 \approx 1.8 - 2.1)$, yeast flocs in rotating tubes $(D_3 \approx 2.66)$ to marine snow composed of diatoms in the ocean $(D_3 \approx 1.52)$.

6.2.1. Generating Initial Aggregates

To generate such aggregates numerically, a number of different algorithms have been proposed, that lead to aggregates with different structures. In general, particles will be released at certain sites in space and then move in a specific manner until they come into contact. The different models refer to different assumptions about the motions of the particles and the collision process. In some cases the assumptions may approximate the real process of aggregate formation, whereas in other cases they are strongly idealized. Here, three different cases are discussed 1) diffusion-limited aggregation, 2) ballistic aggregation and 3) reaction-limited aggregation and for each case there are two different subcases, particle-cluster and cluster-cluster aggregation.

1. Diffusion-Limited Aggregation



Figure 6.1.: A diffusion-limited particle-cluster aggregate consisting of 1000 primary particles $(r_1 = 4\mu m)$.

Diffusion-limited aggregation is probably the most widely used model for the generation of aggregates with a fractal structure. It has been introduced first by Witten and Sander (1981, 1983) who studied a numerical model for aggregation processes on a lattice. A particle is placed at the center of the lattice, then a second particle is added to a random site far away from the center. This particle moves randomly along the lattice until it reaches a site next to the center particle. Then it becomes part of the aggregate. Another particle is now introduced and the process continues. If a particle touches the boundaries of the system it is removed and a new particle introduced at a random site. This approach has since then been extended to offlattice simulations, where each new particle performs a Brownian motion until it connects with the aggregate. The structure of such diffusion-limited aggregates has been extensively studied, see e.g. Meakin and Vicsek (1985); Vicsek et al. (1990).



Figure 6.2.: Determining the fractal scaling of aggregates. 1000 DLA aggregates in the range of $10^1 - 10^5$ primary particles $(r_1 = 4\mu m)$ were created. The log-log plot shows the number of particles in the aggregate as a function of the radius of the aggregate. The slope of the linear fit (red) gives the fractal dimension $f_d = 2.5 \pm 0.002$.

Aggregates created in this manner exhibit a fractal scaling, in two dimensions the fractal dimension is $f_d \approx 1.71$ and in three dimensions $f_d \approx 2.5$, see Fig. 6.2. They show a clear center and fairly symmetric branches going off to the sides.

This procedure to create diffusion-limited aggregates is a particle-cluster aggregation processes. By contrast, diffusion-limited aggregates can also be created via cluster-cluster aggregation. For cluster-cluster aggregation, a number of smaller aggregates are created as described above, but these clusters also move around in a Brownian motion and collide to form still larger aggregates. Cluster-cluster aggregation generally leads to more open structures than particle-cluster aggregation, for diffusion-limited cluster-cluster aggregation in three dimension the fractal dimension is $f_d \approx 1.80$. Their overall structure is more chain-like and does not have the clear center of the particle-cluster aggregates.

2. Ballistic aggregation

If the mean free path of particles is long compared to their size, their motion can be approximated as ballistic instead of the Brownian motion used for the diffusionlimited aggregation. For ballistic aggregation, the primary particles can penetrate deeply into the fixed aggregate, leading to very compact structures with $f_d \approx 3$ in three dimensions. Ballistic cluster-cluster aggregation again works the same way, where now also the smaller aggregate clusters move ballistically. Again, the collision of smaller clusters leads to more open structures than for particle-cluster aggregation, in this case ballistic cluster-cluster aggregates leads to a fractal structure with $f_d \approx 1.95$ in three dimensions.

3. Reaction-limited aggregation

The term reaction-limited aggregation generally refers to an extension of the diffusion-limited aggregation process. For diffusion-limited aggregation every collision leads to an attachment. However, in many cases there is a repulsive force between particles, for example from electrostatic double-layer forces which can hinder adhesion upon collision. This is then modeled by requiring a certain number of collisions before a particle sticks. In general, this leads to more compact aggregates than the simple diffusion-limited aggregation process. The asymptotic value of the fractal dimension depends on the required number of collisions but is close to 3 for particle-cluster aggregation and close to 2 for cluster-cluster aggregation.

In this work aggregates created via diffusion-limited particle-cluster aggregation and reaction-limited particle-cluster aggregation are studied (Fig. 6.1). The reason for this choice is that efficient implementations of these aggregation algorithms are freely available on the internet. Here the program by Stock (2006), an arbitrary-dimensional, off-lattice diffusion-limited aggregation simulator is used to create aggregates with different numbers of primary particles.

6.2.2. Particle-Particle Interactions

In this section the interaction forces between primary particles in an aggregate are described, these include both normal forces such as van der Waals attraction, elasto-plastic deformation, repulsion and adhesion as well as tangential friction forces.

First, we discuss normal intecation forces, i.e. those acting in the direction of the contact vector $\mathbf{n}_{ij} := (\mathbf{X}_i - \mathbf{X}_j) / ||\mathbf{X}_i - \mathbf{X}_j||$ between two particles *i* and *j*. The forces are typically expressed in terms of the overlap $\delta_{ij} := (r_i + r_j) - ||\mathbf{X}_i - \mathbf{X}_j||$ between two particles.

The model for the normal forces acting between particles consist of three regimes. At large distances, there are no interactions between particles as long-range interactions such as particle-particle interactions through the fluid are neglected. When particles approach each other to a distance δ , with $\delta_{\min} < \delta < 0$, van der Waals forces lead to an attractive interaction between the particles. For simplicity, more complex force models, such as retardation of van der Waals forces in a liquid and double-layer forces are neglected. Instead, van der Waals forces are modeled by the simplified Hamaker model (Hamaker, 1937)

$$\boldsymbol{F}_{ij}^{vdW} = -A \frac{r_{\text{eff}}}{12\delta_{ij}^2} , \qquad (6.4)$$

where $r_{\text{eff}} = r_i r_j / (r_i + r_j)$ and A is the Hamaker constant. Strictly speaking, A is not a
constant but can depend on a number of factors, such as the distance δ_{ij} between particles. However, the variations in A are small, typically the values range between $10^{-20}J$ and $10^{-19}J$. Rigorous expressions can be obtained from the Lifschitz theory involving quantum field theory calculations, but here a simple approximation as a constant is used. The Hamaker equation has a singularity at $\delta_{ij} = 0$, however this is not a problem in practice as real surfaces are never perfectly smooth and therefore only approach to a minimum distance. Therefore, the force is shifted by a value δ_{\min} so that the maximum van der Waals force $\mathbf{F}_{ij}^{vdW(max)} = -A \frac{r_{\text{eff}}}{12\delta_{\min}^2}$ is reached for $\delta_{ij} \geq 0$. Typically, δ_{\min} should be in the range of the molecules involved.

If particles come into contact, their elasto-plastic deformation, repulsion and adhesion is modeled with a piecewise linear hysteretic spring model (Luding, 2008). The model includes an increasing stiffness of the particles with increasing deformation. Dissipation takes place due to the hysteretic nature of the contact force. However, small amplitude deformations may not be damped enough and therefore, a small viscous, velocity depended dissipative force is added to the contact force.

The total force in normal direction, as a function of the overlap δ_{ij} is given by the sum of the van der Waals force, hysteretic contact force and viscous dissipation.

In reality, a nonlinear hysteresic model is more likely close to the reality than the piecewise linear model used here, but since experimental information is missing, it is believed that such a model represents a reasonable compromise between capturing the essential details and simplicity.

In addition to the normal forces, tangential (friction) forces play an important role in determining the stability and stiffness of aggregates. In addition to forces, tangential interactions can lead to torques that produce a rotation of the primary particles. Within the model of Luding (2008) that is used here the tangential interactions are implemented as three different components, (1) sticking and sliding friction, (2) rolling resistance and (3) torsion resistance, corresponding to different rotational degrees of freedom. The tangential interaction model for each component follows the original approach of Cundall and Strack (1979), where at contact virtual springs are attached, measuring the sliding, rolling and torsion of the contact point.

6.2.3. Fluid Forces

In addition to the contact forces, each primary particle is subject to hydrodynamic forces and torques from the surrounding fluid. Here, only quasi-steady drag forces and torques following Stokes law are used, which is a reasonable approximation for very small Reynolds numbers. The quasi-steady drag force from the point-force approximation, using the undisturbed flow field at the position of the particle, is given by (see e.g. Sec. 2.3.2 of this work)

$$\boldsymbol{F}_D = -6\pi\eta_f r_i (\boldsymbol{V}_i - \boldsymbol{u}) , \qquad (6.5)$$

where u is the velocity of the undisturbed surrounding fluid at the position of the particle. The torque on a particle associated with the quasi-steady drag force in the point force approximation is given by (see e.g. Landau and Lifschitz, 1991, p. 83-84)

$$\boldsymbol{M}_D = -8\pi\eta_f r_i^3(\boldsymbol{\omega}_i - \boldsymbol{\Omega}) , \qquad (6.6)$$

where $\mathbf{\Omega} = \frac{1}{2} \nabla \times \boldsymbol{u}$ is the vorticity of the fluid.

This is the so-called free draining approach, where the interaction of particles through the fluid is neglected. In particular the drag force and torque on particles which are shielded from the flow due to the presence of neighboring particles are overestimated in the free-draining approach. Eggersdorfer et al. (2010) compared their numerical results of aggregate fragmentation in the free-draining approach with the studies of Higashitani et al. (2001) and Harada et al. (2006) and found some differences but similar scaling laws. A recent discussion of the limitations of the free-draining approach in the context of aggregate deformation can be found in the paper of Becker et al. (2009).

A simple alternative to the free-draining approach that does not lead to a significant increase in computational cost is the approach presented in Sec. 2.3.4 of this work. In a suspension of spheres, the effect of a finite volume fraction C_V (see 2.2.1 for a definition) of particles can be estimated in terms of a correction to the drag force and torque on a single particle. This correction factor f_C relates the drag force and torque at a given volume fraction $F_D(C_V)$ and $M_D(C_V)$ to the undisturbed drag force and torque F_D and M_D of a single particle, i.e.

$$f_C := \frac{\boldsymbol{F}_D(C_V)}{\boldsymbol{F}_D} = \frac{\boldsymbol{M}_D(C_V)}{\boldsymbol{M}_D} \ . \tag{6.7}$$

For a given particle Reynolds number, the most commonly used formulation for the drag correction is given by

$$f_C = (1 - C_V)^{1 - b_0} , (6.8)$$

with an empirical coefficient $b_0 \approx 4.5$ (see e.g Ch. 2.3.4 of this work).

6.2.4. Implementation

Finally, we discuss the specifics of the numerical implementation used in the following. For many systems of interest, such as marine aggregates, the detailed contact properties between particles are not known. Most systems where some range for the contact properties is available in the literature are systems from engineering applications, such as suspensions of polystere latex particles in water, which is one of the most studied particle aggregation systems (Eggersdorfer et al., 2010). For example, for the case of marine aggregates discussed in Ch. 4 only data for the primary particle radius (approximately $1\mu m$ to $10\mu m$) and densities (approximately $1\rho_f$ to $3\rho_f$) can be found in the literature, but no information on contact properties (Logan, 1999).

Here, we choose primary particles of $4\mu m$ radius suspended in water, where the particles have twice the density of water. For the contact parameters we take values suggested in Luding (2008) for cohesive powders of micrometer size. The complete list of parameters can be found in Appendix B. However, we emphasize that tests with different ranges of parameter values showed no significant changes to the qualitative behavior of the system. We therefore believe that our results will also be applicable to other systems, as long as the same microscopic mechanisms are at work.

Three dimensional aggregates with different numbers of primary particles are created using an arbitrary-dimensional, off-lattice diffusion-limited particle-cluster aggregation simulator (Stock, 2006). With the standard settings for particle-cluster aggregation the algorithm creates aggregates with a fractal dimension of $f_d = 2.5$. Since we are also interested in the dependence of the simulation results on the fractal dimension, we also create aggregates with a different fractal dimension. Since diffusion-limited particlecluster aggregation always leads to the same fractal dimension the algorithm is modified so that not every collision between particles leads to an attachment. By tuning this number of collisions before attachment such reaction-limited particle-cluster aggregates can achieve any fractal dimension between $f_d = 2.5$ and $f_d = 3$.

The particles are suspended in a simple linear shear flow in x_1 -direction where the fluid velocity changes linearly in x_2 -direction and does not depend on the other two coordinates, i.e.

$$\boldsymbol{u}(\boldsymbol{x}) = S_f \cdot \begin{pmatrix} x_2 \\ 0 \\ 0 \end{pmatrix} . \tag{6.9}$$

The aggregate center of mass is placed in the center of the coordinate system, where the flow velocity is zero. The shear stress acting in the flow is $G = \mu_f S_f$, where S_f is the fluid shear (see for example Ch. 3.2.3 for a definition) and μ_f is the fluids dynamic viscosity.

To determine the number and size of individual fragment after fragmentation we use a cluster algorithm to identify groups of connected particles (see e.g. Stauffer and Aharony, 1995). Primary particles are assumed to belong to the same cluster if the overlap δ_{ij} :=

 $(r_i + r_j) - ||\mathbf{X}_i - \mathbf{X}_j||$ is greater than zero.

A final, important criterion for the stability of the contact algorithm is the time scale of the molecular dynamics time step Δt_{MD} . In particular the time step needs to be chosen small enough to resolve the contact duration between two particles. Since the particle contacts are implemented as springs they have a typical time scale given by the eigenfrequencies of their oscillation. These frequencies depend on the spring stiffness parameters. For a given parameter set the time step therefore need to be chosen small enough to be below the characteristic time scales of the particle contacts, given by the eigenfrequencies of the contact springs. Both the normal and the tangential and rotational directions have to be considered here. For the parameter values used here, a time step of $\Delta t_{MD} = 10^{-5} \mu s$ was found to be sufficient to ensure the stability of the system.

6.3. Simulation Results

In this section we show the simulation results for the fragmentation of an individual, diffusion-limited aggregate in a linear shear flow. It is examined how the fragmentation behavior of an aggregate changes with changing magnitude of the applied shear stress and with changing fractal dimension of the aggregate.

6.3.1. Critical Behavior

We start out with a diffusion-limited particle-cluster aggregate with a fractal dimension of f_d in a linear shear flow. When examining the dynamics of an aggregate for different values of the applied shear stress one finds two distinct regimes, separated by a critical shear stress value. For small values of the fluid shear stress below this critical point the aggregate rotates around its center of mass but remains intact. For larger values of the fluid shear stress, above the critical point the aggregate is first deformed and stretched and then the connections between certain particles are broken until the aggregate fragments into separate pieces. The fragments are then advected in opposite directions in the flow, see Fig. 6.3. This can easily be seen when plotting the number of fragments after a certain time in the flow (here $1000\mu s$) as a function of the applied fluid shear stress. Figure 6.4 shows this for the case of an aggregate consisting of 250 primary particles with the parameter values from Tab. B.1 in Appendix B, but the same qualitative behavior can be found for any other choice of aggregate.

For small values of the shear stress there is only one fragment, the initial aggregate, whereas beyond a certain point the number of fragments increases with the applied fluid shear stress. There is a critical point, i.e. a certain critical shear stress beyond which



6.3. Simulation Results

the aggregate breaks. This critical shear stress is what has been used in the aggregationfragmentation modeling in the previous sections as the relevant fragmentation property of an aggregate. It provides the connection between the microscopic properties of individual aggregates and the individual particle based model discussed in Ch. 4 of this work, as well as a rate-equation based approach as for example discussed in Ch. 5. Determining the dependence of this critical shear stress on the aggregate size and microscopic properties of the aggregates, such as the fractal dimension is the main goal of this chapter.

The problem with determining the exact value of the critical shear stress for a given aggregate is that close to the critical shear stress level it is very difficult to determine whether an aggregate will fragment. The time until breaking diverges as one approaches the critical point and the critical point is therefore not directly accessible in a numerical simulation. We therefore fix a maximum time of $t_{\text{max}} = 1000 \mu s$ and if for a given shear stress the aggregate has not broken by this time we will assume that it does not fragment at this shear stress level. One should therefore keep in mind that the critical shear stress determined with this method only represents an upper boundary for the real critical shear stress and other choices of t_{max} will lead to slightly different scaling relationships.

In addition, for each realization of a diffusion-limited aggregate the structure of the aggregate will be different and therefore the critical point will be found at a slightly different shear stress for each individual aggregate even if all parameters are identical. For each parameter set we therefore use 10 different realizations of the initial aggregates and calculate all results as averages over these realizations of the aggregates.

Figure 6.5 (blue markers) shows the computed critical shear stress as a function of the aggregate size for diffusion-limited aggregates with four different initial sizes, each as an average over ten realizations of the initial aggregates. The critical shear stress decreases with the aggregate size, i.e. large aggregates are less stable than smaller ones. The critical shear stress G_c changes approximately linearly as a function of the number of primary particles per aggregate α on a log-log plot. This suggests a power-law relationship of the form $G_c \propto \alpha^{-\theta}$ with some exponent θ for the critical shear, similarly to what has been used in Ch. 4 and 5 of this work. For $f_d = 2.5$ and the system parameters detailed in Tab. B.1 in Appendix B we find that the exponent $\theta \approx 1.08 \pm 0.03$. To see the dependence of the critical shear on the fractal dimension the same set of computations is repeated for the case of reaction-limited initial aggregates with a fractal dimension of $f_d = 2.6$ (green markers in Fig. 6.5). Again, the critical shear changes as a power law function of the number of primary particles in the initial aggregate. However, for this increased fractal dimension the critical shear decreases much slower with α than for the case of $f_d = 2.5$. The value of θ can be obtained from a fit as $\theta \approx 0.63 \pm 0.04$.

It is interesting to compare these results for $\theta(f_d)$ with the simple model that was used



Figure 6.4.: Number of fragments created during the breaking of an aggregate consisting of 250 primary particles with $r_1 = 4\mu m$ for different values of the applied fluid shear stress. The arrow indicates the critical point at a shear stress of $G_c \approx 3.5$ beyond which the aggregate fragments.

in Ch. 4 of this work for the critical shear. There it was argued that the break-up of an aggregate occurs when the hydrodynamical forces $F_{\rm hyd}$ acting on the aggregate exceed the forces $F_{\rm agg}$ holding the particles in the aggregate together. The criterion for breakup can therefore be expressed as

$$F_{\rm hyd}/F_{\rm agg} > const.$$
 (6.10)

Assuming that the hydrodynamical force is proportional to the shear force integrated over the surface of the aggregate and the force F_{agg} holding an aggregate together is proportional to the area of constituent matter in a cross-section of the aggregate we found that the splitting condition becomes

$$G_c \propto \alpha^{-(2/f_d - 2/3)}$$
 (6.11)

This model therefore predicts that $\theta(f_d) = \frac{2}{f_d} - \frac{2}{3}$. This would lead to $\theta(f_d = 2.5) \approx 0.13$ and $\theta(f_d = 2.6) \approx 0.1$, respectively. This implies a much weaker dependence of the critical shear on the aggregate structure than what was actually observed in the simulations performed in this section. Such a model under-estimates the fragmentation rate, compared to what our microscopic simulations suggest.

A simple alternative model, that leads to a faster decrease of the critical shear with the aggregate size and therefore to an increased fragmentation rate, could be to assume that the force F_{agg} holding the aggregate together is proportional to the fraction of constituent matter in a cross section of the aggregate, instead of the absolute area. The area of constituent matter is proportional to $A_{\alpha,\text{solid}} \propto \alpha^{\frac{2}{3}}$. Dividing this by the total area in a cross section, $A_{\alpha,\text{total}} \propto \alpha^{2/f_d}$ and again equating F_{agg} and $F_{\text{hyd}} \propto \alpha^{2/f_d}$ leads



Figure 6.5.: Critical shear required for the fragmentation of a DLA aggregate as a function of the number of primary particles in the aggregate for two different values of the fractal dimension. The linear fit suggests a power-law relationship with an exponent $\theta(f_d = 2.5) \approx 1.08 \pm 0.03$ and $\theta(f_d = 2.6) \approx 0.63 \pm 0.04$

 to

$$G_c \propto \alpha^{-(4/f_d - 2/3)}$$
 (6.12)

Such an approach predicts $\theta(f_d) = \frac{4}{f_d} - \frac{2}{3}$ which implies a much faster decrease of the critical shear stress with the aggregate size. This would lead to $\theta(f_d = 2.5) \approx 0.93$ and $\theta(f_d = 2.6) \approx 0.87$, respectively. While still not exactly the result found in the simulations this is much closer to the results than what was predicted by the model from Ch. 4.

A number of other models for the critical shear stress have been proposed in the literature. For example Potanin (1993) and Baldyga and Bourne (1995) suggested in a similar analysis that the critical shear stress should be proportional

$$G_c \propto \alpha^{-\frac{1}{2q}}$$
, where $q = \frac{f_d}{2n(3-f_d)}$ (6.13)

and n is a measure of the average number of bonds of each individual particle. Sonntag and Russel (1987a) found for this that in flocculated networks $n \approx 2.5 - 4.4$. With this, the model predicts that $\theta(f_d = 2.5) \in [0.5, 0.88]$ and $\theta(f_d = 2.6) \in [0.38, 0.68]$, which is slightly lower than what was found in the simulations here. However, it was pointed out by Potanin (1993) that the analysis is only valid for completely rigid particles, whereas particle deformation should lead to larger values of θ . While within our model aggregates possess tangent interactions and are therefore able to support a bending moment, this tangential interaction is weak and aggregates are getting deformed, as can be seen directly from Fig. 6.3. This may be one reason for the fairly high values of θ found in our study.

6.3.2. Supercritical Behavior

In addition to the critical shear stress one can also look at the supercritical behavior of the aggregates, i.e. what happens for shear stresses above the critical point. Close to the critical point the aggregates always fragment into two fragments of similar size, corresponding to the large-scale fragmentation model discussed in Ch. 4 of this work. However, for increasing shear stress Fig. 6.4 already indicated that the number of frag-



Figure 6.6.: Average number of primary particles per fragment $\langle \alpha(t) \rangle$ during the breakup of an individual diffusion-limited particle-cluster aggregate as a function of time. Each linecolor indicates a different level of the applied fluid shear stress, whereas each linestyle corresponds to a different initial number of primary particles of the aggregate.

ments increases. Figure 6.6 shows the change in the average number of primary particles per fragment as a function of time for different initial aggregate sizes and different levels of the applied fluid shear stress.

The average number of primary particles per fragment decreases over time as more and more fragments are created. The results in Fig. 6.6 are for an individual aggregate and not taken as an average over an ensemble of initial aggregates and are therefore quite noisy. Nonetheless, it is clear that after some time the number of particles per fragment approaches a minimum value. In this example that happens around $t = 100 \mu s$. Here, the fragmentation process stops and the remaining fragments are once again stable and will not break up any further in the currently applied fluid shear stress.

What is interesting about this result is that the average number of primary particles per fragment does not seem to depend significantly on the initial size of the aggregate but only on the level of the applied fluid shear stress. For supercritical shear stress aggregates undergo a breaking cascade where each fragment is repeatedly broken up into two or rarely three new fragments, until the fragments are small enough so that that the applied shear stress is below their critical shear stress. The size of these fragments is therefore approximately independent of the original aggregate size but depends on the applied shear stress because they break until a stable fragment size is reached.

6.4. Discussion

In this chapter we discussed the application of a discrete-element model to the fragmentation of an individual aggregate in a laminar shear flow. The key feature of such a model is how to implement the contact properties between individual, primary particles. Here, we used a spring-dashpot model to simulate both central forces, such as adhesion and elasto-plastic repulsion and tangent forces, such as friction and torsion resistance. While such an aggregate possesses a certain rigidity, the model still allows for aggregate deformation and restructuring, as is usually the case for most applications. However, there are some limitations to the approach.

First, it should be noted that the model approach used here only represents a simple, piecewise-linear approximation of a full hysteretic contact model. In reality, the full contact laws will most likely be much more complex, including real hysteresis, non-linear behavior and other effects. For active systems, such as the formation of marine aggregates in the ocean where biological processes on the aggregates play an important role, the contact properties may also vary strongly over time, both on short scales for example over a tidal cycle and on long scales, such as between seasons. Nonetheless, while we do not capture fully every aspect of the particle contacts the model represents a compromise between numerical effort and detail and is still able to capture qualitatively the main macroscopic aggregate properties that result from the microscopic contact details.

Second, finding appropriate values for the system parameters is rather difficult since not all the quantities required correspond directly to real physical properties. Here we used a parameterization for cohesive powders with particles in the micrometer range. Nonetheless, we do not believe that this is a big restriction for the generality of our results, since tests with different parameter ranges showed a qualitatively similar behavior. Therefore, we conclude that as long as the principal mechanism remains unchanged, systems with different values for the contact parameters will still exhibit a similar behavior.

Third, while diffusion-limited aggregates represent a simple approach to construct aggregates with a fractal-like structure for computer simulations, it should be kept in mind that in many cases this is only a very rough approximation. The structure of aggregates can be much more complex, consisting of very irregular primary particles and may vary strongly between individuals and also over time. Using such a simple model to construct the aggregates can therefore only be viewed as a first step, or as an approximation for cases where no further details about the aggregate structure are available. In principle, for each specific problem one would have to construct aggregates with an appropriate structure.

Fourth, in addition to a simple linear shear there may other structures in a real turbulent flow that can be more efficient at breaking up aggregates. For example, for extensional flows, Higashitani et al. (2001) found that the critical strain decreases much faster with the aggregate size, implying that aggregates may be much more sensitive to fragmentation in such a flow type than in a simple linear shear.

Finally, the interaction between particles and fluid is only captured as a lowest order approximation, where two-way particle-flow coupling and particle-particle interactions through the fluid are neglected. It was shown by Becker et al. (2009) that this so-called free draining approximation leads to an over-estimation of the drag force and therefore under-estimating the critical shear stress. In particular the particles in the center of the aggregate are in reality shielded by the surrounding particles and therefore do not feel the full fluid shear stress. While this is certainly a big limitation of the model results presented here, we believe that neglecting these effects is still a reasonable first approximation. At the very least it can serve to illustrate in principle how such a model could be used to go from the microscopic aggregate properties to a macroscopic expression that is needed for the individual, inertial particle based model discussed in Ch. 4 and the rate-equation based approach of Ch. 5. However, it is likely that including such coupling effects, either by direct numerical simulation or by appropriate approximate coupling schemes, such as those discussed in Sec. 2.3.4 of this work will lead to some modification of our results.

Despite these limitations, using the model approach discussed here we were able to show how to connect the microscopic properties of the contacts between individual particles into a macroscopic quantity of the whole aggregate, namely the critical shear stress required for fragmentation. Individual aggregates of different size were placed in a linear shear flow and their breaking behavior was studied. First, we confirmed that there are indeed two different regimes in the particle behavior as a function of the fluid shear stress, separated by a critical shear stress value as has been assumed in the simplified fragmentation models in the previous chapters. For 'small' values of the shear stress, i.e. smaller than a certain critical value aggregates remain intact and simply rotate in a shear flow whereas for large shear stress values aggregates are broken up into smaller fragments. We showed how the critical shear stress changes as a function of the number of primary particles in the initial aggregate. The function for the critical shear stress is a power-law, where the exponent depends on the fractal dimension of the aggregate. For more compact aggregates, i.e. with a higher fractal dimension the critical shear stress decreases slower with the number of primary particles per aggregate. Such a power-law expression was also used in the aggregation-fragmentation models in the previous chapters. We found that while there are small changes in the value of the critical shear stress between individual aggregates of the same size the deviations are not very large. This implies that at least for the case of DLA/RLA aggregates studied here the characterization of the aggregate structure via the fractal dimension is a reasonable approximation to determine the fragmentation behavior of different aggregates.

We compared the results for the slope of the power-law function for the critical shear stress with different theoretical predictions. While it seems clear that the simple assumption of the aggregate strength being proportional to the area of constituent matter in a cross-section of the aggregate underestimates the aggregate stability, our results are not sufficient to differentiate between the other discussed possibilities. Here, future studies with more simulations and a detailed analysis of the fragmentation process are required for a more conclusive answer.

With regard to the number and size distributions of the fragments, at least for the current contact model between the primary particles, our results suggest that close to the critical shear stress level aggregates tend to break into two fragments of similar size. This corresponds to the large-scale fragmentation mechanism discussed in Ch. 4 of this work, supporting the assumptions made there. If the shear stress greatly exceeds the critical shear stress, the discrete-element simulations of this chapter suggest a cascade mechanism similar to what was used in the individual, particle based models in Chs. 3 and 4 of this work. Initially, two fragments are created, however if these are too large to be stable at the given shear stress level they fragment again. This is repeated until all fragments are below the critical size defined by the current level of shear stress.

In addition to what was already found here further studies with a discrete element model such as the one presented here may help to greatly enhance our understanding of the details of the fragmentation mechanism for an individual aggregate. This can then be used to generate macroscopic expressions of the aggregate properties, such as the critical shear stress that are based on the microscopic interactions within the individual aggregate. A deeper understanding of this connection is essential in the formulation of better models for full aggregation-fragmentation systems and as already shown in the previous sections is a crucial feature for the determination of the overall aggregate size distribution in such systems.

7. Conclusions and Outlook

In this thesis we discussed aggregation and fragmentation processes of particles in a fluid flow. We introduced a model approach based on the motion of individual, inertial particles as an addition to the usual rate-equation models that are widely used. While less detailed than the full hydrodynamical simulation of individual aggregates, which is still computationally not feasible for larger systems, our approach is able to provide a new perspective on aggregation and fragmentation processes. It has the potential to be a powerful tool, in particular in the investigation of different approximation strategies for the usual rate-equation models.

We presented two applications of our model, for two very different scenarios. On the one hand we studied the dynamics of spherical, viscous droplets in a turbulent flow. For this, we discussed two different fragmentation mechanisms, fragmentation due to shear and fragmentation due to particles exceeding a maximum size. In many natural systems, for example for raindrops in a cloud, fragmentation due to a maximum limiting size is the main mechanism for fragmentation. Shear fragmentation is more relevant when both particle and fluid viscosity are similar, for example when creating emulsions. Using our model we were able to reproduce steady state size distributions, resulting from the balance of aggregation and fragmentation, as well as the dependence of the steady state on the particle and flow properties. In this context, the main advantage of our approach lies in its ability to directly include particle inertia into the model description.

On the other hand we studied the application of our model to systems of solid particles, where aggregation leads to clusters with a complex, often fractal-like structure. We discussed how to approximate such a fractal-like structure within the framework of our model, by using a description based on an increased radius and modified density. Again, we examined steady state solutions and how these changed as a function of particle and flow properties. In this case, the fractal dimension of the aggregates was found to be the most significant system parameter. Additionally, while for spherical, viscous droplets the fragmentation behavior is well understood and suitable models exist, for solid aggregates with a fractal-like structure this is not the case. A number of different, possible fragmentation mechanisms are discussed in the literature and it seems different systems may possess very different mechanisms. Here our model approach has the advantage that different fragmentation models can directly be incorporated in terms of easy to interpret macroscopic properties of an individual aggregate, such as the critical shear required for the breakup of an individual aggregate. This makes it particularly easy to compare the effect of different fragmentation mechanisms, for example large-scale fragmentation into fragments of similar size with erosion-like processes where fragments of very different size are created. Our results show that this distribution of the fragments during breaking is indeed the most important parameter for the shape of the steady state aggregate size distribution. Comparing the size distributions obtained with our model with measured size distributions for different systems may give some indication what mechanism is active in that system. For example, large-scale fragmentation always leads to an exponential size distribution such as those measured for marine aggregates in coastal waters, indicating that this might be the primary fragmentation mechanism for such aggregates.

Both of these examples illustrate the important role of fragmentation for aggregation processes of particles in fluid flows, something that has been neglected in many works for a long time and has only recently started to come into focus. For all systems studied here, in particular the steady state size distribution depends strongly on the fragmentation behavior, while aggregation seems to be more relevant for the transients, for example affecting the time to reach the steady state.

While these two applications illustrated the wide usefulness of our model approach in very different systems and showed how our approach is able to predict steady state size distributions as well as the dependence of the steady state on the particle and flow properties another question immediately comes to mind. That is, how does our model fit in with the established rate-equation approach as well as the full hydrodynamical simulations of individual aggregates? By examining the connection between our model and these approaches we showed how our model is able to bridge the gap between the existing models. While using rate-equation based approaches one is able to simulate very large systems in a reasonable amount of time such an approach relies on many approximation and parameterizations. Our model is able to provide such approximations, for example by directly simulating collision and fragmentation for inertial particles, which can then be translated into suitable collision and fragmentation rates for a rate-equation based model. Additionally, our model approach can serve as a basis of comparison to check the accuracy of different approximation strategies within a rate-equation model. In this work we presented a rate-equation based model for fractal-like aggregates and examined how to calculate fragmentation rates that correspond to our particle-based model. We showed how one can in principle calculate steady state solutions. These calculations once again revealed the great importance of fragmentation for the steady state of the aggregation-fragmentation system.

On the other end, while full hydrodynamical simulations of aggregates are able to provide very detailed information about the behavior of individual aggregates, they are limited to very small systems. Using such a discrete-element model for the fragmentation of an individual aggregate we showed how to take the detailed microscopic information obtained there and translate this into a macroscopic single-aggregate property, the critical shear, that can directly be incorporated in our model. This can then in turn be used to generate approximations for rate-equation based approaches, linking the detailed microscopic simulation of individual aggregate properties to models than can be used for large-scale simulations.

In summary, in this thesis we were able to introduce a new approach for the description of aggregation and fragmentation processes, pointed out the connection to existing approaches and laid a solid foundation for work to come. While many questions were answered during the course of this work and in particular the important role of fragmentation became very apparent from our results some open questions remained. To conclude, we will therefore point out some of these open questions and discuss possible directions for further work, based on what we have shown here.

In the first part of this thesis we discussed collision rates between inertial particles. This is an important aspect for the formulation of rate-equation based models. While much progress has been made in this area, there are still many aspects that are not fully understood. Most approaches to calculate collision rates work with the so-called ghost collision approach, where collisions between particles are computed but they do not have a physical consequence. This assumption only works well for very dilute suspensions, where the ensemble dynamics is not strongly affected by collisions. However, for example Medrano et al. (2008) pointed out the existence of bursts in the particle dynamics if collisions are taken into account. Such interactions may lead to strong modifications of the collision rates. Sundaram and Collins (1997) argued that it may be possible to capture such effects as additive corrections to the collision rate obtained using a ghost-collision approach.

In addition, what has so far been neglected in many cases is the enhancement of the collision rates due to the settling of the particles under gravity. For example, for raindrop formation it is believed that the extremely fast growth of droplets in clouds may be due to a few larger particles collection many smaller ones in a very short amount of time as they fall downwards through the cloud. Preliminary calculations suggested that such strong enhancements can indeed be found when including a sinking velocity in the equations of motion for the particles. However, particularly in this context the role of coupling between the particles and the fluid as well as between particles through the fluid may become very important. While a number of studies exist for spherical particles, the situation is much less clear for particles with a complex structure, where intra-particle flows may lead to strong modifications of the surrounding flow field. For a first discussion on this we refer

to the work of Stolzenbach and Elimelech (1994).

For the dynamics of spherical, viscous particles this work mainly focused on the steady state behavior, when a balance between aggregation and fragmentation was obtained. However, in many situations the transient behavior can be very relevant. For example, for weather forecasts, the time it takes to initiate rainfall from a cloud is still a great unknown. Here in particular including particle sinking may lead to much better predictions. As already stated above, it is believed that the fast growth of a second peak in the particle size distribution that has been observed (see e.g. Shaw, 2003) may be due to a few larger droplets collecting a host of small ones as they sink through a cloud. In addition, it may be an interesting question whether the simple, synthetic turbulent flow field that was used here to approximate the dissipative range of a real turbulent flow is really able to sufficiently capture all important effects. Real turbulent flows may exhibit intermittency, as well as clustering effects on scales larger than the Kolmogorov scale. The particle Stokes number may change along the trajectory of a particle, leading to fluctuations in the equations of motion for the particles. All this can in principle be incorporated within the framework of our model, but judging the impact of such effects requires further study.

For the modeling of aggregates with a fractal-like structure one of the most interesting questions would be how to describe polydisperse suspensions of particles, i.e. suspensions where different types of particles appear. The easiest case would be having particles of different density in the system. For each aggregate one would not only have to keep track of the number of primary particles in that aggregate, but also of the ratio between the different types of particles. This immediately leads to a modification for the equation for the aggregate density, which instead of the density of the primary particles now contains the weighted average of the densities of all the different types of primary particles. This could be a first approximation for describing aggregates from different sources, e.g. marine aggregates consisting of inorganic matter such as clay or silt and organic matter such as phytoplankton cells. More complex approaches could include particles with different radii or different binding strengths. In addition, in many cases restructuring effects of the aggregates may be very interesting. For example Maggi et al. (2007) discussed the effect of a variable fractal dimension in the framework of a rate-equation based model. In many systems the fractal dimension may change as a function of time, either due to repeated aggregating and fragmenting of the aggregates or due to other effects, such as changing biological activity, for example from bacteria on the aggregates which may affect the aggregate strength as well.

Within the rate-equation based approach discussed here one of the main questions that remained was how to approximate fragmentation rates for real turbulent flows. The assumption of Gaussian distribution of the velocity increments is usually only a very poor model, except for the largest scales of a turbulent flow. Generally, the tails of the distribution of the velocity increments tend to become longer as the scale decreases. For example, Sreenivasan (1999) suggested a description based on stretched exponentials. The incorporation of such a model for the fluid flow may lead to more realistic expressions for the fragmentation rate than those that are currently widely in use. In addition, the approach discussed here raised the question whether a more general solution than the specific case of an exponential aggregate size distribution can be obtained. In particular, the question of what is really required for a distribution to be a solution of the Smoluchowski equation and whether this would lead to similar steady state solutions as those suggested here has not been answered in this work. For this, it may prove fruitful to compare the analytical results discussed in this thesis with numerical solutions of the Smoluchowski equation and see whether for example a similar dependence on the fragmentation mechanism can be found.

In our opinion discrete-element approaches for fragmentation, such as the one discussed here may prove to be the most useful direction for a better understanding of fragmentation processes in the future. With the ever-increasing computational resources available, a large number of possibilities exist. Some questions that immediately come to mind are examining the dependence of the critical shear on the strength of individual, microscopic bonds, as well as comparing the influence of normal and tangent interactions. For example, changing the strength of the tangential bonds, i.e. making the aggregate more or less rigid one should be able to see the transition between the two theoretical limit cases of completely rigid and completely floppy aggregates, which have both been discussed separately in the literature so far (see e.g. Potanin, 1993). The inclusion of particlefluid coupling, as well as modifications for example of the drag forces due to shielding from neighboring particles may lead to drastic changes in the critical shear required for fragmentation (see e.g. Becker et al., 2009). Additionally, in many realistic systems not all particles are identical, i.e. one may have to deal with polydisperse aggregates with primary particles of varying density and size and there may be a distribution of bonds of different strength between different particles in a single aggregate, which may greatly modify the breaking behavior. Also, using such a discrete-element model one may be able to answer in detail some of the questions already raised above, such as the effect of aggregate restructuring and changing fractal dimensions over time. For a given type of particles, does the influence of shear or repeated aggregation and fragmentation lead to some kind of a 'limit' structure that the aggregate approaches?

Finally, one of the most interesting questions with respect to applications is how to include biological and chemical processes on the aggregates in this context. So far the model contains only physical interactions, such as the van der Waalsinteraction between particles. Biological and chemical processes, e.g. the presence of bacteria or polymer coatings on the aggregates may greatly influence the connections between the primary particles. In the framework of such a discrete-element aggregate model one is able to directly model the colonization of an aggregate with bacteria in a flow, as well as the dynamics of bacteria on such an aggregate that can have a significant influence on the aggregate composition, its structure, sinking and fragmentation behavior.

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Appendix and Bibliography

A. Incompressible, Synthetic Turbulence using a Spectral Approach

In this chapter we discuss how to construct a 'random flow' that models certain characteristics of a real turbulent flow. Such flows are called 'synthetic' turbulence. While they do not capture all features of a real turbulent flow, they can serve as simple models for a certain flow characteristic and can be simulated with much less computational effort than real turbulent flows. Here, we discuss a specific model for synthetic turbulence that models features of the dissipative range of a turbulent flow by imposing an energy spectrum.

The idea behind the spectral approach to synthetic turbulence is to write a flow $\boldsymbol{u}(\boldsymbol{x},t) = \{u_n(\boldsymbol{x},t)\}_{n=1...d}$ as a Fourier spectral representation. The Fourier components are then described by a stochastic process with statistics suitable to reproduce certain features of turbulent flows, e.g. the energy spectrum or the structure functions.

If one is interested in systems with periodic boundary conditions, the Fourier spectrum is discrete and a flow $\boldsymbol{u}(\boldsymbol{x},t)$ can be written as a Fourier series

$$u_n(\boldsymbol{x},t) = \sum_{\boldsymbol{k} \in \mathbb{Z}^d \setminus \{\boldsymbol{0}\}} \hat{u}_n(\boldsymbol{k},t) e^{i\frac{2\pi}{L}k_m \cdot x_m} , \qquad (A.1)$$

where L is the size of the periodic domain and $\hat{\boldsymbol{u}}(\boldsymbol{k},t) \in \mathbb{C}^d$ are the Fourier components, with the property $\hat{u}_n(-\boldsymbol{k},t) = \hat{u}_n^*(\boldsymbol{k},t)$ because $u_n(\boldsymbol{x},t)$ is real-valued. The star denotes complex conjugation.

In the Fourier spectral representation the condition of incompressibility of the flow field $\frac{\partial}{\partial x_n}u_n(\boldsymbol{x},t) = 0$ reads as $\frac{2\pi i}{L}k_n \cdot \hat{u}_n(\boldsymbol{k},t) = 0$, i.e. each Fourier component needs to be perpedicular to the corresponding wave vector \boldsymbol{k} . In two dimensions the easiest way to do this is to introduce a stream function and its Fourier components. An approach to synthetic turbulence using this can be found in the work of Sigurgeirsson and Stuart (2002). Here, a different idea is shown, that can also be applied to higher dimensional flows.

By taking for $\hat{\boldsymbol{u}}(\boldsymbol{k},t)$ the projection of a different stochastic process $\hat{\boldsymbol{v}}(\boldsymbol{k},t) \in \mathbb{C}^d$ onto the plane perpendicular to the wave vector \boldsymbol{k} , i.e.

$$\hat{u}_n(\boldsymbol{k},t) = \hat{v}_n(\boldsymbol{k},t) - \frac{(\hat{v}_m(\boldsymbol{k},t) \cdot \boldsymbol{k}_m)}{|\boldsymbol{k}|^2} k_n$$
(A.2)

incompressibility can be ensured for any *d*-dimensional flow field. The stochastic process $\hat{v}_n(\mathbf{k}, t)$ is assumed to be an Ornstein-Uhlenbeck process. It is a solution of the complexvalued stochastic differential equation

$$d\hat{v}_n = -\alpha(\mathbf{k})\hat{v}_n dt + \sigma(\mathbf{k})dW_n , \qquad (A.3)$$

with $\alpha(\mathbf{k}), \sigma(\mathbf{k}) \in \mathbb{R}$, where dW_n is an complex Wiener increment. The stationary distribution corresponding to this stochastic differential equation is a Gaussian distribution with mean $\langle \hat{v}_n \rangle = 0$ and variance

$$\langle \hat{v}_n^* \hat{v}_n \rangle = \frac{\sigma(\mathbf{k})^2}{\alpha(\mathbf{k})} , \qquad (A.4)$$

without summing over n. The parameters $\alpha(\mathbf{k}), \sigma(\mathbf{k})$ need to be chosen so that the flow $u(\mathbf{x}, t)$ reproduces some features of a real turbulent flow.

For the situations studied in this work it is often sufficient to consider only very few Fourier modes. In that case it may be computationally appropriate to calculate the flow field directly at the position of the particles by evaluating the Fourier sum (A.1). In that case the flow is not calculated on a grid, as is usually the case and no interpolation is required. This allows for the resolution of very fine structures in the particle dynamics. To this end a k_{max} is set, and the Fourier sum is only taken to this value. The lowest value where isotropy can be ensured is $k_{\text{max}} = 1$, which results in 8 Fourier modes in two dimensions and 26 modes in three dimensions. In computer simulations it is convenient to use expressions involving real and imaginary parts of the Fourier modes $\hat{u}_n(-\mathbf{k}, t)$. Using the property $\hat{u}_n(-\mathbf{k}, t) = \hat{u}_n^*(\mathbf{k}, t)$ of the Fourier modes allows the elimination of some Fourier modes and the rewriting of the Fourier series. Using this, only non-negative values of the first component k_1 of the wave vector \mathbf{k} need to be considered. For the case of $k_1 = 0$ only non-negative values of the second component k_2 of the wave vector need to be considered, and so on. For example in three dimensions the Fourier sum then reads as

$$u_n(\boldsymbol{x},t) = \sum_{k_1=1}^{k_{\max}} \sum_{k_2=-k_{\max}}^{k_{\max}} \sum_{k_3=-k_{\max}}^{k_{\max}} \left[\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\} \cos(\frac{2\pi}{L}(k_1x_1+k_2x_2+x_3x_3)) + \operatorname{Im}\{\hat{u}_n(\boldsymbol{k},t)\} \sin(\frac{2\pi}{L}(k_1x_1+k_2x_2+k_3x_3)) \right] \\ + \sum_{k_2=1}^{k_{\max}} \sum_{k_3=-k_{\max}}^{k_{\max}} \left[\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\} \cos(\frac{2\pi}{L}(k_2x_2+k_3x_3)) \right]$$

+ Im{
$$\hat{u}_{n}(\boldsymbol{k},t)$$
} sin($\frac{2\pi}{L}(k_{2}x_{2}+k_{3}x_{3}))$]
+ $\sum_{k_{3}=1}^{k_{\max}} \left[\text{Re}\{\hat{u}_{n}(\boldsymbol{k},t)\} \cos(\frac{2\pi}{L}(k_{3}x_{3})) + \text{Im}\{\hat{u}_{n}(\boldsymbol{k},t)\} \sin(\frac{2\pi}{L}(k_{3}x_{3})) \right]_{k_{1},k_{2}=0}$. (A.5)

A.1. Reproducing an Energy Spectrum

One possible feature of real turbulent flows that can be reproduced using synthetic turbulence is the correct energy spectrum E.

The velocity spectrum tensor is defined as the Fourier transform of the two-point correlation of the velocity field, i.e.

$$\phi_{mn}(\mathbf{k},t) := \langle \hat{u}_m^*(\mathbf{k},t)\hat{u}_n(\mathbf{k},t)\rangle \quad . \tag{A.6}$$

The energy contained in each Fourier mode is $\frac{1}{2}$ the trace of the velocity spectrum tensor. The energy spectrum is obtained as the total energy per Fourier mode, without any directional information, i.e. by summing over the energy of all Fourier modes with wave numbers of the same magnitude $|\mathbf{k}| = k$

$$E(k) = \sum_{\substack{\boldsymbol{k} \in \mathbb{Z}^d \setminus \{\mathbf{0}\}\\ |\boldsymbol{k}| = k}} \frac{1}{2} \phi_{nn}(\boldsymbol{k}, t) .$$
(A.7)

Kolmogorov's hypothesis of local isotropy states that in turbulent flows for sufficiently high Reynolds numbers for any length scales l smaller than some maximum length scale l_{EI} the turbulent motion is isotropic. This is the so-called universal equilibrium range, which is below the energy-containing range of the largest eddies of the flow that are affected by the boundary conditions of the flow. It consists of all wave-vectors \mathbf{k} with $k > l_{EI}^{-1}$. Kolmogorov's first similarity hypothesis then states that in this universal equilibrium range the velocity statistics are uniquely determined by the kinematic viscosity ν of the fluid and the mean dissipation rate of turbulent kinetic energy ϵ (see e.g. Pope, 2008). This universal relationship can be written as

$$E(k) = \epsilon^{2/3} k^{-5/3} \Psi(k\eta) , \qquad (A.8)$$

where η is the Kolmogorov length scale, which is the length scale of the smallest, dissipative eddies in the flow. The part of the universal equilibrium range with $l > \eta$ is generally referred to as the inertial subrange, while the part with $l < \eta$ is called the dissipation subrange. The function $\Psi(k\eta)$ is the compensated Kolmogorov spectrum function. In the inertial subrange, where the second similarity hypothesis states that E(k) is independent of ν , $k\eta$ tends to zero and Ψ becomes constant. This leads to the famous Kolmogorov -5/3 spectrum in the inertial subrange.



Figure A.1.: Energy spectra E(k) in homogeneous turbulence, (a) model spectrum as proposed by Pope (2008), Eq. (A.9) and (b) dissipation range with comparison to other model spectra: 'Pope Exponential' (Eq. (A.11)), 'Kraichnan Spectrum' (Eq. (A.12)) and 'Kraichnan Square' spectrum which is the Kraichnan spectrum from Eq. (A.12), but with the argument of the exponential squared.

Different models for $\Psi(k\eta)$ have been proposed to model the different regimes of turbulent flows. A model spectrum that confirms well to experimental data for all ranges of k can be found in Pope (2008). It reads as

$$E(k) = C\epsilon^{2/3}k^{-5/3}f_L(k)\Psi(k\eta) , \qquad (A.9)$$

where f_L and f_η are non-dimensional functions that model the behavior of E(k) for large scales > L and dissipative scales < η respectively and tend to unity for k in the inertial subrange. The definition of f_L is

$$f_L(k) = \left(\frac{2\pi k}{\left[(2\pi k)^2 + c_L\right]^{1/2}}\right)^{5/3+2} , \qquad (A.10)$$

where c_L is a positive constant, for high Reynolds numbers $c_L \approx 6.78$. The definition of f_η is

$$\Psi(k\eta) = \exp\{-\beta([(2\pi k\eta/L)^4 + c_\eta^4]^{1/4} - c_\eta)\}, \qquad (A.11)$$

where c_{η} is a positive constant that is for high Reynolds numbers $c_{\eta} \approx 0.40$. For the

constant β of the exponential decay several experiments suggest a value of $\beta = 5.2$ (see e.g. Pope, 2008, pg. 233). The qualitative shape of the energy spectrum function (A.9) at high Reynolds numbers can be seen in Fig. A.1(a). It shows both a quadratic increase in the energy containing range and the Kolmogorov -5/3 spectrum in the inertial subrange.

However, for this work generally only the dissipative scales of the flow are relevant since the properties of particles much smaller than the Kolmogorov scale are investigated. It is therefore sufficient to consider the spectral energy function restricted to this subrange. Here, an exponential decay of E(k) is expected. Instead of taking e.g. the full model spectrum (A.9) by Pope, simplifications can be considered. One possibility is to take for the spectrum in the dissipative subrange the shape function $\Psi(k\eta)$, with an appropriate normalization constant, i.e. $E(k) = C\Psi(k\eta)$. For $c_{\eta} = 0$ this can be simplified further to the exponential spectrum suggested by Kraichnan (Martinez et al., 1997)

$$E(k) = C(2\pi k\eta/L)^3 \exp(-\beta [2\pi k\eta/L]) . \qquad (A.12)$$

In some of the literature, e.g. Sigurgeirsson and Stuart (2002), a different version of the Kraichnan spectrum appears, that contains the square of $2\pi k\eta/L$ instead. These three spectra are compared, with appropriate normalizations, to the full model spectrum in Fig. A.1(b). It can be seen that both the exponential spectrum by Pope (A.11) and the simpler Kraichnan spectrum (A.12) lead to very similar approximations. Therefore, in this work the Kraichnan spectrum with $\beta = 5.2$ is used, as it seems to be a simple approximation that is still very close to experimental data. However, different versions of E(k) are of course possible.

The normalization of the energy spectrum E(k) can be done in one of two possible ways. Either, the normalization constant is chosen so that E(k) sums to a desired value of the total energy \mathcal{E} or the constant is chosen so that $2\nu(2\pi k/L)^2 E(k)$ sums to a desired value of the dissipation of turbulent kinetic energy ϵ . Here, the second version is preferred since ϵ is the quantity most often set in experiments. The constant C is then given by

$$C = \epsilon \left[\sum_{k} 2\nu (2\pi k/L)^2 (2\pi k\eta/L)^3 \exp(-\beta [2\pi k\eta/L]) \right]^{-1} .$$
 (A.13)

The question that needs to be answered now is how E(k) is related to the parameters $\alpha(\mathbf{k})$ and $\sigma(\mathbf{k})$ of the stochastic differential equation for $\langle \hat{v}_n \rangle$. To see this the relationship between the variances $\langle \hat{u}_n^*(\mathbf{k},t)\hat{u}_n(\mathbf{k},t)\rangle$ and $\langle \hat{v}_n^*(\mathbf{k},t)\hat{v}_n(\mathbf{k},t)\rangle$ needs to be determined.

From Eq. (A.2) it follows that

By using

where the first line¹ follows from \hat{v}_n and \hat{v}_m being independent random variables with mean zero for $m \neq n$, one obtains

$$\langle \hat{u}_n^*(\boldsymbol{k},t)\hat{u}_n(\boldsymbol{k},t)\rangle = (d-1)\frac{\sigma(\boldsymbol{k})^2}{\alpha(\boldsymbol{k})}$$
 (A.16)

This result can also easily be understood intuitively. The distribution of $\hat{\boldsymbol{v}}(\boldsymbol{k},t)$ is a *d*-dimensional complex-valued Gaussian distribution. By projecting this on a (d-1)-dimensional subspace perpendicular to any vector \boldsymbol{k} , one obtains a (d-1)-dimensional complex-valued Gaussian distribution, which has a variance as given by Eq. (A.16).

To assure statistical isotropy of the flow, it is assumed that $\alpha(\mathbf{k})$ and $\sigma(\mathbf{k})$ only depend on $|\mathbf{k}|$. From Eq. (A.7) it then follows that

$$\frac{\sigma(k)^2}{\alpha(k)} = \frac{2}{(d-1)} \frac{E(k)}{\#(k)} , \qquad (A.17)$$

where

$$\#(k) = \sum_{\substack{\boldsymbol{k} \in \mathbb{Z}^d \setminus \{\mathbf{0}\}\\ |\boldsymbol{k}| = k}} 1 \tag{A.18}$$

denotes the number of \boldsymbol{k} that have the same modulus k.

There are many possible ways to satisfy this condition. One convenient way is to set $\alpha(k) = c$ and $\sigma(k) = \sqrt{c\lambda(k)}$. The constant c is then the correlation time of the flow and $\lambda(k)$ equals the right hand side of Eq. (A.17). With the definition of E(k) from Eq. (A.12) the system can now be solved. This is usually done numerically. In this work an explicit order 1.0 strong scheme (Heun) is used, see e.g. Kloeden and Platen (1999).

¹there is no summation over n on the right hand side of the first line

A.2. Calculating Mean, Variance and Spatial Correlations of the Flow

The mean, variance and spatial correlations of a flow constructed in the manner described in the previous subsection can be calculated from the properties of the individual Fourier modes. For simplicity, only the result for d = 2 is shown, but the same calculations can be done for other values of d. For the case of d = 2 the components of the flow u(x, t)are given by

$$u_{n}(\boldsymbol{x},t) = \sum_{k_{1}=1}^{k_{\max}} \sum_{k_{2}=-k_{\max}}^{k_{\max}} \left[\operatorname{Re}\{\hat{u}_{n}(\boldsymbol{k},t)\} \cos(\frac{2\pi}{L}(k_{1}x_{1}+k_{2}x_{2})) + \operatorname{Im}\{\hat{u}_{n}(\boldsymbol{k},t)\} \sin(\frac{2\pi}{L}(k_{1}x_{1}+k_{2}x_{2}+k_{3}x_{3})) \right] + \sum_{k_{2}=1}^{k_{\max}} \left[\operatorname{Re}\{\hat{u}_{n}(\boldsymbol{k},t)\} \cos(\frac{2\pi}{L}(k_{2}x_{2})) + \operatorname{Im}\{\hat{u}_{n}(\boldsymbol{k},t)\} \sin(\frac{2\pi}{L}(k_{2}x_{2})) \right]_{k_{1}=0}.$$
(A.19)

The ensemble mean $\langle u_n(\boldsymbol{x},t)\rangle$ of Eq. (A.19) is zero, because the mean of $\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}\$ and $\operatorname{Im}\{\hat{u}_n(\boldsymbol{k},t)\}\$ is zero for all \boldsymbol{k} .

The variance $\langle u_n(\boldsymbol{x},t)^2 \rangle$ can also be calculated from Eq. (A.19). Using the independence of the real and imaginary part of $\langle u_n(\boldsymbol{x},t) \rangle$, both for the same and different \boldsymbol{k} , the variance reduces to

$$\langle u_n(\boldsymbol{x},t)^2 \rangle = \sum_{k_1=1}^{k_{\max}} \sum_{k_2=-k_{\max}}^{k_{\max}} \left[\langle \operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)^2\} \rangle \cos^2(\frac{2\pi}{L}(k_1x_1+k_2x_2)) + \langle \operatorname{Im}\{\hat{u}_n(\boldsymbol{k},t)\}^2 \rangle \sin^2(\frac{2\pi}{L}(k_1x_1+k_2x_2+k_3x_3)) \right] + \sum_{k_2=1}^{k_{\max}} \left[\langle \operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}^2 \rangle \cos^2(\frac{2\pi}{L}(k_2x_2)) + \langle \operatorname{Im}\{\hat{u}_n(\boldsymbol{k},t)\}^2 \rangle \sin^2(\frac{2\pi}{L}(k_2x_2)) \right]_{k_1=0} .$$
(A.20)

The problem therefore reduces to finding the variance of the real and imaginary part of the Fourier modes $\hat{u}_n(\mathbf{k}, t)$. From Eq. (A.16) and (A.17) it follows that

$$\langle \operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}\rangle = \frac{E(k)}{\#(k)},$$
 (A.21)

and the same also for the variance of the imaginary part. It should be noted that this is the result for the sum of the variances of all components and the variance of the *n*-th component $\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}$ is not necessarily given by $\frac{1}{d}\langle\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}\rangle$! Due to the projection perpendicular to \boldsymbol{k} , for some values of k some components may be zero while others contain the full variance. However, for the calculation of Eq. (A.20) this does not matter since the sum goes over all values of k. We can therefore use $\langle\operatorname{Re}\{\hat{u}_n(\boldsymbol{k},t)\}^2\rangle = \langle\operatorname{Im}\{\hat{u}_n(\boldsymbol{k},t)\}^2\rangle = \frac{1}{d} \cdot \frac{E(k)}{\#(k)}$ in the sum in Eq. (A.20). This results in

$$\left\langle u_n(\boldsymbol{x},t)^2 \right\rangle = \sum_{k_1=1}^{k_{\max}} \sum_{k_2=-k_{\max}}^{k_{\max}} \left[\frac{1}{d} \cdot \frac{E(k)}{\#(k)} \right] + \sum_{k_2=1}^{k_{\max}} \left[\frac{1}{d} \cdot \frac{E(k)}{\#(k)} \right]_{k_1=0} .$$
(A.22)

This expression can then be evaluated for a given parameter set. For example, for $k_{\text{max}} = 1$, $L = 2\pi$, $\eta = 1$, c = 1 which are the parameters typically used in this work, the variance of each velocity component is approximately $\langle u_n(\boldsymbol{x},t)^2 \rangle \approx 0.1\epsilon$.

The calculation of spatial correlations $\langle u_n(\boldsymbol{x},t)u_n(\boldsymbol{x}+\delta\boldsymbol{e}_m,t)\rangle$ (no summation over n) is somewhat more involved. In particular, due to the projection of the Fourier modes perpendicular to \boldsymbol{k} , the spatial correlations for each component depend on the direction of the unit vector \boldsymbol{e}_m . In d = 2 it is $\langle u_1(\boldsymbol{x},t)u_1(\boldsymbol{x}+\delta\boldsymbol{e}_1,t)\rangle \neq \langle u_2(\boldsymbol{x},t)u_2(\boldsymbol{x}+\delta\boldsymbol{e}_1,t)\rangle$ and $\langle u_1(\boldsymbol{x},t)u_1(\boldsymbol{x}+\delta\boldsymbol{e}_2,t)\rangle \neq \langle u_2(\boldsymbol{x},t)u_2(\boldsymbol{x}+\delta\boldsymbol{e}_1,t)\rangle$, where \boldsymbol{e}_1 is the unit vector in x_1 direction. It should be noted that this does not violate isotropy, because $\langle u_1(\boldsymbol{x},t)u_1(\boldsymbol{x}+\delta\boldsymbol{e}_1,t)\rangle = \langle u_2(\boldsymbol{x},t)u_2(\boldsymbol{x}+\delta\boldsymbol{e}_2,t)\rangle$ and $\langle u_1(\boldsymbol{x},t)u_1(\boldsymbol{x}+\delta\boldsymbol{e}_1,t)\rangle = \langle u_2(\boldsymbol{x},t)u_2(\boldsymbol{x}+\delta\boldsymbol{e}_2,t)\rangle$.

To calculate the spatial correlation $\langle u_n(\boldsymbol{x},t)u_n(\boldsymbol{x}+\delta\boldsymbol{e}_m,t)\rangle$ the product of two sums of the form of the right-hand side of Eq. (A.19) needs to be calculated. For simplicity, we only show the case of $k_{\text{max}} = 1$, where the right hand side of Eq. (A.19) has only four summands. Using that the variance of the real and imaginary part of $\hat{u}_n(\boldsymbol{x},t)$ is the same, one obtains for the *n*-th component for the spatial correlation in \boldsymbol{e}_1 direction (neglecting the dependence on t)

$$\langle u_{n}(\boldsymbol{x})u_{n}(\boldsymbol{x}+\delta\boldsymbol{e}_{1})\rangle = \langle \operatorname{Re}\{\hat{u}_{n}([0,1])\}^{2}\rangle + \langle \operatorname{Re}\{\hat{u}_{n}([1,-1])\}^{2}\rangle \cos(\frac{2\pi}{L}(-x_{1}+x_{2}))\cos(\frac{2\pi}{L}(-x_{1}-\delta+x_{2})) + \langle \operatorname{Re}\{\hat{u}_{n}([1,-1])\}^{2}\rangle \sin(\frac{2\pi}{L}(-x_{1}+x_{2}))\sin(\frac{2\pi}{L}(-x_{1}-\delta+x_{2})) + \langle \operatorname{Re}\{\hat{u}_{n}([1,0])\}^{2}\rangle \cos(\frac{2\pi}{L}x_{1})\cos(\frac{2\pi}{L}(x_{1}+\delta)) + \langle \operatorname{Re}\{\hat{u}_{n}([1,0])\}^{2}\rangle \sin(\frac{2\pi}{L}x_{1})\sin(\frac{2\pi}{L}(x_{1}+\delta))$$
(A.23)

+
$$\langle \operatorname{Re}\{\hat{u}_n([1,1])\}^2 \rangle \cos(\frac{2\pi}{L}(x_1+x_2)) \cos(\frac{2\pi}{L}(x_1+\delta+x_2))$$

+ $\langle \operatorname{Re}\{\hat{u}_n([1,1])\}^2 \rangle \sin(\frac{2\pi}{L}(x_1+x_2)) \sin(\frac{2\pi}{L}(x_1+\delta+x_2))$.

This can be expanded in a Taylor series for small δ , resulting in

$$\langle u_n(\boldsymbol{x})u_n(\boldsymbol{x}+\delta\boldsymbol{e}_1)\rangle = \langle u_n(\boldsymbol{x})^2 \rangle - \frac{\delta^2}{2} \left(\frac{2\pi}{L}\right)^2 \left[\langle \operatorname{Re}\{\hat{u}_n([1,-1])\}^2 \rangle + \langle \operatorname{Re}\{\hat{u}_n([1,1])\}^2 \rangle \right] + \mathcal{O}(\delta^4) \cdot (A.24)$$

In a similar manner the spatial correlation in the e_2 direction can be obtained, resulting in

$$\langle u_n(\boldsymbol{x})u_n(\boldsymbol{x}+\delta\boldsymbol{e}_2)\rangle = \langle u_n(\boldsymbol{x})^2 \rangle - \frac{\delta^2}{2} \left(\frac{2\pi}{L}\right)^2 \left[\langle \operatorname{Re}\{\hat{u}_n([1,-1])\}^2 \rangle + \langle \operatorname{Re}\{\hat{u}_n([0,1])\}^2 \rangle + \langle \operatorname{Re}\{\hat{u}_n([1,1])\}^2 \rangle \right] + \mathcal{O}(\delta^4) .$$
(A.25)

The reason for the dependence of the spatial correlations of the different velocity components on the direction of \boldsymbol{e}_m now becomes apparent. Because of the projection perpendicular to \boldsymbol{k} , $\langle \operatorname{Re}\{\hat{u}_1([1,0])\}^2 \rangle = 0$, whereas $\langle \operatorname{Re}\{\hat{u}_2([1,0])\}^2 \rangle = \frac{E(k)}{\#(k)}$ and similar $\langle \operatorname{Re}\{\hat{u}_1([0,1])\}^2 \rangle = \frac{E(k)}{\#(k)}$, whereas $\langle \operatorname{Re}\{\hat{u}_2([0,1])\}^2 \rangle = 0$. This means that spatial correlations in the direction perpendicular to the direction of the velocity decay faster. With $\langle \operatorname{Re}\{\hat{u}_n([1,-1])\}^2 \rangle = \langle \operatorname{Re}\{\hat{u}_n([1,1])\}^2 \rangle = \frac{1}{d} \cdot \frac{E(k)}{\#(k)}$ the spatial correlations can now be evaluated. For example, for $L = 2\pi$, $\eta = 1$, c = 1 one obtains

$$\langle u_n(\boldsymbol{x})u_n(\boldsymbol{x}+\delta\boldsymbol{e}_m)\rangle = \begin{cases} \epsilon \left(0.1 - 0.05 \cdot \delta^2\right) + \mathcal{O}(\delta^4), & n \neq m \\ \epsilon \left(0.1 - 0.0125 \cdot \delta^2\right) + \mathcal{O}(\delta^4), & n = m \end{cases}$$
(A.26)

Variance of the velocity gradients

With the results from the previous section the variance of the velocity gradients $\frac{\partial u_n}{\partial x_m}$ can be calculated. Again, for simplicity only the case of d = 2 is shown and only the specific result for $k_{\text{max}} = 1$, $L = 2\pi$, $\eta = 1$, c = 1 is calculated.

In this work the velocity gradients are typically estimated as the difference quotient over the distance $\delta = 2r$, where r is the radius of a particle. Therefore,

$$\left\langle \left(\frac{\partial u_n}{\partial x_m}\right)^2 \right\rangle \approx \left\langle \left(\frac{u_n(\boldsymbol{x}+\delta \boldsymbol{e}_m,t)-u_n(\boldsymbol{x},t)}{\delta}\right)^2 \right\rangle$$

$$= \frac{2}{\delta} \left[\left\langle \left(u_n(\boldsymbol{x}, t)^2 \right\rangle - \left\langle \left(u_n(\boldsymbol{x}, t) \, u_n\left(\boldsymbol{x} + \delta \boldsymbol{e}_m, t \right) \right\rangle \right] \right]$$
(A.27)

Using the result for the spatial correlation in Eq. (A.26), one obtains

$$\left\langle \left(\frac{\partial u_n}{\partial x_m}\right)^2 \right\rangle \approx \begin{cases} 0.1\epsilon, & n \neq m\\ 0.025\epsilon, & n = m \end{cases}$$
(A.28)

B. Parametrization of the Discrete Element Aggregate Model

The Table B.1 below lists the parameters used in our implementation of the discrete element aggregate model in Chapter 6. The fourth column contains the parameter values in SI units. The third column contains these values in rescaled units, where time, length and mass are μs , mm and mg, respectively. The second column is the parameter as it is used in the numerical implementation.

Parameter	Value	Rescaled Units	SI Units	
time	1	$1\mu s$	10^{-6} s	
space	1	1mm	10^{-3} m	
mass	1	1mg	10^{-6} kg	
radius	0.004	0.004mm	$4 \cdot 10^{-6} \mathrm{m}$	
density (particle)	2	2mg/mm ³	2000kg/m^3	
density (fluid)	1	$1 \mathrm{mg/mm^3}$	1000kg/m^3	
viscosity (fluid)	10^{-6}	$10^{-6} \mathrm{mg}/(\mathrm{mm} \cdot \mu \mathrm{s})$	10^{-3} kg/(m · s)	
normal forces				
Hamaker constant	10^{-20}	$10^{-20} (mg \cdot mm^2) / \mu s^2$	$10^{-20} \rm kgm^2/s^2$	
δ_{min}	$4 \cdot 10^{-7}$	$4 \cdot 10^{-7} \mathrm{mm}$	$4 \cdot 10^{-10} \mathrm{m}$	
δ_{max}	$1.1 \cdot (r_1 + r_2)$			
reloading stiffness k	5	$5 mg/\mu s^2$	$5 \cdot 10^6 \mathrm{kg/s^2}$	
loading stiffness	$0.5 \cdot k$			
adhesion	$0.5 \cdot k$			
plastic range	0.05			
visc. Dissipation γ	$5 \cdot 10^{-5}$	$5 \cdot 10^{-5} \mathrm{mg}/\mu \mathrm{s}$	$5 \cdot 10^{-5} \mathrm{kg/s}$	
tangent forces				
sliding stiffness	$0.2 \cdot k$			
rolling stiffness	$0.1 \cdot k$			
Continued on next page				

Table B.1.: Microscopic parameters used for the discrete-element aggregate model. See Luding (2008) for a detailed description of the meaning of each parameter.

Parameter	Value	Rescaled Units	SI Units
torsion stiffness	$0.1 \cdot k$		
static friction (sliding)	1		
dyn. friction ratio	1		
rolling friction	0.1		
torison friction	0.1		
sliding visc. dissipation	$0.2 \cdot \gamma$		
rolling visc. dissipation	$0.05 \cdot \gamma$		
torsion visc. dissipation	$0.05 \cdot \gamma$		

Table B.1 – continued from previous page

C. Additional Results on Inertial Particles in Chaotic Advection

The following pages contain our work on aggregation/coagulation of inertial particles in chaotic advection. These works are not included in the main part of this thesis because there we focused on a slightly different question, namely (synthetic) turbulent flows. However, we include the full publications on this topic for the sake of completeness. The following publications are included in this Appendix:

 Zahnow, J. C., Vilela, R. D., Feudel, U., and Tél, T. (2008). "Aggregation and fragmentation dynamics of inertial particles in chaotic flows." *Physical Review E*, 77(5):055301.

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• Zahnow, J. C., Vilela, R. D., Feudel, U., and Tél, T. (2009). "Coagulation and fragmentation dynamics of inertial particles." *Physical Review E*, **80(2)**:026311. This work is under copyright of The American Physical Society (2009), included with permission.

In these works we applied the individual, inertial-particle based model approach for aggregation and fragmentation, discussed in Chapter 3 and 4 of this thesis, to the problem of spherical particles in chaotic advection and show how our model is able to capture the basic properties of aggregation and fragmentation processes.

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Aggregation and fragmentation dynamics of inertial particles in chaotic flows

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Inertial particles advected in chaotic flows often accumulate in strange attractors. While moving in these fractal sets they usually approach each other and collide. Here we consider inertial particles aggregating upon collision. The new particles formed in this process are larger and follow the equation of motion with a new parameter. These particles can in turn fragment when they reach a certain size or shear forces become sufficiently large. The resulting system consists of a large set of coexisting dynamical systems with a varying number of particles. We find that the combination of aggregation and fragmentation leads to an asymptotic steady state. The asymptotic particle size distribution depends on the mechanism of fragmentation. The size distributions resulting from this model are consistent with those found in raindrop statistics and in stirring tank experiments.

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There is an increasing recent interest in the advection of inertial particles in fluid flows [1]. This comes in part from the fact that the dynamics of these particles are dissipative, which leads in most flows to a preferential accumulation on chaotic, fractal attractors. Previous studies concentrated mainly on noninteracting particles, in spite of the fact that accumulation leads unavoidably to strong mutual interactions of different kinds.

Here we consider the interaction of these particles in the form of aggregation and fragmentation. When two particles come sufficiently close, they aggregate to a larger one, observing mass and momentum conservation. If the size exceeds a certain threshold value, which depends on a property, the stickiness γ of the particles, or other conditions are fulfilled, they break up into smaller pieces. This is the basic mechanism underlying such processes in nature like raindrop formation in clouds [2] or the sedimentation of marine aggregates [3] in the ocean. We demonstrate that for the study of such processes a particle based approach may be a useful addition to the usual population-balance equation approach [4]. The latter is based on the assumption of well-mixed particles while our approach takes the incomplete mixing of inertial particles in fluids explicitly into account.

Although concepts of dynamical systems theory can usefully be applied, we show that the entire dynamics is much more complex than that of any usual dynamical system. The dynamics of particles of any size are governed by the same type of equations of motion, but with different parameters since new particles will have new radii. Even if one considers a finite number *n* of possible sizes (size classes), there are n equations of motion with different size-dependent parameters. We thus have a union of n dynamical systems and, moreover, the number of particles in each size class is changing in time. It is useful to interpret the attractors of the different size classes (of the noninteracting problem) as the skeleton of the full dynamics. Aggregation and fragmentation generates transitions from one attractor to another one. It is this permanent wandering among different attractors which characterizes the new dynamics.

We show that the combination of aggregation and frag-

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and this steady state is unique for the cases studied here. We find that the dynamics and the steady state depend on the fragmentation rule. For fragmentation due to shear we present a simple scaling relationship for the asymptotic average size of the particles. Furthermore, the shape of the asymptotic size distribution can be represented in a scaled form independent of the stickiness γ . For simplicity we consider spherical aerosoles, i.e., par-

mentation, superimposed on chaotic inertial advection dy-

namics, leads to a convergence to an asymptotic steady state,

For simplicity we consider spherical aerosoles, i.e., particles much denser than the ambient fluid, and assume that the difference between their velocity $\dot{\mathbf{r}}$ and the fluid velocity $\mathbf{u}=\mathbf{u}(\mathbf{r}(t), t)$ at the same position is sufficiently small so that the drag force is proportional to this difference (Stokes drag). The dimensionless form of the governing equation for the path $\mathbf{r}(t)$ of such aerosols subjected to drag and gravity, reads as [5]:

$$\ddot{\mathbf{r}} = A(\mathbf{u} - \dot{\mathbf{r}} - W\mathbf{n}),\tag{1}$$

where **n** is a unit vector pointing upwards in the vertical direction. Throughout this paper we consider the vertical direction along the axis y. The inertia parameter A (larger values for smaller particle size) can be written in terms of the densities ρ_p and ρ_f of the aerosol and of the fluid, respectively, the radius a of the aerosols, the fluids kinematic viscosity ν , and the characteristic length L and velocity U of the flow. It is A=R/St, where $R=\rho_f/\rho_p \ll 1$ is the density ratio and $St=(2a^2U)/(9\nu L)$ is the so-called Stokes number of the aerosol [8]. $W=2a^2\rho_p g/(9\nu\rho_f U)$ is the dimensionless settling velocity in a medium at rest.

Every particle produces perturbations in the flow that decay inversely proportional to the distance from the particle [6]. Here we assume a dilute regime, where the local concentration of particles is low enough, so that particle-particle interaction can be neglected [7].

During aggregation and fragmentation the radius of particles changes and so do the parameters A and W. The smallest (*primary*) particles considered in this model have dimensionless radius $a_1=5/30^{1/3} \times 10^{-5}$, mass $m_1=\rho_p 4/3\pi a_1^3$,

ZAHNOW et al.

inertia parameter A_1 =7, and settling velocity W_1 =0.4/ A_1 . All larger particles are assumed to consist of an integer number of these primary particles. Thirty different size classes are considered. A particle that consists of α (α =1,...,30 is called *size class index*) primary particles has a radius $a_{\alpha} = \alpha^{1/3}a_1$, an inertia parameter $A_{\alpha} = (a_1/a_{\alpha})^2A_1 = \alpha^{-2/3}A_1$ and a settling velocity $W_{\alpha} = \alpha^{2/3}W_1$. The largest particle therefore has a radius $a_{30} = 5 \times 10^{-5}$.

Aggregation takes place upon collision, i.e., if two particles, say of radius a_i and a_j , come closer than a threshold. Mass conservation requires the radius of the new particle to be $a_{new}^3 = a_i^3 + a_j^3$. For the size class index this implies a linear rule: $\alpha_{new} = \alpha_i + \alpha_j$, which determines the new inertia parameter via $A_{\alpha_{new}} = \alpha_{new}^{-2/3} A_1$. The velocity of the new particle follows from momentum conservation.

Fragmentation. We apply one of the following rules: (i) *Size-limiting fragmentation*: If a particle becomes larger than the maximum radius a_{30} , it is broken up into two smaller fragments whose radii are chosen randomly, with a uniform distribution between a_1 and half the original radius. If any fragment is larger than a_{30} this process is repeated, until no fragment exceeds a_{30} . (ii) *Shear fragmentation* takes place if the velocity gradient is too large. More specifically, the velocity gradient is evaluated across each particle both in the horizontal and in the vertical direction. If the maximum in any direction exceeds a threshold value, the particle is broken up into two smaller parts in the same way as for size-limiting fragmentation. While size-limiting fragmentation is dominant for raindrops [2], shear fragmentation determines the breakup of marine aggregates [9].

Since for marine aggregates the threshold gradient becomes smaller for larger particles [9], we write

$$(\operatorname{grad}(\mathbf{u}))_{th} = \gamma a_1 / a = \gamma \alpha^{-1/3}.$$
 (2)

Coefficient γ represents the "stickiness" of the particles. Whatever rule is taken, the result is the reversed process of aggregation: two new particles are formed from an old one with the size class indices: $\alpha_{i,new} + \alpha_{j,new} = \alpha_{old}$. The centers of the new particles are placed along a line segment in a random direction so that their distance equals the sum of their radii. Momentum is conserved. For simplicity we assume that the new particles have the same velocity as the old one. Shear fragmentation is applied together with size-limiting fragmentation to keep the maximum number of occurring size classes at 30.

At the instant of both the aggregation and the fragmentation process there is a sudden change in the dynamics: the number of particles jumps in three among the 30 available dynamical systems defined by the size classes.

For convenience, we treat the case where the fluid flow is two dimensional, therefore the phase space of the advection dynamics is four dimensional. We use the convection model of [10] with dimensionless velocity field

$$\mathbf{u}(x,y,t) = \left[1 + k\sin(\omega t)\right] \begin{pmatrix} \sin(2\pi x)\cos(2\pi y) \\ -\cos(2\pi x)\sin(2\pi y) \end{pmatrix}, \quad (3)$$

where k=2.72 is the amplitude and $\omega=\pi$ is the frequency of the periodic forcing. The fluid flow itself is laminar, but the

PHYSICAL REVIEW E 77, 055301(R) (2008)

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FIG. 1. Poincaré section of the attractors of Eq. (1) projected onto the plane of the flow for inertia parameters (a) A=7 (size class 1), (b) A=2.778 (size class 16), and (c) A=2.253 (size class 30). The positive Lyapunov exponents are (a) $\lambda_1=0.108$, (b) $\lambda_1=0.061$, and (c) $\lambda_1=0.119$, $\lambda_2=0.014$. The settling velocity is W=0.4/A.

dynamics of the inertial particles can be chaotic. Because of the spatial periodicity of the flow and the resulting spatial periodicity of the attractors the total particle mass M in each 1×1 unit cell remains the same over time. The dynamics can therefore be restricted to one cell. The characteristic size and velocity of the flow are therefore L=1, U=1, respectively.

In the numerical realization of the problem the particles are advected without any interaction over a time interval δt =T/20 at the end of which first aggregation and then fragmentation take place, instantly. This is repeated after every time step δt . To carry out the aggregation process, the distance between particles is calculated and all particles within a distance less than the sum of their radii aggregate.

As an initial condition we take 10^5 particles in the smallest size class and no particles in other size classes. Furthermore, particles are uniformly distributed over the entire configuration space with velocities matching that of the fluid. This choice fixes the total mass of the system to be $M = 10^5 m_1$.

Before presenting the results obtained for the full dynamics, it is instructive to see the attractors of the noninteracting problem. Figure 1 presents the attractors for the smallest, an intermediate, and the largest size classes. The extension of the attractor seems to grow almost monotonically with the size class index, except for a few intermediate size classes ($\alpha = 9, ..., 14$), where the attractor size decreases or the attractor becomes periodic.

In order to understand the full dynamics, we include first the simplest fragmentation process, the size-limiting fragmentation. Figure 2(a) shows the time dependence of the number $N_{\alpha}(t)$ of particles in a few size classes. The particles leave the initial size class very quickly. After 20 time units nearly all other size classes are considerably occupied. In fact, the population in size class 16 reaches a maximum here, but decreases again later on. It is the occupation of the largest size classes which continuously increases and then saturates. The total number N(t) of particles (bold line) rapidly decreases first, but saturates later on. The spatial distribution of particles [Figs. 2(b) and 2(c)] shows that they move initially among the more localized attractors characteristic of small size class indices. Later, the distribution becomes more



AGGREGATION AND FRAGMENTATION DYNAMICS OF ...

FIG. 2. Particle numbers vs time, and space distributions for size-limiting fragmentation. (a) Total number N(t) of particles (bold—left axis) and the number of particles $N_{\alpha}(t)$ in size class α (gray) for $\alpha=1$ (left axis), $\alpha=16$ (right axis), and $\alpha=30$ (right axis). Distribution of all particles in configuration space at time (b) t=5, and (c) t=100.

extended in configuration space when size classes with extended attractors become well occupied, although the total number of particles is much less than in the initial phase. While the full dynamics is dominated by transients in between attractors, the shape of the backbone attractors is clearly recognizable in the plots.

To follow the convergence towards an asymptotic state, we found it useful to consider the average size class index $\langle \alpha(t) \rangle = \sum_{i=1}^{30} \alpha_i N_{\alpha_i}(t) / N(t)$. Figure 3(a) shows the time dependence of this index for both types of fragmentation. It illustrates the convergence to an asymptotic steady state for both fragmentation rules. Initially, aggregation leads to a fast increase in the average particle size class for both fragmentation rules. Then fragmentation is reached, with a different asymptotic average particle size $\alpha_{\infty} = \lim_{t \to \infty} \langle \alpha(t) \rangle$ for the



FIG. 3. (a) Average size class index $\langle \alpha(t) \rangle$ vs time for sizelimiting fragmentation (upper curve) and shear fragmentation with γ =9 (lower curve). (b) Asymptotic average size class index for the same initial particle distribution as a function of the stickiness parameter γ . Squares, numerical results; continuous line, fit $\alpha_{\infty}(\gamma)$ by a linear function of γ^3 , based on the data for $\gamma < 10$.



PHYSICAL REVIEW E 77, 055301(R) (2008)

FIG. 4. Histogram of the particle size distribution. (a) Size class percentage N_{α}/N in the steady state vs the dimensionless radius *a*, for size limiting and shear fragmentation with $\gamma=9$. Results for fragmentation into three parts is also shown (open markers). (b) Normalized number of particles versus the relative radius $a/\langle a \rangle$ for different values of γ (binary splitting).

two rules. For size-limiting fragmentation the value of α_{∞} is almost constant over time, while for shear fragmentation α_{∞} oscillates with the period *T* of the flow. This is caused by the periodic change in the fluid flow and the corresponding change in the shear forces.

For size-limiting fragmentation, α_{∞} is, in a broad range, independent of M. For shear fragmentation with $M < 3 \times 10^5 m_1$, $\alpha_{\infty}(M)$ increases approximately linearly with M, while for higher values a saturation of $\alpha_{\infty}(M)$ sets in, which is due to size-limiting fragmentation.

By considering other initial conditions than those mentioned above, while keeping the total mass M fixed, the asymptotic state is found for both rules to be independent of the chosen initial condition, but for shear fragmentation the asymptotic state does depend on the value of the stickiness γ .

To illustrate this dependence of the steady state on the stickiness γ , Fig. 3(b) shows how α_{∞} changes with the stickiness parameter at a fixed *M*. A drastic increase of $\alpha(\gamma)$ can be observed in the interval $4 < \gamma < 10$. It is clear that α_{∞} increases with γ , because particles become more resistant to shear. A quantitative estimate of the shape of this $\alpha_{\infty}(\gamma)$ curve can be derived by assuming that the threshold velocity gradient is approximately constant for the size class index α_{∞} . From Eq. (2) it then follows that α_{∞} depends linearly on γ^3 . This simple dependence is expected to hold for relatively small values of γ and α_{∞} , where shear fragmentation dominates. It can be seen that for higher values of γ , when size-limiting fragmentation becomes important the $\alpha_{\infty}(\gamma)$ curve deviates from this estimate and converges towards a limiting value $\alpha_{\infty}^{(lim)}$ [Fig. 3(b)].

In addition to the average quantities it is natural to investigate the occupation of the different size classes in the steady state. Figure 4(a) shows the steady-state histograms vs the dimensionless radius. For size-limiting fragmentation the distribution shows one broad peak around smaller size classes and a second, smaller peak at large size classes with a sharp drop-off towards zero beyond the maximum size. This behavior, with two maxima and a sudden drop after the second peak, is similar to that of observed cloud drop spectra

RAPID COMMUNICATIONS
[2]. For shear fragmentation the steady-state distribution also shows two peaks, but much closer together, with a long tail in the particle distribution towards larger sizes that goes smoothly towards zero. For $\gamma < 5$ this distribution is not fully developed and only shows one peak. In the intermediate γ range, where the distribution is fully developed, but sizelimiting fragmentation is not important, a scaling form $N_{\alpha}/\max(N_{\alpha}) = f(a/\langle a \rangle)$ is found ($\langle a \rangle$ represents the average radius), independently of γ . All distributions in this range collapse then onto a single master curve as shown in Fig. 4(b). This behavior, along with the long tail in the distribution towards the right-hand side, is typically observed in shear-fragmentation experiments in stirring tanks [11].

We note that our findings are robust with respect to the number of new particles formed by fragmentation. For instance, in Fig. 4(a) we see that the distributions of particles for ternary fragmentation are similar to the ones for binary splitting and only show a slight shift towards smaller size classes. The same result is found in population balance equation models, e.g., [11].

Finally, we mention that in spite of the different steady states, the size distribution in the initial phase is similar in the different cases. After short times, we find a roughly exponential decay. In this early phase, fragmentation is yet inactive, and the process is dominated by aggregation. This

RAPID COMMUNICATIONS

PHYSICAL REVIEW E 77, 055301(R) (2008)

decay in the short time distribution can be found for all initial conditions.

In conclusion, we illustrated that an individual modeling of particles is able to reflect typical properties of aggregation and fragmentation processes. We found the development of a balance between aggregation and fragmentation, and a steady state. The steady-state particle size distributions found here correspond to those observed in raindrops (size-limiting fragmentation) and stirring tank experiments (shear fragmentation). For shear fragmentation the size distributions are found to follow a scaled form. In addition, the approach shown here can reflect spatial inhomogeneity and take actual particle dynamics into account, and could possibly allow for a much more detailed description of particle interaction. It is thus more adequate than the usual stochastic, mean-field-like approach which relies on the assumption that the particles are well mixed [2]. The presence of chaotic attractors can ensure a partial mixing only, and hence the assumption is not valid. An interesting open problem is to extend our study to three-dimensional (3D) flows.

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- A. Babiano, J. H. E. Cartwright, O. Piro, and A. Provenzale, Phys. Rev. Lett. 84, 5764 (2000); T. Nishikawa, Z. Toroczkai, and C. Grebogi, *ibid.* 87, 038301 (2001); J. Bec, Phys. Fluids 15, L81 (2003); 17, 073301 (2005); R. D. Vilela and A. E. Motter, Phys. Rev. Lett. 99, 264101 (2007); J. C. Zahnow and U. Feudel, Phys. Rev. E 77, 026215 (2008).
- [2] H. R. Pruppacher and J. D. Klett, *Microphysics of Clouds and Precipitation* (Kluwer Academic, Dordrecht, 1997).
- [3] T. Kiorboe and G. A. Jackson, Limnol. Oceanogr. 46, 1309 (2001).
- [4] D. N. Thomas et al., Water Res. 33, 1579 (1999).
- [5] M. R. Maxey and J. J. Riley, Phys. Fluids 26, 883 (1983); T.
 R. Auton *et al.*, J. Fluid Mech. 197, 241 (1988); E. E.
 Michaelides, J. Fluids Eng. 119, 233 (1997).

- [6] J. Happel and H. Brenner, Low Reynolds Number Hydrodynamics (Martinus Nijhoff, The Hague, 1983).
- [7] For the cases studied here, typical distances between particles are $\geq 50a_1$.
- [8] For raindrops, typical numbers are $R \approx 10^{-3}$ and $a \approx 1$ mm. For advection of these drops in a convective cloud cell of linear size $L \approx 100$ m and typical velocity fluctuations $U \approx 1$ m/s, we obtain St $\approx 2 \times 10^{-4}$ and $A \approx 5$, since $\nu \approx 10^{-5}$ m²/s for air.
- [9] J. C. Flesch et al., AIChE J. 45, 1114 (1999).
- [10] S. Chandrasekhar, Hydrodynamic and Hydromagnetic Stability (Oxford University Press, New York, 1961); T. Nishikawa, Phys. Rev. E 65, 026216 (2002).
- [11] P. T. Spicer and S. E. Pratsinis, AIChE J. 42, 1612 (1996).

PHYSICAL REVIEW E 80, 026311 (2009)

Coagulation and fragmentation dynamics of inertial particles

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Inertial particles suspended in many natural and industrial flows undergo coagulation upon collisions and fragmentation if their size becomes too large or if they experience large shear. Here we study this coagulation-fragmentation process in time-periodic incompressible flows. We find that this process approaches an asymptotic dynamical steady state where the average number of particles of each size is roughly constant. We compare the steady-state size distributions corresponding to two fragmentation mechanisms and for different flows and find that the steady state is mostly independent of the coagulation process. While collision rates determine the transient behavior, fragmentation determines the steady state. For example, for fragmentation due to shear, flows that have very different local particle concentrations can result in similar particle size distributions if the temporal or spatial variation in shear forces is similar.

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I. INTRODUCTION

The dynamics of inertial particles in fluid flows plays an important role in many natural and industrial contexts and has received an increasing interest in recent years. Questions where inertial particles play an important role are ubiquitous in biology, chemistry, oceanography, astrophysics, and geophysics. Recent works from dynamical systems [1-8] to atmospheric science [9–11] and turbulence [12–14] have added greatly to the understanding of these phenomena. Almost all these works have been devoted to the dynamics of noninteracting inertial particles. A major reason for this is that this problem is already very rich, displaying features yet to be understood in their full complexity, as inhomogeneous spatial distributions [15,16] and multivalued velocity fields [12,17,18]. Interestingly, these very same features yield an increased rate of collisions [13], the consequences of which are in most cases not explicitly taken into account. Typically one assumes a *dilute regime* and fully neglects the collisions. In some other works, one keeps track of the collisions numerically without actually addressing the outcome of such events (ghost collisions) [13,19]. To our knowledge, only very recent works have addressed effects of collisions on the dynamics of inertial particles [20–22]. In Ref. [21], we have reported our first results on the dynamics of inertial particles coagulating [23] upon collisions and fragmenting under certain conditions. In Ref. [22], the authors considered elastic collisions in a monodisperse system and pointed out the existence of bursts in the spread of the particles out of the attractors of the purely advective dynamics. In Ref. [20] the authors treated coagulation and shear fragmentation of dust particles in an astrophysical context. There small dust particles can grow into larger fractal clusters due to turbulent collisions.

In this paper we extend the work of Ref. [21] to different flows and to a broadened parameter set. Our motivation lies primarily on natural phenomena such as the collisional growth of cloud droplets [24], sediments in lakes and rivers,

1539-3755/2009/80(2)/026311(8)

and marine snow in the ocean [25]. Here we focus on the description of spherical droplets, i.e., we do not take into account any fractal structures that often appear in sediments or marine snow.

Our main result is that coagulation and fragmentation dominate the behavior of different time spans of the process and fragmentation rather than coagulation is the dominating process for the steady-state size distribution. For different flows, the collision rates between inertial particles can be very different, leading to great changes in the coagulation of particles. While this might be an important effect for transient processes, such as the initiation of rain in clouds, it will turn out that for the steady state fragmentation plays a much greater role. In fact, the steady-state size distributions are mainly determined by the fragmentation process. The specific flow structure is only relevant for the steady-state size distribution when it directly affects the fragmentation process. This can for example be the case for fragmentation due to shear when the spatial and temporal variations in the shear are very different for the two flows.

We consider fragmentation to be of two possible origins. First, particles break up if their size exceeds a certain maximum allowed size. This is motivated by the hydrodynamical instability of large water drops (e.g., cloud drops) settling due to gravity [26]. Second, particles fragment if the shear forces due to the fluid flow are sufficiently large. This mechanism has been reported to be the dominant one in the case of marine aggregates [27].

At a first glance, one might be tempted to pursue a fieldtheoretical approach, in the framework of which one treats the problem of particle motion as a multiphase flow and then applies the Smoluchowski equation [28] to model coagulation and fragmentation for the particle distribution. However, the inertial particle dynamics is dissipative and contracts to an attractor in a 2*d*-dimensional phase space, where *d* is the spatial dimension of the flow. This attractor can be folded in phase space, meaning that the particle velocity may take on several values even at the same location. Due to the presence of such "caustics" [12,18,29], a field-theoretical approach

cannot be well founded. Therefore a study based on an individual tracking of the particles, as the one presented here, becomes necessary.

Here we consider the fluid flow to be spatially smooth and to have a single macroscopic time scale. We are motivated by flows having coherent (e.g., convective) structures on length scales much larger than the ones at which turbulence plays a major role. The effect of turbulence can then be taken into account as a stochastic perturbation described by an eddy diffusivity [30] at small scales. For simplicity, we neglect this small scale noise in the present work and focus only on the large scale motion of the fluid.

We study the dynamics of the system formed by the transported inertial particles undergoing coagulation and fragmentation in three different fluid flows, as described in Sec. II. We find that the system tends to approach a steady state where several size classes coexist (Sec. III). The average number of particles in each size class is roughly constant with a mild periodic time dependence-with a period identical to the one of the advecting fluid flow. The distribution of particles as well as the mean average size in the steady state depends on the type of fragmentation mechanism taking place. First, when fragmentation occurs solely due to particles exceeding a maximum allowed size, the distribution is in general quite broad. Second, for fragmentation occurring also under sufficiently large shear, the distributions typically decay exponentially beyond a certain size class. The distributions depend on the fluid flow for both types of fragmentation. However, for shear fragmentation the differences are very small as long as the variation in the fluid shear over time is qualitatively similar.

In the case of shear fragmentation, we derive a scaling relation for the average size class in the steady state as a function of the coagulate strength parameter γ . Finally, we show that our results are robust with respect to the total mass of particles, the number of allowed size classes and the initial particle size distribution. Also, in the case of shear fragmentation the size distribution in the steady state has a scaled functional form which does not depend on the coagulate strength γ .

II. COAGULATION AND FRAGMENTATION MODEL

A. Dynamics of inertial particles

First, we present the equations of motion for the motion of finite-size particles that will be used here. For simplicity we consider heavy spherical aerosols, i.e., particles much denser than the ambient fluid and assume that the difference between their velocity $\dot{\mathbf{x}}$ and the fluid velocity $\mathbf{u}=\mathbf{u}(\mathbf{x}(t),t)$ at the same position is sufficiently small so that the drag force is proportional to this difference (Stokes drag). The dimensionless form of the governing equation for the path $\mathbf{x}(t)=(x_1(t),x_2(t))$ of the center of mass for such heavy aerosols subjected to drag and gravity reads in this case as [31–33]

$$\ddot{\mathbf{x}} = \frac{1}{\tau} [\mathbf{u}(\mathbf{x}(t), t) - \dot{\mathbf{x}} - W\mathbf{n}], \tag{1}$$

where \mathbf{n} is a unit vector pointing upwards in the vertical direction. Throughout this paper we consider the vertical di-

rection along the axis x_2 . Under the assumption that the density ratio $\rho_f/\rho_p \ll 1$, the particle response time τ can be written in terms of the density ρ_p of the particle, the radius *r* of the aerosols, the fluids dynamic viscosity η , and the characteristic length *L* and characteristic velocity *U* of the flow as $\tau = (2r^2\rho_p U)/(9\eta L)$. We note that the response time τ is nothing but the Stokes number, which can be written in our case as $\tau = \tau_p/T$, where τ_p is the particle's dimensional Stokesian relaxation time and *T* is the characteristic time of the flow. The dimensionless settling velocity in a medium at rest is given by $W = (2r^2\rho_p g)/(9\eta U)$. Note that W/τ is independent of the particle radius *r*.

Every particle produces perturbations in the flow that decay at least inversely proportional to the distance from the particle [34,35]. Here we assume a dilute regime, where the local concentration of particles is low enough, so that particle-particle interaction can be neglected [36] unless particles come into direct contact.

The assumption that the particle radii a are small also means that the feedback from the particle motion on the flow will be small as well [33] and is therefore neglected in the following.

B. Coagulation

Second, we present a model for the coagulation of finitesize particles.

The smallest particles considered will be called primary particles. These primary particles can combine to form larger particles, called coagulates. Coagulation takes place upon collision. All particles are assumed to consist of an integer number of these primary particles, i.e., the primary particles can never be broken up. The number α of primary particles in a coagulate is called the *size class index*. We consider ndifferent size classes, i.e., coagulates can consist of a maximum of *n* primary particles. A coagulate of size class α has a radius $r_{\alpha} = \alpha^{1/3} r_1$, where r_1 is the radius of the primary particles. The response time is $\tau_{\alpha} = (r_{\alpha}/r_1)^2 \tau_1 = \alpha^{2/3} \tau_1$ and the settling velocity in still fluid is $W_{\alpha} = \alpha^{2/3} W_1$. Here τ_1 and W_1 are the response time and the settling velocity for the primary particles, respectively. The largest coagulates therefore have a radius $r_n = n^{1/3}r_1$. We note that particles of different sizes have different parameters τ_{α} and W_{α} and therefore follow the flow with different parameters in the equation of motion [Eq. (1)].

We define a collision of two particles if the centers of the particles, say, of radius r_i and r_j , come closer than a distance $d=r_i+r_j$. In that case the particles coagulate and form a larger particle. Mass conservation requires the radius of the new particle to be $r_{new}^3 = r_i^3 + r_j^3$. For the size class index this implies a linear rule, $\alpha_{new} = \alpha_i + \alpha_j$, which determines the new response time and settling velocity via $\tau_{\alpha_{new}} = \alpha_{new}^{2/3} \tau_1$ and $W_{\alpha_{new}} = \alpha_{new}^{2/3} W_1$, respectively.

The velocity of the new particle follows from momentum conservation. The position of the new coagulate is the center of gravity of the two old particles.

C. Fragmentation

Third, we present a model for the fragmentation of particles. Primary particles cannot be broken up. In the follow-

COAGULATION AND FRAGMENTATION DYNAMICS OF ...

ing, we will compare two different fragmentation rules.

(i) Size-limiting fragmentation. If a particle becomes larger than the maximum radius a_n , it is broken up into two smaller fragments (binary fragmentation) whose radii are chosen randomly, from a uniform distribution between a_1 and half the original radius. If any fragment is larger than a_n this process is repeated, until no fragment exceeds a_n .

(ii) Shear fragmentation takes place when the hydrodynamical force F_{hyd} acting on the particle exceeds the forces F_{coag} holding the coagulate together by a certain factor. The criterion for breakup can therefore be expressed as

$$F_{hyd}/F_{coag} > \tilde{\gamma},$$
 (2)

where $\tilde{\gamma}$ is a constant.

The hydrodynamical force in this case is proportional to the local velocity gradients in the flow. It is expected that larger particles are more likely to break up, therefore, the critical force required for fragmentation should decrease with the coagulate size. For liquid spherical particles (drops) in the size range where viscous forces dominate, Taylor [37] and later Delichatsios [38] derived an expression for the critical velocity difference Δu across the drop required for breakup. Under the condition that the characteristic time of drop deformations is small compared to the time where this velocity gradient occurs, we rewrite this condition with a single parameter as

$$\frac{\Delta u}{a_{\alpha}} = \gamma \left(\frac{r_1}{r_{\alpha}}\right) = \gamma \alpha^{-1/3}, \qquad (3)$$

where γ is a constant, the coagulate strength parameter (the same quantity is called stickiness in [21]). The radius has been normalized with the radius of a primary particle. If the maximum velocity difference across the radius of the drop exceeds the threshold value given by Eq. (3), the particle is broken up into two smaller fragments (binary fragmentation) in the same way as for size-limiting fragmentation.

At the instant of both coagulation and fragmentation there is a sudden change in the dynamics: the number of particles changes in 2 or 3 among the n available dynamical systems defined by the size classes.

D. Fluid flows

For convenience, we treat the case where the fluid flow depends only on two coordinates, i.e., we study a threedimensional flow where the velocity in the third direction is negligible compared to the other two velocities. This can then be represented by a two-dimensional flow. Therefore, the phase space of the particles dynamics is four dimensional. We choose three simple paradigmatic flow situations with different characteristics to indicate the generality of our results.

All flow domains are spatially periodic, with a characteristic length *L*. More specifically, the flows are (a) a convection cell flow with moving vortex centers (in the following referred to as the *moving convection flow*), (b) a convection cell flow with fixed vortices (referred to as the *fixed convection flow*), and (c) a sinusoidal shear flow.

PHYSICAL REVIEW E 80, 026311 (2009)

The two convection cell flows (a) and (b) consist of a regular pattern of vortices, or roll cells. Flow (b) was first introduced by Chandrasekhar [39] as a solution to the Rayleigh-Bénard problem and since then it has been used in the context of different theoretical studies [1,40-42]. The moving convection flow (a) is a slightly modified version, with moving vortex centers, to yield a more realistic chaotic regime for the particle motion. Convection flows are chosen because they contain vortices (convection cells) and uprising/sinking regions, which are characteristic features of realistic flows often found in nature. The flows are defined by the velocity field as the following:

(a) Moving convection flow

$$\mathbf{u}(x_1, x_2, t) = [1 + k_1 \sin(\omega_1 t)] \begin{pmatrix} \sin(2\pi \hat{x}_1) \cos(2\pi \hat{x}_2) \\ -\cos(2\pi \hat{x}_1) \sin(2\pi \hat{x}_2) \end{pmatrix},$$
(4)

where $\hat{x}_1 = x_1 + k_2 \sin(\omega_2 t)$ and $\hat{x}_2 = x_2 + k_2 \cos(\omega_2 t)$. The parameters $k_1 = 2.72$ and $\omega_1 = \pi$ are the amplitude and the frequency of the periodic forcing of the flow, respectively. $k_2 = 1/(2\pi)$ and $\omega_2 = \pi/4$ determine the amplitude and the frequency of the periodic motion of the centers of the vortices in the flow. The period of the flow is T=2 and the characteristic length and characteristic velocity are L=1 and U=1.

(b) Fixed convection flow with the same equation for the flow as in (a), but with $k_2=0$.

The sinusoidal shear flow (c) consists of alternating horizontal and vertical velocity components, where each velocity component consists of two plateaus in time. It was introduced in Refs. [43,44] and has been used many times in chaotic advection studies. Here we consider a timecontinuous version (see [45]) defined by

(a) sinusoidal shear flow

$$\mathbf{u}(x_1, x_2, t) = 0.5 \begin{pmatrix} \{1 + \tanh[\beta \sin(2\pi t)]\}\sin(2\pi x_2) \\ \{1 - \tanh[\beta \sin(2\pi t)]\}\sin(2\pi x_1) \end{pmatrix}, \quad (5)$$

where the parameter β describes how rapidly the transition between two values, a zero and a nonzero velocity, takes place for each velocity component. The typically used value $\beta = 20/\pi$ corresponds to a very rapid transition.

The period of the flow is T=1 and the characteristic length and characteristic velocity are L=1 and U=1.

The fluid flows are laminar and time periodic, but the dynamics of the inertial particles moving in these flows can be chaotic.

To emphasize the difference between the flows, Fig. 1 shows the maximum of the velocity gradient vs time for the convection flows (there is no difference between the moving and the fixed convection flows) and the sinusoidal shear flow. The difference in magnitude and also in the temporal evolution between these two flows is clearly visible, indicating a possibly very different behavior with respect to shear fragmentation.

E. Numerical implementation

After presenting the model, we describe some details about the implementation. In the bulk of the paper we con-



FIG. 1. Maximum of the velocity gradient grad $\mathbf{u}(x_1, x_2, t)$ vs time for the two different flows described by Eqs. (4) and (5).

sider n=30 size classes. The primary particles considered here have dimensionless radius $r_1=5/30^{1/3} \times 10^{-5}$, response time $\tau_1=1/55$, and settling velocity $W_1=3.2\tau_1$. As initial condition we take $N(t=0)=10^5$ particles of the smallest size and no larger particles. Furthermore, particles are uniformly distributed over the 1×1 unit cell of the configuration space. The initial particle velocity matches that of the fluid at their position in all cases.

The simulation is based on the following ingredients:

(a) All particles move in the flow over some time step dt according to Eq. (1). This integration time step dt needs to be chosen small enough to allow for the detection of every collision. After each time step dt there is an interaction between particles in the form of coagulation if they are too close to each other. Our experience shows that a choice dt=T/20 is sufficiently small for the conditions considered here.

Because of the spatial periodicity of the flow, the particle dynamics is folded back onto the 1×1 unit cell, using periodic boundary conditions (see, e.g., [40,42]).

(b) Particles coagulate if their distance is smaller than the sum of their radii. Computationally, the coagulation process is the most costly component of the simulation. Here a link-cell algorithm [46] is used to compute the distance between particles, which scales as O(N) and is thus much faster than simply summing over all particles.

(c) Coagulates can fragment either due to size-limiting fragmentation or due to shear fragmentation.

(1) Size-limiting fragmentation. If the coagulate size α exceeds the predefined maximum size, which is in the following fixed at n=30 unless mentioned otherwise, the coagulate is broken up.

(2) Shear fragmentation. If the shear at the position of the coagulate exceeds a critical value, determined by Eq. (3) the coagulate breaks up. Due to the symmetry of the flows chosen here, the maximum velocity difference is always in the direction of one of the coordinate axes, therefore only these values have to be calculated. Shear fragmentation is always applied together with size-limiting fragmentation to keep the maximum number of occurring size classes fixed at n.

Whatever rule is applied, the result is the reversed process of coagulation: two new particles are formed from an old one with the size class indices: $\alpha_{i,new} + \alpha_{j,new} = \alpha_{old}$. As indicated earlier, $\alpha_{i,new}$ can take on any value between α_1 and $\alpha_{n/2}$ with equal probability. The centers of the new particles are placed along a line segment in a random direction so that the distance *d* between the particle centers equals the sum of their radii, i.e., $d=r_i+r_j$, and the center of mass remains un-



FIG. 2. Stroboscopic sections of the attractors of Eq. (1) projected onto the configuration space for particles for size class α = 16 for: (a) moving convection flow, (b) fixed convection flow, and (c) sinusoidal shear flow. The difference in the degree of clustering in the three flows is clearly visible.

changed. Momentum is conserved. For simplicity we assume that the new particles have the same velocity as the old one.

III. SIMULATION RESULTS

In this section we show simulation results using the model described above and compare the influence of the different flows and the effect of size-limiting fragmentation and shear fragmentation.

Before presenting any results for the complete model, it is worth showing the attractors for the noninteracting problem in the different flows. Figure 2 shows the stroboscopic section (taken with the period T of the flow) of the attractors for flows (a)–(c) for one specific size class projected onto the plane of the coordinates. The figure illustrates the difference in the geometric properties of the particles dynamics in the different flows.

For the moving convection flow and the sinusoidal shear flow the degree of clustering of the particles in the attractors, quantified by their fractal dimension, decreases monotonically with the size class. The parameter region is chosen in such a way that the attractors are either area filling or fractal with dimension smaller than 2, which we consider to be closer to a realistic situation than, for example, fixed-point attractors. For the fixed convection flow, the particles tend to cluster on a quasiperiodic attractor. This leads to a much larger collision rate than in the other two cases.

In all flows we find convergence to an asymptotic steady state. Initially, coagulation leads to a fast increase in the average particle size class, independent of the fragmentation rules. Then fragmentation sets in and a balance between coagulation and fragmentation is reached, with an asymptotic average coagulate size

$$\alpha_{\infty} = \lim_{t \to \infty} \frac{1}{T} \int_{t}^{t+T} ds \langle \alpha(s) \rangle \tag{6}$$

that depends on the fragmentation rule and the different flows. The average $\langle \alpha(s) \rangle$ is taken over the coagulate sizes at time *s* and Eq. (6) corresponds to time averaging $\langle \alpha(s) \rangle$ over one time period of the flow to remove the periodicity. For the transient behavior of the coagulation-fragmentation process the geometric properties, in particular the degree of clustering of the particles, are very important since they affect the time scales of the transients. This is not the case for the



COAGULATION AND FRAGMENTATION DYNAMICS OF ...

FIG. 3. Histogram of the particle size distribution in steady state for size-limiting fragmentation and shear fragmentation with (a) moving convection cell flow, (with γ =50 for shear fragmentation), (b) fixed convection cell flow, (with γ =45 for shear fragmentation), and (c) sinusoidal shear flow (with γ =17 for shear fragmentation). The insets show the exponential tail of the size distributions for shear fragmentation in the case of the moving convection cell flow and the fixed convection cell flow.

steady state. This becomes very clear when looking at the size distributions of coagulates in steady state for the different flows and fragmentation mechanisms (Fig. 3). One might expect that the steady state for flow (b) is very different from the other two cases, due to the large difference in particle clustering. Our results show however that the steady state of the particle dynamics for flows (a) and (b) are almost identical, while flow (c) produces different results. For example for both flows (a) and (b), the size distribution has a long tail toward larger size classes that decays exponentially for shear fragmentation. Similar exponential tails for the particle size distribution have been found in observations of aggregates in the ocean (see, e.g., [47]).

By contrast, for the sinusoidal shear flow the size distribution has two peaks and then drops off sharply toward zero beyond the second peak. In this case the size distribution for shear fragmentation is almost identical to that of size-limiting fragmentation, but for a lower value of α_{max} . This is due to the fact that in the sinusoidal shear flow for the chosen parameters the shear a particle experiences is almost constant over time and space, except for a small "dip" every half period. This very narrow distribution of the shear is very similar to having a single maximum stable size, as is the case

PHYSICAL REVIEW E 80, 026311 (2009)



FIG. 4. Histogram of the particle size distribution in steady state for the moving convection cell flow (γ =50) and the smoothed out sinusoidal shear flow (γ =8.5, β = π /20) for shear fragmentation.

for size-limiting fragmentation. In this case a value of $\gamma = 17$ for shear fragmentation corresponds to a value of $\alpha_{max} = 20$. We have also checked the size distributions in subregions

We have also checked the size distributions in subregions of the flows. We found, that as long as the number of particles in the subregions allowed sufficiently good statistics, the normalized size distributions coincided with the global distributions. This means that there is no significant spatial dependence of the size distribution.

While the specific shape of the size distributions found may not be very general, both because of the limitation to only thirty size classes and the very simplified flows, our approach illustrates clearly that the geometric properties of the particle motion related to preferential concentration are not the most relevant ones for the steady-state distributions. Instead, the most important effect for the steady state of the particles seems to be the fragmentation process.

Size-limiting fragmentation is the same in all flows, as it does not depend on properties of the flow. In this case the differences in the size distributions are small for the different flows. This indicates that the size distributions are mainly determined by the fragmentation process because the flow specific differences, e.g., differences in coagulation rates, do not affect the shape of the size distribution. However, when fragmentation depends on shear, the different flows produce very different size distributions. This difference is mainly due to the different properties of the shear forces in the flow (Fig. 1), which lead to differences in fragmentation. To see that it does not depend, e.g., on the detailed characteristics of the particle motion or on the different collision rates, we can adjust the flow parameters. Decreasing the value of the parameter β for the sinusoidal shear flow to a much smaller one, e.g., $\beta = \pi/20$, greatly increases the variation in the shear forces over space and time. We obtain two sinusoidal peaks per period for the shear forces, similar to what happens in the convection flow (cf. Fig. 1), except that for the sinusoidal shear flow both peaks have the same height. For an appropriate choice of γ , so that the average size classes match, it can be seen that the shape of the particle size distributions for both flows have become almost identical (Fig. 4) and show the characteristic exponential tail. Again, this indicates that the shape of the size distribution is mostly affected by fragmentation and any flow specific differences in the distribution result mainly from differences in the shear distribution in the flow, which in turn change the fragmentation rates.

This strong dependence of the steady state on the fragmentation process can also clearly be seen by how α_{∞}



FIG. 5. Asymptotic average size class index α_{∞} as a function of the coagulate strength parameter γ . Squares: numerical results, solid line: fit by $\alpha_{\infty}=c_1+c_2\gamma^3$ [see Eq. (8)]. (a) moving convection cell flow ($c_1=0.521$, $c_2=3.7\times10^{-5}$), (b) fixed convection cell flow ($c_1=-0.256$, $c_2=6.2\times10^{-5}$), and (c) sinusoidal shear flow ($c_1=0.945$, $c_2=1.6\times10^{-3}$).

changes with the coagulate strength γ (see Fig. 5). We note that the shear forces in the convection flows and the sinusoidal shear flow have a different magnitude, as seen in Fig. 1. Therefore three different values of the coagulate strength parameter γ need to be chosen to yield a size distribution that is not only determined by size-limiting fragmentation. For the convection flows the coagulate strength γ needs to be approximately a factor of 3 larger than for the sinusoidal shear flow.

It is clear that α_{∞} increases with γ because particles become more resistant to shear. The exact functional relationship is however not trivial. A first qualitative estimate of the shape of this $\alpha_{\infty}(\gamma)$ curve can be derived by assuming that over one period of the flow the particles experience an "average shear"

$$\bar{G} = \frac{1}{T} \int_0^T dt \int_D d\mathbf{x} p(\mathbf{x}, t) G(\mathbf{x}, t), \tag{7}$$

where $G(\mathbf{x},t)$ is the modulus of the local velocity gradient, $p(\mathbf{x},t)$ is the distribution of particles, and *D* is the unit square domain. From Eq. (3) we then get for the average critical size at this velocity gradient

PHYSICAL REVIEW E 80, 026311 (2009)



FIG. 6. In the case of shear fragmentation the steady state is not static, instead it fluctuates with the period of the flow: (a) average size class vs time for the fixed convection flow and (b) close up of (a).

$$\bar{\alpha}_{crit} = \bar{G}^{-3} \gamma^3. \tag{8}$$

Particles that exceed this size will therefore typically break up during one period of the flow. Since particles break into two parts due to shear, the average size would then be $\alpha_{\infty} \ge \overline{\alpha}_{crit}/2$. The average shear \overline{G} is, however, somewhat complicated to estimate. It would have to be calculated as a mean over the positions of all particles in the flow at a given time. Additionally, how much larger than the critical size particles get before they break up depends on the coagulation probabilities, and therefore also on the local concentrations of particles. The exact dependency of $\alpha_{\infty}(\gamma)$ is therefore not easily calculated. What can be seen from Eq. (8) is however that the average size is expected to scale with γ as

$$\alpha_{\infty} \propto \gamma^3$$
. (9)

This dependence is expected to hold for all values of γ and α_{∞} , where shear fragmentation dominates. A fit with Eq. (9) for the different flows is shown in Fig. 5 and for lower values of γ the fits agree very well with the simulation results. It can be seen that for higher values of γ , when size-limiting fragmentation becomes important, the $\alpha_{\infty}(\gamma)$ curves deviate from this estimate and converge toward the limiting value $\alpha_{\infty}^{(lim)}$ (see Fig. 5). This result demonstrates how the steady state depends very strongly on the fragmentation process. However, the different proportionality constants for Eq. (9) still depend on the flow and can also depend on the spatial distribution of the particles since different regions of the flow might exhibit different shear.

Finally, we mention some further results from our model. First, for all flows the steady state in the case of shear fragmentation is not static, instead due to the periodic time dependence of the flows the steady state also varies periodically over time. This is very clear for example for the fixed convection flows when looking at the average size class index $\langle \alpha(t) \rangle = \sum_{i=1}^{30} \alpha_i N_{\alpha_i}(t) / N(t)$, where N_{α_i} denotes the number of particles in size class α_i (Fig. 6).

Such a time dependence of the average particle size, and therefore of the whole particle size distribution, can have important physical consequences. For example the settling of coagulates in the ocean, which is an important part of the biological carbon pump in the ocean, is greatly affected by the size distribution of the coagulates. In coastal areas, where the fluid may be periodically forced by the tides, such a time COAGULATION AND FRAGMENTATION DYNAMICS OF ...



FIG. 7. Normalized number of particles versus relative radius in steady state in the case of shear fragmentation for different values of the coagulate strength parameter γ . The normalized size distributions collapse onto a master curve that depends on the fragmentation mechanism and the shear forces in the flow for (a) moving convection cell flow [γ =44 (circle), 50 (square), 56 (triangle)], (b) fixed convection cell flow [γ =42.5 (circle), 45 (square), 50 (triangle)], and (c) sinusoidal shear flow [γ =16 (circle), 17 (square), 18 (triangle), β =20/ π].

dependence of the distribution can greatly affect the settling rates and therefore the deposition of coagulates [47].

Second, since the range of possible parameter values for our model is very large, we also mention the robustness of our findings with respect to the following different parameters:

(i) For each flow there is a certain range of the coagulate strength parameters γ where the size distribution for shear fragmentation is "fully developed." By this we mean that γ is large enough so that a sufficiently large fraction of particles has left the smallest size class, but γ is small enough so that break up due to size-limiting fragmentation does not play a significant role. In this intermediate γ range, where the particle size distribution is fully developed, a scaling form

$$\frac{N_{\alpha}}{\max(N_{\alpha})} = f\left(\frac{r}{\langle r \rangle}\right) \tag{10}$$

is found to hold (Fig. 7), where $\langle r \rangle$ represents the average radius. Note that the form of the size distribution is independent of γ . All distributions in this parameter range collapse then onto a single master curve.

While this scaling form is independent of the parameters of the coagulation and of the coagulate strength, the difference between the different flows remains. More specifically, the scaling form f changes when shear forces in the flow or the fragmentation mechanism, for example the distribution of fragments, is varied.

(ii) When investigating cases with different total mass M, we find that for size-limiting fragmentation α_{∞} is largely independent of M. For shear fragmentation with $M < 3 \times 10^6 m_1$, $\alpha_{\infty}(M)$ increases approximately linearly with M, while for higher values a saturation of $\alpha_{\infty}(M)$ sets in, which is due to the fact that size-limiting fragmentation dominates in this case.

(iii) By considering other initial particle size distributions than that mentioned above, for example any single size class with $\alpha > 1$ or a uniform distribution of sizes, and keeping the total mass *M* fixed, the asymptotic state is found to be independent of the chosen initial distribution for each flow and for both fragmentation rules.

PHYSICAL REVIEW E 80, 026311 (2009)

(iv) We also investigated the role of the number of size classes and found that in the chosen range of γ values the size distributions for shear fragmentation are not influenced by the number of size classes.

(v) The effect of the number of new particles formed by fragmentation has been considered. For instance, the distributions of particles for ternary fragmentation are similar to the ones for binary splitting and only show a slight shift toward smaller size classes [48].

IV. SUMMARY

We discussed the formation of a steady-state size distribution in a coagulation-fragmentation process of inertial particles transported by different flows. Our most important finding is that fragmentation rather than coagulation is the dominating process for the steady-state size distribution. For size-limiting fragmentation we found almost no differences in the shape of the steady-state size distribution for various flows. Even in flows with great differences in coagulation rates, e.g., due to differences in local particle concentrations, particle size distributions remained very similar. For the case of shear fragmentation differences in the shape of the size distribution for the different flows appeared. It was found that these were due to differences in the spatial and temporal variation in the shear, which in turn affected the fragmentation.

We have shown that an individual particle based modeling approach is able to reflect typical properties of coagulation and fragmentation processes of inertial particles. The appearance of a steady state is demonstrated. We outlined some of the differences in the convergence to the steady state and the particle size distribution that can result from different types of fragmentation and flow. Altogether, our results suggest that coagulation dominates different time spans of the process than fragmentation. While coagulation is most important for the transients in the beginning, the steady-state size distribution is mainly determined by fragmentation. As a consequence the spatial distribution of particles plays only a transient role. The underlying flow is important for the steady state in the case of shear fragmentation, as the spatial and temporal variation in the shear can greatly influence the fragmentation rates.

The generalization to a fully three-dimensional system is straightforward. The relaxation toward the steady state would slow down, due to the decreased probability of collisions. However, our conclusions regarding the dependence of the steady-state size distribution on the fragmentation remain valid.

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- T. Nishikawa, Z. Toroczkai, and C. Grebogi, Phys. Rev. Lett. 87, 038301 (2001).
- [2] I. J. Benczik, Z. Toroczkai, and T. Tél, Phys. Rev. Lett. 89, 164501 (2002).
- [3] C. Lopez, Phys. Rev. E 66, 027202 (2002).
- [4] J. H. E. Cartwright, M. O. Magnasco, O. Piro, and I. Tuval, Phys. Rev. Lett. 89, 264501 (2002).
- [5] Y. Do and Y. C. Lai, Phys. Rev. E 70, 036203 (2004).
- [6] I. J. Benczik, G. Karolyi, I. Scheuring, and T. Tél, Chaos 16, 043110 (2006).
- [7] R. D. Vilela and A. E. Motter, Phys. Rev. Lett. 99, 264101 (2007).
- [8] G. Haller and T. Sapsis, Physica D 237, 573 (2008).
- [9] R. A. Shaw, Annu. Rev. Fluid Mech. 35, 183 (2003).
- [10] G. Falkovich and A. Pumir, J. Atmos. Sci. 64, 4497 (2007).
- [11] A. Jaczewski and S. P. Malinowski, Q. J. R. Meteorol. Soc. 131, 2047 (2005).
- [12] M. Wilkinson and B. Mehlig, Europhys. Lett. 71, 186 (2005).
- [13] J. Bec, A. Celani, M. Cencini, and S. Musacchio, Phys. Fluids 17, 073301 (2005).
- [14] E. Calzavarini, M. Cencini, D. Lohse, and F. Toschi, Phys. Rev. Lett. 101, 084504 (2008).
- [15] M. R. Maxey and S. Corrsin, J. Atmos. Sci. 43, 1112 (1986).
- [16] M. R. Maxey, J. Fluid Mech. 174, 441 (1987).
- [17] M. Wilkinson, B. Mehlig, S. Östlund, and K. P. Duncan, Phys. Fluids 19, 113303 (2007).
- [18] G. Falkovich, A. Fouxon, and M. G. Stepanov, Nature (London) 419, 151 (2002).
- [19] Y. Zhou, A. S. Wexler, and L.-P. Wang, J. Fluid Mech. 433, 77 (2001).
- [20] L.-P. Wang, A. S. Wexler, and Y. ZhouJ. Fluid Mech. 415, 117 (2000).
- [21] J. C. Zahnow, R. D. Vilela, U. Feudel, and T. Tél, Phys. Rev. E 77, 055301(R) (2008).
- [22] Rene O. Medrano-T., A. Moura, T. Tél, I. L. Caldas, and C. Grebogi, Phys. Rev. E 78, 056206 (2008).
- [23] In Ref. [21] coagulation was referred to as aggregation. However, in the context of liquid particles coagulation is the more widely used term.
- [24] H. R. Pruppacher and J. D. Klett, *Microphysics of Clouds and Precipitation* (Kluwer, Dordrecht, 1997).
- [25] A. L. Alldredge, T. Granata, C. Gotschalk, and T. Dickey, Lim-

nol. Oceanogr. 35, 1415 (1990).

- [26] E. Villermaux, Annu. Rev. Fluid. Mech. 39, 419 (2007).
- [27] D. N. Thomas, S. Judd, and N. Fawcett, Water Res. 33, 1579 (1999).
- [28] M. Smoluchowski, Z. Phys. Chem. 92, 129 (1917).
- [29] J. Bec, Phys. Fluids 15, L81 (2003).
- [30] P. K. Kundu and I. M. Cohen, *Fluid Mechanics*, 4th ed. (Academic, New York, 2008).
- [31] M. R. Maxey and J. J. Riley, Phys. Fluids 26, 883 (1983).
- [32] T. R. Auton, J. Hunt, and M. Prud'homme, J. Fluid Mech. 197, 241 (1988).
- [33] E. E. Michaelides, ASME Trans. J. Fluids Eng. **119**, 233 (1997).
- [34] J. Happel and H. Brenner, Low Reynolds Number Hydrodynamics (Martinus Nijhoff Publishers, The Hague, 1983).
- [35] I. M. Jánosi, J. O. Kessler, and V. K. Horváth, Phys. Rev. E 58, 4793 (1998).
- [36] For the cases studied here, typical distances between particles are $\geq 50a_1$.
- [37] G. I. Taylor, Proc. R. Soc. London, Ser. A 146, 501 (1934).
- [38] M. Delichatsios, Phys. Fluids 18, 622 (1975).
- [39] S. Chandrasekhar, Hydrodynamic and Hydromagnetic Stability
- (Oxford University Press, New York, 1961). [40] J. C. Zahnow and U. Feudel, Phys. Rev. E **77**, 026215 (2008).
- [41] M. R. Maxey, Phys. Fluids 30, 1915 (1987).
- [42] T. Nishikawa, Z. Toroczkai, C. Grebogi, and T. Tél, Phys. Rev. E 65, 026216 (2002).
- [43] M. Liu, F. Muzzio, and R. Peskin, Chaos, Solitons Fractals 4, 869 (1994).
- [44] R. T. Pierrehumbert, Chaos, Solitons Fractals 4, 1091 (1994).
- [45] R. D. Vilela, T. Tél, A. P. S. de Moura, and C. Grebogi, Phys. Rev. E 75, 065203(R) (2007).
- [46] R. W. Hockney and J. W. Eastwood, *Computer Simulations Using Particles* (McGraw-Hill, New York, 1981).
- [47] M. Lunau, A. Lemke, O. Dellwig, and M. Simon, Limnol. Oceanogr. 51, 847 (2006).
- [48] A change in the fragmentation rule has, however, important consequences. By replacing the rule of uniform distribution for the possible size classes after fragmentation by the rule that coagulates always split into two halves of similar sizes makes the size distribution exponential, practically over all size classes.

Bibliography

- Abrahamson, J. (1975). Collision rates of small particles in a vigorously turbulent fluid. Chemical Engineering Science, 30(11):1371–1379.
- Alexander, S. (1998). Amorphous solids: Their structure, lattice dynamics and elasticity. *Physics Reports*, 296(2-4):65–236.
- Alldredge, A. L., Granata, T. C., Gotschalk, C. C., and Dickey, T. D. (1990). The physical strength of marine snow and its implications for particle disaggregation in the ocean. *Limnology And Oceanography*, 35(7):1415–1428.
- Aref, H. (1984). Stirring by chaotic advection. Journal of Fluid Mechanics, 143:1–21.
- Aref, H. and Balachandar, S. (1986). Chaotic advection in a stokes-flow. *Physics of Fluids*, 29(11):3515–3521.
- Argyris, J., Faust, G., and Haase, M. (1994). An Exploration of Chaos. North Holland, Amsterdam.
- Arneodo, A., Benzi, R., Berg, J., Biferale, L., Bodenschatz, E., Busse, A., Calzavarini,
 E., Castaing, B., Cencini, M., Chevillard, L., Fisher, R. T., Grauer, R., Homann,
 H., Lamb, D., Lanotte, A. S., Leveque, E., Luthi, B., Mann, J., Mordant, N., Muller,
 W. C., Ott, S., Ouellette, N. T., Pinton, J. F., Pope, S. B., Roux, S. G., Toschi, F., Xu,
 H., and Yeung, P. K. (2008). Universal intermittent properties of particle trajectories in highly turbulent flows. *Physical Review Letters*, 100(25):254504.
- Ayala, O., Rosa, B., and Wang, L. P. (2008a). Effects of turbulence on the geometric collision rate of sedimenting droplets. Part 2. Theory and parameterization. New Journal of Physics, 10:075016.
- Ayala, O., Rosa, B., Wang, L. P., and Grabowski, W. W. (2008b). Effects of turbulence on the geometric collision rate of sedimenting droplets. Part 1. Results from direct numerical simulation. *New Journal of Physics*, 10:075015.
- Ayyalasomayajula, S., Gylfason, A., Collins, L. R., Bodenschatz, E., and Warhaft, Z. (2006). Lagrangian measurements of inertial particle accelerations in grid generated wind tunnel turbulence. *Physical Review Letters*, 97(14):144507.

- Ayyalasomayajula, S., Warhaft, Z., and Collins, L. R. (2008). Modeling inertial particle acceleration statistics in isotropic turbulence. *Physics of Fluids*, 20(9):095104.
- Bäbler, M. U., Morbidelli, M., and Baldyga, J. (2008). Modelling the breakup of solid aggregates in turbulent flows. *Journal of Fluid Mechanics*, 612:261–289.
- Bäbler, M. U., Sefcik, J., Morbidelli, M., and Baldyga, J. (2006). Hydrodynamic interactions and orthokinetic collisions of porous aggregates in the Stokes regime. *Physics* of Fluids, 18(1):013302.
- Baldyga, J. and Bourne, J. R. (1995). Interpretation of turbulent mixing using, fractals and multifractals. *Chemical Engineering Science*, 50(3):381–400.
- Balkovsky, E., Falkovich, G., and Fouxon, A. (2001). Intermittent distribution of inertial particles in turbulent flows. *Physical Review Letters*, 86(13):2790–2793.
- Basset, A. (1888). A Treatise on Hydrodynamics, volume 2. Deighton Bell, London.
- Batchelor, G. (1967). An Introduction to Fluid Dynamics. Cambridge University Press, Cambridge, UK.
- Batchelor, G. K. (1970). The stress system in a suspension of force-free particles. *Journal* of Fluid Mechanics, 41(03):545–570.
- Bec, J. (2003). Fractal clustering of inertial particles in random flows. *Physics of Fluids*, 15(11):L81–L84.
- Bec, J. (2005). Multifractal concentrations of inertial particles in smooth random flows. Journal of Fluid Mechanics, 528:255–277.
- Bec, J., Biferale, L., Boffetta, G., Celani, A., Cencini, M., Lanotte, A., Musacchio, S., and Toschi, F. (2006a). Acceleration statistics of heavy particles in turbulence. *Journal* of Fluid Mechanics, 550:349–358.
- Bec, J., Biferale, L., Boffetta, G., Cencini, M., Musacchio, S., and Toschi, F. (2006b). Lyapunov exponents of heavy particles in turbulence. *Physics of Fluids*, 18(9):091702.
- Bec, J., Biferale, L., Cencini, M., Lanotte, A., Musacchio, S., and Toschi, F. (2007). Heavy particle concentration in turbulence at dissipative and inertial scales. *Physical Review Letters*, 98(8):084502.
- Bec, J., Celani, A., Cencini, M., and Musacchio, S. (2005). Clustering and collisions of heavy particles in random smooth flows. *Physics of Fluids*, 17:073301.

- Becker, V., Schlauch, E., Behr, M., and Briesen, H. (2009). Restructuring of colloidal aggregates in shear flows and limitations of the free-draining approximation. *Journal* of Colloid and Interface Science, 339(2):362–372.
- Behringer, R. P., Meyers, S. D., and Swinney, H. L. (1991). Chaos and mixing in a geostrophic flow. *Physics of Fluids A*, 3(5):1243–1249.
- Benczik, I. J., Karolyi, G., Scheuring, I., and Tel, T. (2006). Coexistence of inertial competitors in chaotic flows. *Chaos*, 16(4):043110.
- Benettin, G., Galgani, L., Giorgilli, A., and Strelcyn, J.-M. (1980). Lyapunov characteristic exponents for smooth dynamical systems and for Hamiltonian systems; a method for computing all of them. Part1: Theory and Part2: Numerical application. *Meccanica*, 15:9–30.
- Biferale, L., Bodenschatz, E., Cencini, M., Lanotte, A. S., Ouellette, N. T., Toschi, F., and Xu, H. (2008). Lagrangian structure functions in turbulence: A quantitative comparison between experiment and direct numerical simulation. *Physics of Fluids*, 20(6):065103.
- Biferale, L., Boffetta, G., Celani, A., Devenish, B. J., Lanotte, A., and Toschi, F. (2004). Multifractal statistics of Lagrangian velocity and acceleration in turbulence. *Physical Review Letters*, 93(6):064502.
- Biferale, L. and Toschi, F. (2005). Joint statistics of acceleration and vorticity in fully developed turbulence. *Journal of Turbulence*, 6:N40.
- Born, M. and Huang, H. (1954). *Dynamical Theory of Crystal Lattices*. Oxford University Press.
- Boussinesq, J. (1903). *Theorie Analytique de la Chaleur*, volume 2. L'École Polytechnique, Paris.
- Brady, J. F. and Bossis, G. (1988). Stokesian dynamics. Annual Review of Fluid Mechanics, 20:111–157.
- Calzavarini, E., Cencini, M., Lohse, D., and Toschi, F. (2008). Quantifying turbulenceinduced segregation of inertial particles. *Physical Review Letters*, 101(8):084504.
- Celani, A. and Vergassola, M. (2001). Statistical geometry in scalar turbulence. *Physical Review Letters*, 86(3):424–427.

- Cencini, M., Bec, J., Biferale, L., Boffetta, G., Celani, A., Lanotte, A., Musacchio, S., and Toschi, F. (2006). Dynamics and statistics of heavy particles in turbulent flows. *Journal of Turbulence*, 7(36):1–16.
- Chaiken, J., Chevray, R., Tabor, M., and Tan, Q. M. (1986). Experimental-study of lagrangian turbulence in a stokes-flow. *Proceedings of the Royal Society of London Series A*, 408(1834):165–&.
- Clift, R., Grace, J., and Weber, M. E. (2005). *Bubbles, Drop and Particles.* Dover Publishing.
- Coimbra, C. F. M. and Kobayashi, M. H. (2002). On the viscous motion of a small particle in a rotating cylinder. *Journal of Fluid Mechanics*, 469:257–286.
- Cristini, V., Guido, S., Alfani, A., Blawzdziewicz, J., and Loewenberg, M. (2003). Drop breakup and fragment size distribution in shear flow. *Journal of Rheology*, 47(5):1283– 1298.
- Cundall, P. A. and Strack, O. D. L. (1979). Discrete numerical-model for granular assemblies. *Geotechnique*, 29(1):47–65.
- Delichatsios, M. (1975). Model for the breakup of spherical drops in isotropic turbulent flows. *Physics of Fluids*, 18:622.
- DiFelice, R. (1994). The voidage function for fluid particle interaction systems. *International Journal of Multiphase Flow*, 20(1):153–159.
- Drew, D. and Passman, S. (1999). Theory of multicomponent fluids. Springer, New York.
- Duncan, K., Mehlig, B., Ostlund, S., and Wilkinson, M. (2005). Clustering by mixing flows. *Physical Review Letters*, 95(24):240602.
- Durst, F. (2006). Grundlagen der Strömungsmechanik. Springer-Verlag, Berlin.
- Eaton, J. K. and Fessler, J. R. (1994). Preferential concentration of particles by turbulence. International Journal of Multiphase Flow, 20:169–209.
- Eckmannn, J.-P. and Ruelle, D. (1985). Ergodic theory of chaos and strange attractors. *Reviews of Modern Physics*, 57:617–656.
- Eggersdorfer, M. L., Kadau, D., Herrmann, H. J., and Pratsinis, S. E. (2010). Fragmentation and restructuring of soft-agglomerates under shear. *Journal of Colloid and Interface Science*, 342(2):261–268.

- Einstein, A. (1906). Eine neue Bestimmung der Moleküldimensionen. Annalen der Physik, 324(2):289–306. Correction (1911), ibid., 339(3):591–592.
- Falkovich, G., Fouxon, A., and Stepanov, M. G. (2002). Acceleration of rain initiation by cloud turbulence. *Nature*, 419(6903):151–154.
- Falkovich, G., Gawedzki, K., and Vergassola, M. (2001). Particles and fields in fluid turbulence. *Reviews of Modern Physics*, 73(4):913–975.
- Falkovich, G. and Pumir, A. (2007). Sling effect in collisions of water droplets in turbulent clouds. Journal of the Atmospheric Sciences, 64(12):4497–4505.
- Faxén, H. (1922). Der Widerstand gegen die Bewegung einer starren Kugel in einer z\u00e4hen Fl\u00fcssigkeit, die zwischen zwei parallelen ebenen W\u00e4nden eingeschlossen ist. Annalen der Physik, 373(10):89–119.
- Feng, Y. T., Han, K., and Owen, D. R. J. (2007). Coupled lattice boltzmann method and discrete element modelling of particle transport in turbulent fluid flows: Computational issues. International Journal for Numerical Methods in Engineering, 72(9):1111–1134.
- Fessler, J. R., Kulick, J. D., and Eaton, J. K. (1994). Preferential concentration of heavy-particles in a turbulent channel flow. *Physics of Fluids*, 6(11):3742–3749.
- Flesch, J., Spicer, P., and Pratsinis, S. (1999). Laminar and turbulent shear-induced flocculation of fractal aggregates. *AIChE Journal*, 45(5):1114.
- Friedlander, S. K. (2000). *Smoke, Dust, and Haze*. Oxford University Press, New York, second edition.
- Glowinski, R., Pan, T. W., Hesla, T. I., and Joseph, D. D. (1999). A distributed Lagrange multiplier fictitious domain method for particulate flows. *International Journal of Multiphase Flow*, 25(5):755–794.
- Guala, M., Luthi, B., Liberzon, A., Tsinober, A., and Kinzelbach, W. (2005). On the evolution of material lines and vorticity in homogeneous turbulence. *Journal of Fluid Mechanics*, 533:339–359.
- Hamaker, H. (1937). The London van der Waals attraction between spherical particles. *Physica*, 4:1058–1072.
- Han, B., Akeprathumchai, S., Wickramasinghe, S., and Qian, X. (2003). Flocculation of biological cells: Experiment vs. theory. AIChE Journal, 49(7):1687.

- Happel, J. and Brenner, H. (1983). Low Reynolds Number Hydrodynamics. Martinus Nijhoff Publishers, The Hague.
- Harada, S., Tanaka, R., Nogami, H., and Sawada, M. (2006). Dependence of fragmentation behavior of colloidal aggregates on their fractal structure. *Journal of Colloid and Interface Science*, 301(1):123–129.
- Herron, I. H., Davis, S. H., and Bretherton, F. P. (1975). Sedimentation of a sphere in a centrifuge. *Journal of Fluid Mechanics*, 68(MAR25):209–234.
- Higashitani, K., Iimura, K., and Sanda, H. (2001). Simulation of deformation and breakup of large aggregates in flows of viscous fluids. *Chemical Engineering Science*, 56(9):2927– 2938.
- Hill, P. J. and Ng, K. M. (1996). Statistics of multiple particle breakage. AiChE Journal, 42(6):1600–1611.
- Hockney, R. and Eastwood, J. (1981). *Computer Simulation Using Particles*. McGraw-Hill International.
- Israelachvili, J. N. (1985). Intermolecular and Surface Forces. Academic Press, London.
- Jaczewski, A. and Malinowski, S. P. (2005). Spatial distribution of cloud droplets in a turbulent cloud-chamber flow. Quarterly Journal of the Royal Meteorological Society, 131(609):2047–2062.
- Jarvis, P., Jefferson, B., Gregory, J., and Parsons, S. A. (2005). A review of floc strength and breakage. Water Research, 39(14):3121–3137.
- Kailasnath, P., Sreenivasan, K. R., and Stolovitzky, G. (1992). Probability density of velocity increments in turbulent flows. *Physical Review Letters*, 68(18):2766–2769.
- Kaplan, J. and Yorke, J. (1979). Chaotic behavior of multidimensional difference equations. In Peitgen, H.-O. and Walter, H.-O., editors, *Proceedings on Functional Differential Equations and Approximations of Fixed Points*, volume 730 of *Lecture Notes in Mathematics*, pages 204–227, Berlin. Springer.
- Károlyi, G., Pentek, A., Scheuring, I., Tél, T., and Toroczkai, Z. (2000). Chaotic flow: The physics of species coexistence. Proceedings of the National Academy of Sciences of the United States of Americs, 97(25):13661–13665.
- Károlyi, G., Péntek, A., Toroczkai, Z., Tél, T., and Grebogi, C. (1999). Chemical or biological activity in open chaotic flows. *Physical Review E*, 59(5):5468–.

- Kloeden, P. E. and Platen, E. (1999). Numerical Solution of Stochastic Differential Equations. Springer-Verlag, Berlin, third edition.
- Knudsen, H. A., Werth, J. H., and Wolf, D. E. (2008). Failure and success of hydrodynamic interaction models. *European Physical Journal E*, 27(2):161–170.
- Kobayashi, M., Adachi, Y., and Ooi, S. (1999). Breakup of fractal flocs in a turbulent flow. Langmuir, 15:4351–4356.
- Kranenburg, C. (1994). The fractal structure of cohesive sediment aggregates. Estuarine, Coastal and Shelf Science, 39:451–460.
- Landau, L. and Lifschitz, E. (1991). Lehrbuch der Theoretischen Physik: Band VI, Hydrodynamik. Akademie Verlag, Berlin, 5. edition.
- Lawrence, C. J. and Mei, R. W. (1995). Long-time behavior of the drag on a body in impulsive motion. *Journal of Fluid Mechanics*, 283:307–327.
- Li, X. Y. and Logan, B. E. (2001). Permeability of fractal aggregates. *Water Research*, 35(14):3373–3380.
- Logan, B. E. (1999). Environmental Transport Processes. John Wiley and Sons, New York.
- Lomholt, S. and Maxey, M. R. (2003). Force-coupling method for particulate two-phase flow: Stokes flow. Journal of Computational Physics, 184(2):381–405.
- Loth. E. (2010).Particles. Drops and Bubbles: Fluid Dynamics and Numerical Methods. Book Draft for Cambridge University Press (www.ae.illinois.edu/~loth/CUP/Loth.htm).
- Loth, E. and Dorgan, A. (2009). An equation of motion for particles of finite reynolds number and size. *Environmental Fluid Mechanics*, 9(2):187–206.
- Luding, S. (2008). Cohesive, frictional powders: contact models for tension. Granular Matter, 10(4):235–246.
- Lunau, M., Lemke, A., Dellwig, O., and Simon, M. (2006). Physical and biogeochemical controls of microaggregate dynamics in a tidally affected coastal ecosystem. *Limnology* and Oceanography, 51:847–859.
- Lüthi, B., Tsinober, A., and Kinzelbach, W. (2005). Lagrangian measurement of vorticity dynamics in turbulent flow. *Journal of Fluid Mechanics*, 528:87–118.

- Maerz, J. and Wirtz, K. (2009). Resolving physically and biologically driven suspended particulate matter dynamics in a tidal basin with a distribution-based model. *Estuarine Coastal and Shelf Science*, 84(1):128–138.
- Maggi, F., Mietta, F., and Winterwerp, J. C. (2007). Effect of variable fractal dimension on the floc size distribution of suspended cohesive sediment. *Journal of Hydrology*, 343(1-2):43–55.
- Mandelbrot, B. B. (1983). *The fractal geometry of nature*. W.H. Freeman and Company, New York.
- Marshall, J. S. and Palmer, W. M. (1948). The distribution of raindrops with size. *Journal* of *Meteorology*, 5(4):165–166.
- Martinez, D. O., Chen, S., Doolen, G. D., Kraichnan, R. H., Wang, L. P., and Zhou, Y. (1997). Energy spectrum in the dissipation range of fluid turbulence. *Journal of Plasma Physics*, 57:195–201.
- Maude, A. D. and Whitmore, R. L. (1958). A generalized theory of sedimentation. British Journal of Applied Physics, 9(12):477–482.
- Maury, B. (1999). Direct simulations of 2d fluid-particle flows in biperiodic domains. Journal of Computational Physics, 156(2):325–351.
- Maury, B. and Glowinski, R. (1997). Fluid-particle flow: a symmetric formulation. Comptes Rendus De L'Academie Des Sciences Serie I-Mathematique, 324(9):1079– 1084.
- Maxey, M. R. (1987). The motion of small sperical particles in a cellular flow field. *Physics of Fluids*, 30:1915.
- Maxey, M. R. (1993). The equation of motion for a small rigid sphere in a nonuniform or unsteady flow. In ASME-FED Intl. Symp. on Gas-Solid Flows, volume 166, pages 57–62.
- Maxey, M. R. and Corrsin, S. (1986). Gravitational settling of aerosol-particles in randomly oriented cellular-flow fields. *Journal of the Atmospheric Sciences*, 43(11):1112– 1134.
- Maxey, M. R. and Patel, B. K. (2001). Localized force representations for particles sedimenting in stokes flow. *International Journal of Multiphase Flow*, 27(9):1603–1626.

- Maxey, M. R. and Riley, J. J. (1983). Equation of motion for a small rigid sphere in a nonuniform flow. *Physics of Fluids*, 26:883–889.
- Mazzitelli, I. M., Lohse, D., and Toschi, F. (2003). On the relevance of the lift force in bubbly turbulence. *Journal of Fluid Mechanics*, 488:283–313.
- Meakin, P. and Vicsek, T. (1985). Internal structure of diffusion-limited aggregates. *Physical Review A*, 32(1):685–688.
- Medrano, R. O., Moura, A., Tel, T., Caldas, I. L., and Grebogi, C. (2008). Finite-size particles, advection, and chaos: A collective phenomenon of intermittent bursting. *Physical Review E*, 78(5):056206.
- Mei, R. W. and Adrian, R. J. (1992). Flow past a sphere with an oscillation in the free-stream velocity and unsteady drag at finite reynolds-number. *Journal of Fluid Mechanics*, 237:323–341.
- Mei, R. W., Klausner, J. F., and Lawrence, C. J. (1994). A note on the history force on a spherical bubble at finite reynolds-number. *Physics of Fluids*, 6(1):418–420.
- Michaelides, E. (1997). Review the transient equation of motion for particles, bubbles and droplets. *Journal of Fluids Engineering*, 119:233.
- Mietta, F., Maggi, F., and Winterwerp, J. C. (2008). Chapter 19 sensitivity to breakup functions of a population balance equation for cohesive sediments. In Tetsuya Kusuda, Hiroyuki Yamanishi, J. S. and Gailani, J. Z., editors, *Sediment and Ecohydraulics INTERCOH 2005*, volume 9 of *Proceedings in Marine Science*, pages 275 286. Elsevier.
- Mordant, N., Crawford, A. M., and Bodenschatz, E. (2004). Three-dimensional structure of the lagrangian acceleration in turbulent flows. *Physical Review Letters*, 93(21):214501.
- Nishikawa, T., Toroczkai, Z., and Grebogi, C. (2001). Advective coalescence in chaotic flows. *Physical Review Letters*, 87(3):038301.
- Oles, V. (1992). Shear-induced aggregation and breakup of polystyrene latex-particles. Journal of Colloid and Interface Science, 154(2):351–358.
- Oseen, C. (1910). Über die Stokessche Formel und über die verwandte Aufgabe in der Hydrodynamik. Arkiv fior Matematik, Astronomi och Fysik, 6(29):1.
- Oseen, C. (1927). Hydrodynamik. Leipzig.

- Ottino, J. (1989). The kinematics of mixing: stretching, chaos and transport. Cambridge University Press, Cambridge, U.K.
- Pandya, J. D. and Spielman, L. A. (1982). Floc breakage in agitated suspensions theory and data-processing strategy. *Journal of Colloid and Interface Science*, 90(2):517–531.
- Pantina, J. P. and Furst, E. M. (2005). Elasticity and critical bending moment of model colloidal aggregates. *Physical Review Letters*, 94(13):138301.
- Poisson, S. (1831). Mémoire sur les Mouvements simultanés d'un pendule et de l'air environnant. *Memoires de l'Academie des Sciences*, 9:521–523.
- Pope, S. (2008). *Turbulent Flows*. Cambridge University Press, Cambridge, UK, 5th edition.
- Pope, S. B. (1994). Lagrangian pdf methods for turbulent flows. Annual Review of Fluid Mechanics, 26:23–63.
- Pöschel, T. and Schwager, T. (2005). Computational Granular Dynamics. Springer, Berlin.
- Potanin, A. A. (1993). On the computer-simulation of the deformation and breakup of colloidal aggregates in shear-flow. *Journal of Colloid and Interface Science*, 157(2):399– 410.
- Prosperetti, A. (2007). Computational Methods for Multiphase Flows, chapter "Averaged equations for multiphase flow", pages 237–281. Cambridge University Press, Cambridge, UK.
- Pruppacher, H. R. and Klett, J. D. (1997). Microphysics of Clouds and Precipitation, volume 18. Kluwer Academic Publishers, Dordrecht, second edition.
- Pumir, A., Shraiman, B. I., and Chertkov, M. (2000). Geometry of lagrangian dispersion in turbulence. *Physical Review Letters*, 85(25):5324–5327.
- Reade, W. C. and Collins, L. R. (2000). Effect of preferential concentration on turbulent collision rates. *Physics of Fluids*, 12(10):2530–2540.
- Richardson, J. F. and Zaki, W. N. (1954). The sedimentation of a suspension of uniform spheres under conditions of viscous flow. *Chemical Engineering Science*, 3(2):65–73.
- Rubinow, S. I. and Keller, J. B. (1961). The transverse force on a spinning sphere moving in a viscous fluid. *Journal of Fluid Mechanics*, 11(3):447–459.

- Ruiz, J. and Izquierdo, A. (1997). A simple model for the break-up of marine aggregates by turbulent shear. Oceanologica Acta, 20(4):597.
- Saffman, P. G. (1965). Lift on a small sphere in a slow shear flow. Journal of Fluid Mechanics, 22:385–&.
- Saffman, P. G. and Turner, J. S. (1956). On the collision of drops in turbulent clouds. Journal of Fluid Mechanics, 1(1):16–30.
- Sandulescu, M., López, C., Hernández-García, E., and Feudel, U. (2007). Plankton blooms in vortices: the role of biological and hydrodynamic timescales. *Nonlinear Processes in Geophysics*, 14:443–454.
- Shaw, R. A. (2003). Particle-turbulence interactions in atmospheric clouds. Annual Review of Fluid Mechanics, 35:183–227.
- Sigurgeirsson, H. and Stuart, A. (2002). A model for preferential concentration. *Physics of Fluids*, 14(12):4352–4361.
- Sigurgeirsson, H., Stuart, A., and Wan, W.-L. (2001). Algorithms for particle-field simulations with collisions. *Journal of Computational Physics*, 172(2):766–807.
- Smoluchowski, M. (1917). Versuch einer mathematischen Theorie der Koagulationskinetik kolloider Lösungen. Zeitschrift für Physikalische Chemie, 92:129–168.
- Sommerer, J. C. and Ott, E. (1993). Particles floating on a moving fluid a dynamically comprehensible physical fractal. *Science*, 259(5093):335–339.
- Sonntag, R. C. and Russel, W. B. (1987a). Elastic properties of flocculated networks. Journal of Colloid and Interface Science, 116(2):485–489.
- Sonntag, R. C. and Russel, W. B. (1987b). Structure and breakup of flocs subjected to fluid stresses .2. theory. *Journal of Colloid and Interface Science*, 115(2):378–389.
- Sonntag, R. C. and Russel, W. B. (1987c). Structure and breakup of flocs subjected to fluid stresses .3. converging flow. *Journal of Colloid and Interface Science*, 115(2):390– 395.
- Spicer, P. and Pratsinis, S. (1996a). Coagulation and fragmentation: universal steadystate particle size distribution. AIChE Journal, 42:1612–1620.
- Spicer, P. T. and Pratsinis, S. E. (1996b). Shear-induced flocculation: The evolution of floc structure and the shape of the size distribution at steady state. *Water Research*, 30(5):1049–1056.

- Squires, K. D. and Eaton, J. K. (1991). Preferential concentration of particles by turbulence. *Physics of Fluids A*, 3(5):1169–1179.
- Sreenivasan, K. R. (1999). Fluid turbulence. Reviews of Modern Physics, 71(2):S383– S395.
- Stauffer, D. and Aharony, A. (1995). Perkolationstheorie. Wiley-VCH, Weinheim.
- Stock, M. (2006). Dla-nd. http://markjstock.org/dla-nd/.
- Stokes, G. (1851). On the effect of internal friction of fluids on the motion of a pendulum. Transactions of the Cambridge Philosophical Society, 9:8–106.
- Stolzenbach, K. D. and Elimelech, M. (1994). The effect of particle density on collisions between sinking particles - implications for particle aggregation in the ocean. *Deep-Sea Research*, 41(3):469–483.
- Sundaram, S. and Collins, L. R. (1997). Collision statistics in an isotropic particle-laden turbulent suspension .1. direct numerical simulations. *Journal of Fluid Mechanics*, 335:75–109.
- Taylor, G. (1934). The formation of emulsions in definable fields of flow. *Proceedings of the Royal Society A*, 146:501.
- Tél, T., DeMoura, A., Grebogi, C., and Károlyi (2005). Chemical and biological activity in open flows: a dynamical systems approach. *Physics Reports*, 413:91–196.
- Tél, T., Nishikawa, T., Motter, A. E., Grebogi, C., and Toroczkai, Z. (2004). Universality in active chaos. *Chaos*, 14(1):72–78.
- Ten Cate, A., Derksen, J. J., Portela, L. M., and Van Den Akker, H. E. A. (2004). Fully resolved simulations of colliding monodisperse spheres in forced isotropic turbulence. *Journal of Fluid Mechanics*, 519:233–271.
- Ten Cate, A. and Sundaresan, S. (2006). Analysis of unsteady forces in ordered arrays of monodisperse spheres. *Journal of Fluid Mechanics*, 552:257–287.
- Thomas, D., Judd, S., and Fawcett, N. (1999). Flocculation modelling: A review. Water Research, 33(7):1579–1592.
- Toschi, F. and Bodenschatz, E. (2009). Lagrangian properties of particles in turbulence. Annual Review of Fluid Mechanics, 41:375–404.

- Tucker III, C. L. and Moldenaers, P. (2002). Microstrucural evolution in polymer blends. Annual Review of Fluid Mechanics, 34(1):177–210.
- Vassileva, N., vandenEnde, D., Mugele, F., and Mellema, J. (2007). Fragmentation and erosion of two-dimensional aggregates in shear flow. *Langmuir*, 23(5):2352–2361.
- Verney, R., Lafite, R., Brun-Cottan, J. C., and Hir, P. L. (2010). Behaviour of a floc population during a tidal cycle: Laboratory experiments and numerical modelling. *Continental Shelf Research*, In Press, Corrected Proof:–.
- Vicsek, T., Family, F., and Meakin, P. (1990). Multifractal geometry of diffusion-limited aggregates. *Europhysics Letters*, 12(3):217–222.
- Vigil, R. D. and Ziff, R. M. (1989). On the stability of coagulation fragmentation population balances. Journal of Colloid and Interface Science, 133(1):257–264.
- Vilela, R. D. and Motter, A. E. (2007). Can aerosols be trapped in open flows? *Physical Review Letters*, 99(26):264101.
- Villermaux, E. (2007). Fragmentation. Annual Review of Fluid Mechanics, 39:419–446.
- Villermaux, E. and Bossa, B. (2009). Single-drop fragmentation determines size distribution of raindrops. *Nature Physics*, 5(9):697–702.
- Visser, A. W. (1997). Using random walk models to simulate the vertical distribution of particles in a turbulent water column. *Marine Ecology-Progress Series*, 158:275–281.
- Wang, L. P. and Maxey, M. R. (1993). Settling velocity and concentration distribution of heavy-particles in homogeneous isotropic turbulence. *Journal of Fluid Mechanics*, 256:27–68.
- Wang, L.-P., Wexler, A. S., and Zhou, Y. (1998). On the collision rate of small particles in isotropic turbulence. i. zero-inertia case. *Physics of Fluids*, 10(1):266–276.
- Wang, L. P., Wexler, A. S., and Zhou, Y. (2000). Statistical mechanical description and modelling of turbulent collision of inertial particles. *Journal of Fluid Mechanics*, 415:117–153.
- Wilkinson, M. and Mehlig, B. (2003). Path coalescence transition and its applications. *Physical Review E*, 68(4):040101.
- Wilkinson, M. and Mehlig, B. (2005). Caustics in turbulent aerosols. *Europhysics Letters*, 71(2):186–192.

- Wilkinson, M., Mehlig, B., and Bezuglyy, V. (2006). Caustic activation of rain showers. *Physical Review Letters*, 97(4):048501.
- Wilkinson, M., Mehlig, B., Ostlund, S., and Duncan, K. P. (2007). Unmixing in random flows. *Physics of Fluids*, 19(11):113303.
- Winterwerp, J. C. (1998). A simple model for turbulence induced flocculation of cohesive sediment. Journal of hydraulic research, 36:309.
- Witten, T. A. and Sander, L. M. (1981). Diffusion-limited aggregation, a kinetic critical phenomenon. *Physical Review Letters*, 47(19):1400–1403.
- Witten, T. A. and Sander, L. M. (1983). Diffusion-limited aggregation. *Physical Review* B, 27(9):5686–5697.
- Zahnow, J. C. and Feudel, U. (2008). Moving finite-size particles in a flow: A physical example of pitchfork bifurcations of tori. *Physical Review E*, 77(2):026215.
- Zahnow, J. C. and Feudel, U. (2009). What determines size distributions of heavy drops in a synthetic turbulent flow? *Nonlinear Processes In Geophysics*, 16(6):677–690.
- Zahnow, J. C., Maerz, J., and Feudel, U. (2010). Particle-based modelling of aggregation and fragmentation processes: Fractal-like aggregates. *submitted to Physica D*.
- Zahnow, J. C., Vilela, R. D., Feudel, U., and Tel, T. (2008). Aggregation and fragmentation dynamics of inertial particles in chaotic flows. *Physical Review E*, 77(5):055301.
- Zahnow, J. C., Vilela, R. D., Feudel, U., and Tel, T. (2009). Coagulation and fragmentation dynamics of inertial particles. *Physical Review E*, 80(2):026311.
- Zeidan, M., Xu, B. H., Jia, X., and Williams, R. A. (2007). Simulation of aggregate deformation and breakup in simple shear flows using a combined continuum and discrete model. *Chemical Engineering Research & Design*, 85(A12):1645–1654.
- Zhang, D. Z. and Prosperetti, A. (1994). Averaged equations for inviscid disperse 2-phase flow. Journal of Fluid Mechanics, 267:185–219.
- Zhou, Y., Wexler, A. S., and Wang, L. P. (1998). On the collision rate of small particles in isotropic turbulence. ii. finite inertia case. *Physics of Fluids*, 10(5):1206–1216.
- Zimmerman, J. T. F. (1986). The tidal whirlpool: A review of horizontal dispersion by tidal and residual currents. *Netherlands Journal of Sea Research*, 20(2-3):133–154.

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Erklärung

Hiermit erkläre ich, dass ich die vorliegende Dissertation selbstständig verfasst habe und keine anderen als die angegebenen Hilfsmittel und Quellen verwendet habe. Teile der Dissertation wurden bereits veröffentlicht bzw. sind zur Veröffentlichung eingereicht, wie an den entsprechenden Stellen angegeben. Die Dissertation hat weder in Teilen noch in ihrer Gesamtheit einer anderen wissenschaftlichen Hochschule zur Begutachtung in einem Promotionsverfahren vorgelegen.

Oldenburg, den 29. September 2010.

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