

INVESTIGATIONS ON THE SEDIMENTATION PROCESS OF POLYDISPERSE COLLOIDAL PARTICULATE SYSTEMS

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1. Introduction

Understanding the settling behaviour of suspensions of small particles, and how such suspensions can be manipulated to maximize a desired effect, is important for the successful formulation of chemical and biological products. In e.g. solid liquid separation flocculants are used to destabilize suspensions and to promote flocculation which yields a higher overall settling velocity of a suspension. The effects of present or prevented flocculation/aggregation can be studied experimentally, but such experiments are often time consuming and costly. This drawback can be overcome by means of numerical simulations, either in addition to experiments or replacing them. There are a number of approaches to solve particulate flow problems numerically. Each of them has its pros and cons, but a discussion would lead beyond the scope of this work. Yet we want to mention the method of our choice. Differing from most other numerical schemes, the Stokesian dynamics (SD) method (Durlofsky *et al.*, 1987) does not rely on a numerical grid. The fluid motion is implicitly incorporated into the system which is solved. This makes it an efficient method for the numerical simulation of particulate flows at low Reynolds number. SD and related methods which are based on the Oseen equations have successfully been applied to the simulation of clouds of non-colloidal particles settling in a quiescent fluid (Subramanian and Koch, 2008; Pignatelli *et al.*, 2011), as well as to the simulation of colloidal suspensions (Wagner and Brady, 2009; Harshe and Lattuada, 2012). (Binder *et al.*, 2006) have compared a Lattice Boltzmann method and SD on the example of agglomerates and aggregates. (Binder *et al.*, 2009) have investigated the drag force acting on settling aggregates of monodisperse particles. As a lot of works on aggregates, the mentioned works by Binder *et al.* do not consider inter-particle forces which lead to the build-up of aggregates. Widely accepted models for such forces arise from the DLVO theory (Derjaguin and Landau, 1941; Verwey, 1948), which we use for the simulations giving the results presented in this work.

2. Objective

To obtain the present results we go one step further than the previously mentioned works. We investigate the sedimentation behavior of polydisperse particulate systems when DLVO forces cannot be neglected. This work is organized as follows. We first give a short introduction into SD and how we model particle-particle interactions of non-hydrodynamic nature. After that we present results we have gained from our simulations. We discuss the settling of a spherical cloud of particles influenced by present DLVO forces.

3. Methods

Based on the integral expression for the fluid velocity field in Stokes flow (Durlofsky *et al.*, 1987) a linear relation between hydrodynamic force, hydrodynamic torque and the particles' translational and angular velocity can be found. This relation is given by

$$M \begin{pmatrix} F_H \\ T_H \end{pmatrix} = \begin{pmatrix} U_p \\ \omega_p \end{pmatrix}, \quad (1)$$

where F_H and T_H are vectors containing the components of the hydrodynamic force and torque on all N_p particles. U_p and ω_p are vectors containing the translational and angular velocities of all particles, respectively. The so-called mobility matrix M depends only on the particle positions and the particle radii. Together with the Langevin equation,

$$m_p \frac{d}{dt} U_p = F_H + F_G + F_I + F_B, \quad (2)$$

and the angular momentum conservation equation,

$$I_p \frac{d}{dt} \omega_p = T_H, \quad (3)$$

a set of ordinary differential equations can be constructed. Here, F_H denotes the hydrodynamic force, F_G the effective gravitational force, F_I inter-particle forces such as van der Waals or electrostatic forces, and F_B the Brownian force resulting from thermal motion of the fluid (Ichiki and Hayakawa, 1995). In this work we only consider relatively large colloids of diameter 500 nm and larger, where Brownian motion can still be neglected. In case of a small Reynolds number and particles with mass density of a similar order of magnitude as the fluid mass density, by a dimensional analysis we find that the left hand side of equations (2) and (3) can be assumed to be negligibly small compared to the terms on the right hand side. Thus substitution of equation (1) into equations (2) and (3) yields a linear relation between inter-particle forces and gravity on the one hand, and particle velocities on the other hand. We use the potentials given in (Hamaker, 1937), (Feke *et al.*, 1984) and (Hogg *et al.*, 1966) to model DLVO forces, since these potentials are widely accepted and valid for polydisperse systems.

4. Results

Initial configuration in our simulations of particle clouds is a spherical cloud in an infinitely extended quiescent fluid. Particles are placed randomly inside such a cloud whose radius is computed such that a given volume fraction inside it is met. In the present case the clouds are comprised of 2000 particles. Volume fractions are $\phi = 0.01$, $\phi = 0.025$ and $\phi = 0.05$. The particle mass density is 1190 kg/m^3 , whereas the fluid mass density is 1000 kg/m^3 . The system is at a temperature of 293 K. Using the mean particle radius (cf. figure 1) and the corresponding Stokes velocity as characteristic scales yields a Reynolds number and Stokes number of order of 10^{-8} , which is reasonably small for our assumption (see section 3). Note that the system is fully polydisperse. The particle radii are distributed according to a log-normal distribution with resulting expected value and standard deviation given in figure 1.

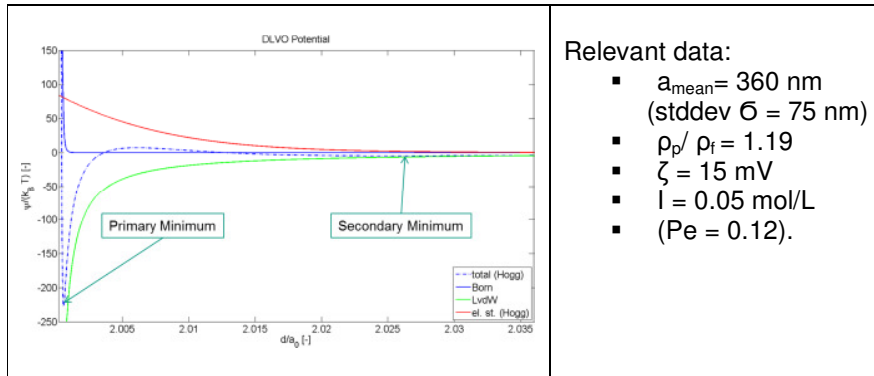


Figure 1: Left: Different potentials taken into account in our simulations. Right: Data relevant for the simulation.

When inter-particle forces such as the van der Waals force and electrostatic forces are non-negligible, aggregation of particles can occur. As we neglect the influence of Brownian motion, particles tend to reach the secondary minimum and not the primary minimum. A factor which is of interest for technical applications is the number of build aggregates. Figure 2 shows the number of particles over time, when the particle cloud initially contained 2000 particles. The number of particle clusters decreases with time and tends to a constant value. This is due to the fact that after a certain time of aggregation inter-particle distances become too large for the relatively short-ranged van der Waals force to have an effect. The build aggregates settle and as big aggregates settle faster than small aggregates, particle clusters diverge more and more, leaving no chance for any more aggregation.

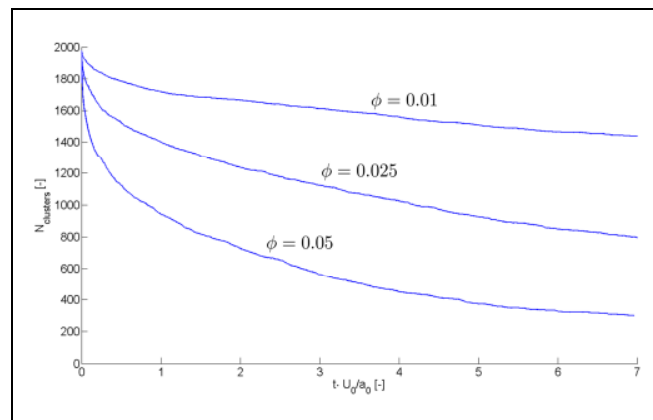


Figure 2: Number of particle clusters (aggregates) over time. As aggregates become bigger, the total number of clusters decreases.

We have implemented a cluster detection algorithm which acts on the particle data gained from our simulations. Due to the fact that the simulations provide us with all necessary data on the particles, we have access to all information on the aggregates. This can be e.g. the Feret diameter, radius of gyration, or the number of particles per aggregate.

5. Conclusion

We have implemented a highly parallel algorithm with which we are able to simulate the settling process of large numbers of particles suspended in a fluid. These simulations include hydrodynamic interactions between particles, as well as other non-hydrodynamic inter-particle interactions. Results gained from the simulation of settling particle clouds show the capability of our implementation. Our code is based on the open source code RYUON, therefore we want to thank K. Ichiki for laying the foundation to our code.

REFERENCES

- Binder, C., Feichtinger, C., Schmid, H. J., Thürey, N., Peukert, W. and Rüde, U., 2006: Simulation of the hydrodynamic drag of aggregated particles; *J. C. I. S.* **301**, 155-167.
- Binder, C., Hartig, M. A. J. and Peukert, W., 2009: Structural dependent drag force and orientation prediction for small fractal aggregates; *J. C. I. S.* **331**, 243-250.
- Derjaguin, B. V. and Landau, L., 1941; *Acta Physicochim. (URSS)* **14**, 633-662.
- Durlofsky, L., Brady, J.F. and Bossis, G., 1987: Dynamic simulation of hydrodynamically interacting particles, *J. F.M.* **180**, 21-49.
- Feke, D. L., Prabhu, N. D., Mann J. A. (Jr) and Mann J. A., 1984: A Formulation of the Short-Range Repulsion between Spherical Colloidal Particles; *J. Phys. Chem.* **88**, 5735-5739.
- Hamaker, H. C., 1937: the London-Van der Waals attraction between spherical particles; *Physica IV* **10**, 1058-1072.
- Harshe, Y. M. and Lattuada, M., 2012: Breakage Rate of Colloidal Aggregates in Shear Flow through Stokesian Dynamics; *Langmuir* **28**, 283-292.
- Hogg, R., Healy, T. W. and Fuerstenau, D. W., 1966: Mutual Coagulation of Colloidal Dispersions; *Trans. Faraday. Soc.* **62**, 1638-1651.
- Ichiki, K. and Hayakawa, H., 1995: Dynamical simulation of fluidized beds: Hydrodynamical interacting granular particles; *Phys. Rev. E* **52**, 658-670.
- Pignatelli, F., Nicolas, M. and Guazzelli, E., 2011: A falling cloud of particles at small but finite Reynolds-number; *J. F. M.* **671**, 34-51.
- Subramanian, G. and Koch, D. L., 2008: Evolution of clusters of sedimenting low-Reynolds-number particles with Oseen interactions; *J.F.M.* **603**, 63-100.
- Verwey, E. J. W., 1948: *Theory of the stability of lyophobic colloids*; Elsevier, Amsterdam.
- Wagner, N. J. and Brady, J. F., 2009: Shear thickening in colloidal dispersions; *Phys. Today* **62**, 27-32.