

Simulation of particle suspensions at the Institute for Computational Physics

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Simulations of Particle Suspensions at the Institute for Computational Physics

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Summary. In this report we describe some of our projects related to the simulation of particle-laden flows. We give a short introduction to the topic and the methods used, namely the Stochastic Rotation Dynamics and the lattice Boltzmann method. Then, we show results from our work related to the behaviour of claylike colloids in shear flow as well as structuring effects of particles in the vicinity of rigid walls.

1 Introduction

Simulating the flow of suspensions is an extremely difficult and demanding problem. Suspensions are mixtures of fluid and granular materials, and each component alone is a challenge to numerical modelers. When they are combined in a suspension, neither component can be neglected, so all the difficulties of both fluids and grains must be solved, in addition to the new problem of describing the interaction between the two. These suspensions are ubiquitous in our daily life, but are not well understood due to their complexity. During the last twenty years, various simulation methods have been developed in order to model these systems. Due to varying properties of the solved particles and the solvents, one has to choose the simulation method properly in order to use the available compute resources most effectively with resolving the system as well as needed. Various techniques for the simulation of particle suspensions have been implemented at the Institute for Computational Physics allowing us to study the properties of clay-like systems, where Brownian motion is important, more macroscopic particles like glass spheres or fibers solved in liquids, or even the pneumatic transport of powders in pipes.

Computer simulation methods are indispensable for such many-particle systems, for the inclusion of inertia effects (Reynolds numbers > 1) and Brownian motion (Peclet number of order 1). These systems often contain a large number of important time scales which differ by many orders of magnitude, but nevertheless have to be resolved by the simulation, leading to a large numerical effort. However, simulations have the potential to increase our knowl-

edge of elementary processes and to enable us to find the aforementioned relations from simulations instead of experiments.

In this paper we will present two approaches which have been applied during the last year: A combined Molecular Dynamics and Stochastic Rotation Dynamics method has been applied to the simulation of claylike colloids and the lattice-Boltzmann method was used as a fluid solver to study the behaviour of glass spheres in creeping shear flows.

2 Simulation of Claylike Colloids: Stochastic Rotation Dynamics

We investigate properties of dense suspensions and sediments of small spherical Al_2O_3 particles by means of a combined Molecular Dynamics (MD) and Stochastic Rotation Dynamics (SRD) simulation. Stochastic Rotation Dynamics is a simulation method developed by Malevanets and Kapral [17, 18] for a fluctuating fluid. The work this chapter is dealing with is presented in more detail in references [7, 6].

We simulate claylike colloids, for which in many cases the attractive Van-der-Waals forces are relevant. They are often called “peloids” (Greek: clay-like). The colloidal particles have diameters in the range of some nm up to some μm . The term “*peloid*” originally comes from soil mechanics, but particles of this size are also important in many engineering processes. Our model systems of Al_2O_3 -particles of about half a μm in diameter suspended in water are often used ceramics and play an important role in technical processes. In soil mechanics [22] and ceramics science [20], questions on the shear viscosity and compressibility as well as on porosity of the microscopic structure which is formed by the particles, arise [26, 16]. In both areas, usually high volume fractions ($\Phi > 20\%$) are of interest. The mechanical properties of these suspensions are difficult to understand. Apart from the attractive forces, electrostatic repulsion strongly determines the properties of the suspension. Depending on the surface potential, one can either observe formation of clusters or the particles are stabilized in suspension and do sediment only very slowly. The surface potential can be adjusted by the *pH*-value of the solvent. Within Debye-Hückel theory one can derive a so-called *2pK* charge regulation model which relates the simulation parameters with the *pH*-value and ionic strength *I* adjusted in the experiment. In addition to the static interactions hydrodynamic effects are also important for a complete description of the suspension. Since typical Peclet numbers are of order one in our system, Brownian motion cannot be neglected.

The colloidal particles are simulated with molecular dynamics (MD), whereas the solvent is modeled with stochastic rotation dynamics (SRD).

In the MD part of our simulation we include effective electrostatic interactions and van der Waals attraction, a lubrication force and Hertzian contact

forces. In order to correctly model the statics and dynamics when approaching stationary states, realistic potentials are needed. The interaction between the particles is described by DLVO theory [9, 24, 16]. If the colloidal particles are suspended in a solvent, typically water, ions move into solution, whereas their counter ions remain in the particle due to a different resolvability. Thus, the colloidal particle carries a charge. The ions in solution are attracted by the charge on the particles and form the electric double layer. It has been shown (see [24]), that the resulting electrostatic interaction between two of these particles can be described by an exponentially screened Coulomb potential

$$V_{\text{Coul}} = \pi \varepsilon_r \varepsilon_0 \left[\frac{2 + \kappa d}{1 + \kappa d} \cdot \frac{4k_{\text{B}}T}{ze} \tanh \left(\frac{ze\zeta}{4k_{\text{B}}T} \right) \right]^2 \times \frac{d^2}{r} \exp(-\kappa[r - d]), \quad (1)$$

where d denotes the particle diameter and r is the distance between the particle centers. e is the elementary charge, T the temperature, k_{B} the Boltzmann constant, and z is the valency of the ions of added salt. Within DLVO theory one assumes linear screening, mainly by one species of ions with valency z (e.g. $z = +1$ for NH_4^+). The first fraction in equation 1 is a correction to the original DLVO potential, which takes the surface curvature into account and is valid for spherical particles [2]. The effective surface potential ζ is the electrostatic potential at the border between the diffuse layer and the compact layer, it may therefore be identified with *the* ζ -potential. It includes the effect of the bare charge of the colloidal particle itself, as well as the charge of the ions in the Stern layer, where the ions are bound permanently to the colloidal particle. In other words, DLVO theory uses a renormalized surface charge. This charge can be related to the pH value of the solvent within Debye-Hückel theory [6]. ε_0 is the permittivity of the vacuum, ε_r the relative dielectric constant of the solvent. κ is the inverse Debye length defined by $\kappa^2 = 8\pi\ell_B I$, with the ionic strength I and the Bjerrum length ℓ_B . We use $\varepsilon_r=81$ for water, which implies $\ell_B = 7 \text{ \AA}$.

The Coulomb term of the DLVO potential competes with the attractive van der Waals term

$$V_{\text{vdW}} = -\frac{A_{\text{H}}}{12} \left[\frac{d^2}{r^2 - d^2} + \frac{d^2}{r^2} + 2 \ln \left(\frac{r^2 - d^2}{r^2} \right) \right]. \quad (2)$$

$A_{\text{H}} = 4.76 \cdot 10^{-20} \text{ J}$ is the Hamaker constant [8] which involves the polarizability of the particles.

For the integration of the translational motion we utilize a velocity Verlet algorithm [1] to update the velocity and position of particle i according to the equations

$$\mathbf{x}_i(t + \delta t) = \mathbf{x}_i(t) + \delta t \mathbf{v}_i(t) + \delta t^2 \frac{F_i(t)}{m}, \quad (3)$$

$$\mathbf{v}_i(t + \delta t) = \mathbf{v}_i(t) + \delta t \frac{F_i(t) + F_i(t + \delta t)}{2m}. \quad (4)$$

For the rotation, a simple Euler algorithm is applied:

$$\omega_i(t + \delta t) = \omega_i(t) + \delta t \mathbf{T}_i, \quad (5)$$

$$\vartheta_i(t + \delta t) = \vartheta_i(t) + F(\vartheta_i, \omega_i, \delta t), \quad (6)$$

where $\omega_i(t)$ is the angular velocity of particle i at time t , \mathbf{T}_i is the torque exerted by non central forces on the particle i , $\vartheta_i(t)$ is the orientation of particle i at time t , expressed by a quaternion, and $F(\vartheta_i, \omega_i, \delta t)$ gives the evolution of ϑ_i of particle i rotating with the angular velocity $\omega_i(t)$ at time t .

The concept of quaternions [1] is often used to calculate rotational motions in simulations, because the Euler angles and rotation matrices can easily be derived from quaternions. Using Euler angles to describe the orientation would give rise to singularities for the two orientations with $\vartheta = \pm 90^\circ$. The numerical problems related to this fact and the relatively high computational effort of a matrix inversion can be avoided using quaternions.

The Stochastic Rotation Dynamics method (SRD) introduced by Malevanets and Kapral [17, 18] is a promising tool for a coarse-grained description of a fluctuating solvent, in particular for colloidal and polymer suspensions. The method is also known as “Real-coded Lattice Gas” [10] or as “multi-particle-collision dynamics” (MPCD) [23]. It can be seen as a “hydrodynamic heat bath”, whose details are not fully resolved but which provides the correct hydrodynamic interaction among embedded particles [15]. SRD is especially well suited for flow problems with Peclet numbers of order one and Reynolds numbers on the particle scale between 0.05 and 20 for ensembles of many particles. The method is based on so-called fluid particles with continuous positions and velocities. Each time step is composed of two simple steps: One streaming step and one interaction step. In the streaming step the positions of the fluid particles are updated as in the Euler integration scheme known from Molecular Dynamics simulations:

$$\mathbf{r}_i(t + \tau) = \mathbf{r}_i(t) + \tau \mathbf{v}_i(t), \quad (7)$$

where $\mathbf{r}_i(t)$ denotes the position of the particle i at time t , $\mathbf{v}_i(t)$ its velocity at time t and τ is the time step used for the SRD simulation. After updating the positions of all fluid particles they interact collectively in an interaction step which is constructed to preserve momentum, energy and particle number. The fluid particles are sorted into cubic cells of a regular lattice and only the particles within the same cell are involved in the interaction step. First, their mean velocity $\mathbf{u}_j(t') = \frac{1}{N_j(t')} \sum_{i=1}^{N_j(t')} \mathbf{v}_i(t)$ is calculated, where $\mathbf{u}_j(t')$ denotes the mean velocity of cell j containing $N_j(t')$ fluid particles at time $t' = t + \tau$. Then, the velocities of each fluid particle in cell j are updated as:

$$\mathbf{v}_i(t + \tau) = \mathbf{u}_j(t') + \mathbf{\Omega}_j(t') \cdot [\mathbf{v}_i(t) - \mathbf{u}_j(t')]. \quad (8)$$

$\mathbf{\Omega}_j(t')$ is a rotation matrix, which is independently chosen randomly for each time step and each cell. The mean velocity $\mathbf{u}_j(t)$ in the cell j can be seen

as streaming velocity of the fluid at the position of the cell j at the time t , whereas the difference $[\mathbf{v}_i(t) - \mathbf{u}_j(t')]$ entering the interaction step can be interpreted as a contribution to the thermal fluctuations.

To couple the two parts of the simulation, MD on the one hand and SRD on the other one, the colloidal particles are sorted into the SRD boxes and their velocities are included in the rotation step. This technique has been used to model protein chains suspended in a liquid [4, 27]. Since the mass of the fluid particles is much smaller than the mass of the colloidal particles, one has to use the mass of each particle – colloidal or fluid particle – as a weight factor when calculating the mean velocity.

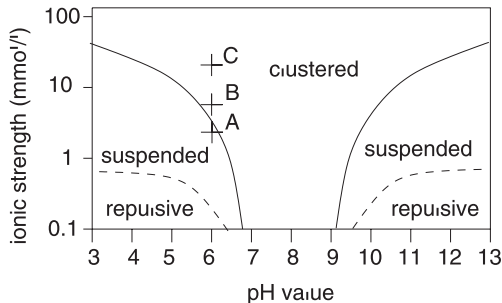


Fig. 1. Schematic phase diagram for volume fraction $\Phi = 35\%$ in terms of pH -value and ionic strength involving three different phases: a clustering regime due to van der Waals attraction, stable suspensions where the charge of the colloidal particles prevents clustering, and a repulsive structure for further increased electrostatic repulsion. This work concentrates on state A ($pH = 6$, $I = 3$ mmol/l) in the suspended phase, state B ($pH = 6$, $I = 7$ mmol/l) close to the phase border but already in the clustered phase, and state C ($pH = 6$, $I = 25$ mmol/l) in the clustered phase [6]

Depending on the experimental conditions, one can obtain three different phases in a Al_2O_3 suspension: A clustered region, a suspended phase, and a repulsive structure. These phases can be reproduced in the simulations and we can quantitatively relate interaction potentials to certain experimental conditions. A schematic picture of the phase diagram is shown in Fig. 1. Close to the isoelectric point ($pH = 8.7$), the particles form clusters for all ionic strengths since they are not charged. At lower or higher pH values one can prepare a stable suspension for low ionic strengths because of the charge, which is carried by the colloidal particles. At even more extreme pH values, one can obtain a repulsive structure due to very strong electrostatic potentials (up to $\zeta = 170$ mV for $pH = 4$ and $I = 1$ mmol/l, according to our model). The repulsive structure is characterized by an increased shear viscosity. In the following we focus on three states: State A ($pH = 6$, $I = 3$ mmol/l) is in the suspended phase, state B ($pH = 6$, $I = 7$ mmol/l) is a point already in

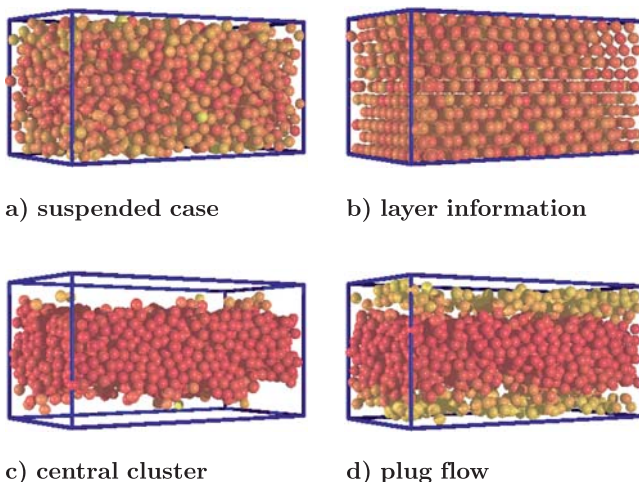


Fig. 2. Images of four different cases. For better visibility we have chosen smaller systems than we usually use for the calculation of the viscosity. The *colors* denote velocities: *Dark particles* are slow, *bright ones* move fast. The potentials do not correspond exactly to the cases *A–C* in Fig. 1, but they show qualitatively the differences between the different states: **a)** Suspension like in state *A*, at low shear rates. **b)** Layer formation, which occurs in the repulsive regime, but also in the suspension (state *A*) at high shear rates. **c)** Strong clustering, like in state *C*, so that the single cluster in the simulation is deformed. **d)** Weak clustering close to the phase border like in state *B*, where the cluster can be broken into pieces, which follow the flow of the fluid (plug flow)

the clustered phase but still close to the phase border, and state *C* ($pH = 6$, $I = 25 \text{ mmol/l}$) is located well in the clustered phase.

Some typical examples for the different phases are shown in Fig. 2a)–d). These examples are meant to be only illustrative and do not correspond exactly to the cases *A–C* in Fig. 1 denoted by uppercase letters. In the suspended case (a), the particles are mainly coupled by hydrodynamic interactions. One can find a linear velocity profile and a slight shear thinning. If one increases the shear rate $\dot{\gamma} > 500/\text{s}$, the particles arrange in layers. The same can be observed if the Debye-screening length of the electrostatic potential is increased (b), which means that the solvent contains less ions ($I < 0.3 \text{ mmol/l}$) to screen the particle charges. On the other hand, if one increases the salt concentration, electrostatic repulsion is screened even more and attractive van der Waals interaction becomes dominant ($I > 4 \text{ mmol/l}$). Then the particles form clusters (c), and viscosity rises. A special case, called “plug flow”, can be observed for high shear rates, where it is possible to tear the clusters apart and smaller parts of them follow with the flow of the solvent (d). This happens in our simulations for $I = 25 \text{ mmol/l}$ (state *C*) at a shear rate of $\dot{\gamma} > 500/\text{s}$. However, as long as there are only one or two big clusters in the

system, it is too small to expect quantitative agreement with experiments. In these cases we have to focus on state B ($I = 7$ mmol/l) close to the phase border.

Within this project we have investigated many details of the phase diagram and also compared our simulations to experimental data of a group in Karlsruhe. Furthermore, a large number of simulations has been carried out to investigate the properties of the simulation method and to improve it where needed. We are currently studying the dependence of autocorrelation functions, structure factors, and the fractality of particle clusters. Until now, two papers have been published describing the results of this projects in much greater detail than possible within this report [7, 6].

3 Transport Phenomena and Structuring in Suspensions: Lattice-Boltzmann Simulations

The object of this project is a theory describing the influence of solid boundary walls in the creeping shear flow of suspensions. To analyze the various effects, the results of theories, simulations, and experiments are compared.

We are interested in structuring effects which might occur in the solid fraction of the suspension. Such effects are known from dry granular media resting on a plane surface or gliding down an inclined chute [21, 25]. In addition, the wall causes a demixing of the solid and fluid components which might have an unwanted influence on the properties of the suspension. Near the wall one finds a thin lubrication layer which contains almost no particles and causes a so-called “pseudo wall slip”. We expect structuring close to a rigid wall at much smaller concentrations than in granular media because of long-range hydrodynamic interactions. In [11], we study these effects by the means of particle volume concentrations versus distance to the wall.

We model the fluid in by means of a lattice-Boltzmann algorithm. The lattice-Boltzmann method is a simple scheme for simulating the dynamics of fluids. By incorporating solid particles into the model fluid and imposing the correct boundary condition at the solid/fluid interface, colloidal suspensions can be studied. Pioneering work on the development of this method has been done by Ladd et al. [12, 13, 14] and we use their approach to model sheared suspensions near solid walls.

The lattice-Boltzmann (hereafter LB) simulation technique which is based on the well-established connection between the dynamics of a dilute gas and the Navier-Stokes equations [3]. We consider the time evolution of the one-particle velocity distribution function $n(\mathbf{r}, \mathbf{v}, t)$, which defines the density of particles with velocity \mathbf{v} around the space-time point (\mathbf{r}, t) . By introducing the assumption of molecular chaos, i.e. that successive binary collisions in a dilute gas are uncorrelated, Boltzmann was able to derive the integro-differential equation for $n(\mathbf{r}, \mathbf{v}, t)$ named after him [3]. The LB technique arose from the realization that only a small set of discrete velocities is necessary to simulate

the Navier-Stokes equations [5]. Much of the kinetic theory of dilute gases can be rewritten in a discretized version. The time evolution of the distribution functions n is described by a discrete analogue of the Boltzmann equation [14]:

$$n_i(\mathbf{r} + \mathbf{c}_i \Delta t, t + \Delta t) = n_i(\mathbf{r}, t) + \Delta_i(\mathbf{r}, t), \quad (9)$$

where Δ_i is a multi-particle collision term. Here, $n_i(\mathbf{r}, t)$ gives the density of particles with velocity \mathbf{c}_i at (\mathbf{r}, t) . In our simulations, we use 19 different discrete velocities \mathbf{c}_i .

To simulate the hydrodynamic interactions between solid particles in suspensions, the lattice-Boltzmann model has to be modified to incorporate the boundary conditions imposed on the fluid by the solid particles. Stationary solid objects are introduced into the model by replacing the usual collision rules at a specified set of boundary nodes by the “link-bounce-back” collision rule [19]. When placed on the lattice, the boundary surface cuts some of the links between lattice nodes. The fluid particles moving along these links interact with the solid surface at boundary nodes placed halfway along the links. Thus, a discrete representation of the surface is obtained, which becomes more and more precise as the surface curvature gets smaller and which is exact for surfaces parallel to lattice planes.

The particle position and velocity are calculated using Newton’s equations in a similar manner as in the section on SRD simulations. However, particles do not feel electrostatic interactions, but behave like hard spheres in the case presented in this section.

The purpose of our simulations is the reproduction of rheological experiments on computers. We simulate a representative volume element of the experimental setup and compare our calculations with experimentally accessible data, i.e. density profiles, time dependence of shear stress and shear rate. We also get experimentally inaccessible data from our simulations like translational and rotational velocity distributions, particle-particle and particle-wall interaction frequencies. The experimental setup consists of a rheoscope with two spherical plates. The upper plate can be rotated either by exertion of a constant force or with a constant velocity, while the complementary value is measured simultaneously. The material between the rheoscope plates consist of glass spheres suspended in a sugar-water solution. The radius of the spheres varies between 75 and 150 μm .

We are currently investigating the occurrence of non-Gaussian velocity distributions of particles for higher particle densities and higher shear rates. For this, improvements of the method are mandatory in order to prevent instabilities of the simulation. By utilizing an implicit scheme for the update of the particle velocities [14, 19] we are able to overcome artefacts caused by numerical inaccuracies at high volume fractions or shear rates. Figure 3 shows a snapshot of a system containing 1536 particles after 28000 timesteps.

t = 28000

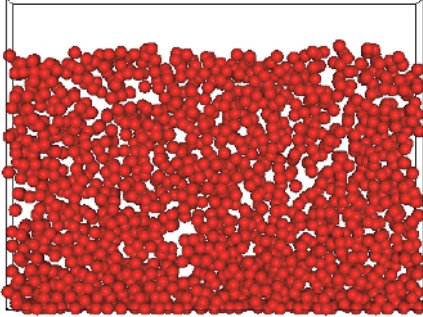


Fig. 3. A snapshot of a suspension with 1536 spheres after 28000 timesteps used to gain statistics of particle velocity distributions

4 Conclusions

We have reviewed two of the projects at the Institute for Computational Physics concerning particle suspensions. Both projects have developed simulations that can be directly compared to experiments. In this way, our numerical work contributes to answering important questions in both physics and engineering. Until now, a number of papers have been published describing the applied models and the results in much more detail [7, 6, 11]. The reader is referred to those in order to gain a better understanding of the implementation details.

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