



Hydrodynamic effects on the aggregation of nanoparticles in porous media

Ngoc H. Pham, Dimitrios V. Papavassiliou *

School of Chemical, Biological, and Materials Engineering, University of Oklahoma, Norman, OK 73019-1004, United States

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ABSTRACT

The aggregation of spherical nanoparticles as they propagate through porous media is explored using lattice Boltzmann simulations and tracking of the trajectories of individual particles. The porous media are modeled as periodic arrays of spheres in different packing configurations. The effects of interparticle interactions on particle aggregation are treated through a single aggregation probability upon particle collision. Fast aggregation is represented by an aggregation probability with values close to one, and slow aggregation is represented by lower aggregation probability. An algorithm that accounts for the different time scales between hydrodynamics and Brownian motion is devised and validated. It is found that primary particle size, initial particle concentration, injection flow rate, and aggregation probability significantly impact the dynamics of the aggregation. Calculations of the transient mean size of the aggregates show that the aggregation rate is high near the entrance of the porous medium, yet dramatically decreases when moving farther downstream. The reason is that the growth of larger aggregates slows the aggregation process. It is also found that the bigger aggregates are formed in the pore space near the pore matrix surface, where particle residence time is long enough to allow the aggregates to grow. An empirical correlation based on measurable parameters is proposed for the prediction of the mean aggregate size.

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1. Introduction

Aggregation of suspended particles in a solvent can have detrimental effects on a host of environmental and industrial processes. In other cases, as in water treatment, aggregation is desirable and is an important step towards improving dewatering of suspensions by gravitational sedimentation or membrane filtration [1–3]. Since the pioneering work by Smoluchowski [4], extensive efforts have been made to understand the stability of suspensions, as well as the kinetics of the aggregation process. Particle properties (e.g., size and shape) and fluid properties, such as the ionic strength and the pH of the suspensions affect the process [5–9]. It has also been noted that aggregation can transition from a reaction-limited to a diffusion-limited process by adding salt to the suspensions [5,10,11]. The salt concentration at which the transition happens (the *critical coagulation concentration*) can be controlled by adjusting the pH of the suspension, or by coating the suspended particles [5,12–14].

Aggregation of suspended particles is also a concern for the stability of dispersed systems used for novel enhanced oil recovery

(EOR) applications. Such applications include the injection of nanoparticle suspensions into hydrocarbon reservoirs to alter the mobility ratio between oil and displacing fluids, and to change the wettability of rock surfaces at oil-rock interfaces [15–17]. However, many researchers have experimentally observed that particle size distributions change in effluent samples when nanofluids go through porous media, indicating that aggregation occurs in porous media columns [18–22]. In such cases, aggregation of the nanoparticles is undesired, since size changes can potentially lead to clogging of the porous medium and to failure of the dispersion process. The dispersing systems contain mixtures of aggregates and primary particles, where the former might give rise to pore clogging, surface attachment, and surface sedimentation, reducing the mobility of the suspensions. Particle retention profiles (retention of particles as a function of location in the porous medium) of such cases usually exhibit an exponential decay from inlet to outlet [22,23].

Similar to aggregation in stationary systems, initial particle concentration and solution pH are among the factors affecting particle aggregation in porous media. Raychoudhury et al. [19] showed that higher particle concentration causes larger average aggregate size. Although higher particle concentration results in more deposition, it simultaneously results in the formation of large aggregates, which might reduce the overall single collector efficiency of the

* Corresponding author.

E-mail address: dvpapava@ou.edu (D.V. Papavassiliou).

process [19]. (Note that a *single collector* is one of the spheres making up the porous medium, and its *efficiency* is defined as the percent of nanoparticles colliding with it that adsorb on its surface.) The effects of pH on particle aggregation, as well as on the attachment of aggregates to the collector surface, have been discussed in Guzman et al. [20] and Solovitch et al. [18], but with contradicting results. Focusing on TiO₂ nanoparticles, Guzman et al. [20] reported that the percent of particles transported through porous structures, fabricated by packing spheres in simple cubic arrays, was at least 80% for all the pH considered, including pH values that resulted in attractive interactions between the particles and the sphere surface. In contrast, Solovitch et al. [18] found zero breakthrough of the same nanoparticle type through silica sand columns at pH of attractive interactions between the particles and the silica sands, yet significant breakthroughs were observed at pH of repulsive interactions.

Other than aggregation, particles in porous media are susceptible to flow-induced disaggregation, a process in which aggregated particles might be fragmented because of shear stresses. The breakup of aggregates due to shear has been discussed as a conceptual model [21], yet no mean size data before and after injection were given. Such measurements reporting aggregate breakup were conducted later [24], when effective particle diameters in the influent were found to be larger than those in the effluent. Physically, disaggregation of an aggregate would occur when the exerted forces due to shear were larger than the force required to breakup the aggregate [25]. It is important to note that disaggregation is not always possible, but there exists a critical shear rate, beyond which the aggregates are breakable [26,27]. The breakup of aggregates is often modeled as an exponential decay, controlled by a disaggregation rate constant [28,29]. Disaggregation in elongational flows would result in smaller aggregate fragments than in shear flows [30].

In this work, irreversible aggregation of nanoparticles in the pore space of ideally-packed and randomly-packed beds with spheres is numerically investigated. Pulse releases of the nanoparticles, represented by passive particles, are conducted by uniformly spreading particles at the inlet of arrays of periodically packed spheres. Aggregation dynamics are explored by monitoring the changes in particle size and the positions of the aggregates over time. Irreversible aggregation among the nanoparticles is modeled by assigning an aggregation probability (p_a) that takes values between zero and one, controlling successful aggregation over the total number of collisions between particle pairs in every simulation time step. The multiscale issue that rises from the disparity between the simulation timescale and the timescale associated with the particle Brownian motion is resolved by applying a collision prediction model. Failure to account for the timescale differences might lead to an underestimation of particle collisions. In our consideration, particle retention by pore surface attachment, sedimentation, and pore clogging are ignored. The focus is instead on the transient size change of the nanoparticles with changes of the primary particle size (D_0), initial particle concentration, aggregation probability (p_a), and pore velocity (u_p) (see Table 1 for details). An empirical correlation combining the effects of these parameters to predict the final mean size of aggregates (denoted as D_m hereafter) is also proposed.

2. Numerical implementation

2.1. Lattice Boltzmann method

In this work, the lattice Boltzmann method (LBM) for single-phase, Newtonian, incompressible fluids was employed to drive water at room temperature with viscosity of 0.001 Pa s through

periodic arrays of spheres packed in different configurations. The pore velocity u_p was set to be between 4.76×10^{-4} cm/s and 7.96×10^{-2} cm/s. The use of the LBM helped to create a steady, on-lattice velocity field, which is used to advance nanoparticles in the pore space by the Lagrangian particle tracking (LPT) algorithm, presented further below. The LBM algorithm implemented is not presented extensively herein, as it has been described in our prior work [31–33]. In brief, the employed LBM scheme was a D3Q15 method (*i.e.*, using 3 dimensions in space and 15 velocity lattice vectors) that utilized the Bhatnagar, Gross and Crook approximation for the collision operator. Simple cubic (SC), body centered cubic (BCC), and face centered cubic (FCC) arrays of rigid, impermeable spheres were generated computationally to model ideally packed beds with spheres, while practical packed beds were represented by packing the spheres in a random manner (see Fig. 1), following a modified Lubachevsky-Stillinger algorithm [34]. The porous bed porosities were 0.49, 0.33, 0.27, and 0.35 for the SC, BCC, FCC and the randomly packed arrangements, respectively. Periodic boundary conditions in X, Y, and Z directions were applied. The size of one periodic simulation unit was $375 \times 375 \times 375 \mu\text{m}^3$, and was the same for all packing configurations considered. In each direction, the periodic simulation unit was discretized into 101 equally-spaced mesh points, resulting in a total number of $101 \times 101 \times 101$ mesh points. The diameters of the spheres composing the SC, BCC, and FCC configuration were 375, 325, and 265 μm , respectively, while that of the spheres in the randomly packed configuration was 150 μm .

2.2. Lagrangian particle tracking with particle-particle interaction

As mentioned already, the release of a number of nanoparticles into the flow field was implemented with the LPT algorithm. These particles would ride on the fluid to advance their positions in time due to convection. While the fluid velocity was only computed on-lattice with the LBM, the particles were released randomly in the pore space at the inlet plane of the computed domain. In the rare event when the particles were on-lattice, their velocity was equal to the fluid velocity at those lattice nodes. In general, the velocities of the off-lattice particles were found by an interpolation between the eight nearest lattice nodes using a trilinear interpolation scheme [35]. During the particle propagation, particle-particle collisions were enabled in the algorithm (as described later in the manuscript) to allow for the formation of particle aggregates. It was assumed that the particle aggregates and the nanoparticles did not have inertial effects despite growing in size. It is also important to note that the presence of the particles and particle aggregates did not disturb the computed flow field, *i.e.*, the particles were assumed to be passive. The assumption of passive particles is applicable to suspension systems of nano-sized particles. It holds for particle aggregates obtained in this work, since their size is comparable, or even one order of magnitude smaller than that of flow-tracing particles [36–38], utilized in particle image velocimetry (PIV), a flow visualization technique using passive tracers. Both convection and diffusion contribute to the particle propagation, leading to the following form of the particle equation of motion [35,39]:

$$\vec{X}_{t+\Delta t} = \vec{X}_t + \Delta t \vec{U}_t + \Delta \vec{X} \quad (1)$$

where \vec{U}_t is the particle velocity at time t , Δt is the simulation time step, and $\Delta \vec{X}$ is the travel distance by Brownian motion, whose direction is random and its magnitude follows a normal distribution with a mean value of zero. The standard deviation for the random movement in each one of the three space directions is related to the molecular diffusivity of the particles, D_n , as follows:

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