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Simulation of agglomerate breakage and restructuring in shear flows: Coupled effects of shear gradient, surface energy and initial structure

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ABSTRACT

Nano- and submicron particles typically exist in the form of agglomerates. The shear flow, a common local flow, plays an important role in the breakage and restructuring of agglomerates. In this work, the discrete element method (DEM) with a cohesive contact model is used to simulate the dynamic evolution of agglomerates under shear flow. The results show that the size and structure of agglomerates at a steady state are the results of competition among the shear gradient of the flow field, the surface energy and the initial structure of the agglomerates. Based on the variation of the radius of gyration (R_g) and fractal dimension (D_f) of agglomerates with time, the following three stages can be distinguished: (a) a stage dominated by stretch and breakage, (b) a stage dominated by agglomeration and densification, and (c) a stage characterized by the steady size and structure of agglomerates. With increasing shear gradient, the agglomerate fragments at the steady stage become smaller, more compact and more uniform. When the shear gradient is high, increasing the surface energy can result in larger fragments with a more compact structure. However, when the shear gradient is low, increasing the surface energy does not lead to larger fragments. The effect of initial structure on the stable size and structure of fragments disappears gradually with the increase in shear gradient. The detailed simulation results can improve the understanding and control of the size and structure of agglomerates in practical devices.

1. Introduction

Nano- and submicron particles have many advantages because of their high specific surface area and special optical and electrical characteristics. For example, metal oxide nanoparticles such as ZnO and SnO_2 can be used in batteries and solar energy conversion [1, 2]. Gold nanoparticles can be used to produce biosensors because of their unique properties [3]. However, high surface energy leads to particle agglomeration [4]. In nature, nanoparticles and submicron particles usually exist in the form of agglomerates that are not easily broken. In product and process engineering, the structure of agglomerates undergoes a series of changes, e.g., agglomeration, breakage, and restructuring, which determines their characteristics and influences device performance. As a common local flow in practical devices, the shear flow plays an important role in agglomerate breakage and restructuring. Therefore, determining the relationship between the shear flow condition and the structure and size of agglomerates is meaningful [5, 6].

Some authors have investigated the breakage and restructuring of agglomerates under different flow fields. For example, Oles [7] found that shear forces resulted in the breakup of aggregates, finally leading to a balance between growth and breakup, and that the stable size of the aggregates decreased with increasing shear rate. Gardner et al. [8] studied aggregation kinetics under different laminar shear stresses and found that a larger shear stress resulted in more rapid aggregation because of a higher frequency of particle collision. Selomulya et al. [9] reported that under shear flow, the aggregate size decreased with time; however, the aggregates became more compact. Soos et al. [10] investigated the aggregation and breakage of fully destabilized polystyrene particles under turbulent conditions and found that the mean cluster size at steady state decreased with increasing shear rate. Harshe et al. [11] found that cluster breakup depended on the initial aggregate structure and flow-field conditions. Pieper et al. [12] reported that the size of stabilized agglomerates in an annular-gap high-shear disperser decreased with increasing shear gradient. Zhu et al. [13] studied the morphological properties of kaolin flocs at a steady state under different shear flow conditions and found that the flocs became more compact and regular with increasing shear rate.

In parallel with experimental investigations, computational simulations were performed to investigate agglomerate behavior under shear flows. Sommer et al. [14] compared different methods of modeling the agglomeration and breakage of nanoparticles in stirred media mills. Moreno-Atanasio et al. [15] studied the effect of surface energy on the breakage of agglomerates and found that the damage ratio

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decreased as the surface energy increased for a given impact velocity. Zeidan et al. [16] found that higher shear rates produced smaller and more uniform aggregates, whereas stronger aggregates resulted in a broader size distribution. Hassanpour et al. [17] simulated the breakage of agglomerates with different size ratios under shear flow and found that the agglomerates with a high size ratio were difficult to break but their structure was deformed. Eggersdorfer et al. [18] studied the restructuring, breakup and relaxation of agglomerates in shear flows by the discrete element method (DEM) and found that lower shear stresses resulted in larger fragments and delayed the onset of agglomerate breakup. Lieu et al. [19] found that the stable structure of aggregates was strongly dependent on shear conditions but weakly dependent on the initial structure of the aggregate. Kroupa et al. [20] found that more adhesive primary particles produced larger agglomerates under a given shear rate. The cluster size decreased with increasing shear gradient; however, the Young's modulus had no effect on the cluster size. Harshe et al. [21] found that open clusters produced dense fragments under shear flow but that dense clusters could not produce denser fragments.

The effects of the shear rate and agglomerate properties on the agglomerate size and structure can interplay with each other, which is an idea that has rarely been explored. In the present study, DEM with a cohesive contact model is used to explore the coupled effects of the initial structure and surface energy on the breakage and restructuring of agglomerates under various shear gradients. The cohesive contact model used in this work is a hysteretic-type model [22–25]. The initial agglomerates are generated by diffusion-limited agglomeration (DLA) and reaction-limited agglomeration (RLA) with fractal dimensions of approximately 2.0 and 2.5, respectively [26, 27]. The particles are uniform, with a diameter of 500 nm, and the agglomerate radius is approximately 8–15 μm. The agglomeration-breakage evolution is characterized through analysis of the dynamic change and steady values of size, fractal dimension, and coordination number of agglomerate fragments.

2. Model

2.1. General model

The motion of each particle is calculated on the basis of Eq. (1), which considers the contact force between particles and the drag force [18]. The particle gravity force is neglected because it is very small.

$$m_i \frac{d^2 x_i}{dt^2} = \sum_j \left(\mathbf{F}_{i,j}^n + \mathbf{F}_{i,j}^t \right) + \mathbf{F}_i^{drag} \tag{1}$$

where
m_i is the particle mass, x_i is the particle position vector, $\mathbf{F}_{i,j}^n$ and $\mathbf{F}_{i,j}^t$ are the normal and tangential components of the contact force, respectively, and $\mathbf{F}_i^{drag}$ is the drag force. The contact force will be described in detail in the next section.

The drag force is calculated using Stokes' drag law because the particle Reynolds number is very small. The computational complexity would be increased dramatically if we considered the shielding of the surrounding particles, which is still under active study [28]. In the future, the difference between the drag force of an isolated particle and a particle within agglomerates should be quantified. The expression for the drag force is

$$\mathbf{F}_i^{drag} = 3\pi\eta_f d_p (\mathbf{v}_i - \mathbf{v}_{f,i}) \tag{2}$$

where $\mathbf{v}_{f,i}$ is the local fluid velocity determined by the shear gradient (G) and the Y-axial position of the particle, which is expressed by $\mathbf{v}_{f,i} = G \cdot y_i$. The other two components of the fluid velocity are zero because the shear gradient only exists in the Y direction.

2.2. Contact model

The contact model between particles is based on a cohesive linear spring-dashpot contact model [25]. The normal component of the contact force can be calculated as

$$\mathbf{F}_{i,j}^n = F_n(\delta)\mathbf{n} + \gamma^n \mathbf{v}_{i,j}^n \tag{3}$$

$$\mathbf{n} = (\mathbf{x_i} - \mathbf{x_j}) / |\mathbf{x_i} - \mathbf{x_j}| \tag{4}$$

$$\gamma^n = 2\sqrt{(m_p k_{load}^n)}\ \ln e_n / \sqrt{\pi^2 + \ln^2 e_n} \tag{5}$$

$$\mathbf{v}_{i,j}^n = (v_i - v_j)\cdot\mathbf{n} \tag{6}$$

$$\delta_{i,j} = (\mathbf{x_i} - \mathbf{x_j})\cdot\mathbf{n} - (R_i + R_j) \tag{7}$$

where $\mathbf{n}$ is the unit normal vector pointing from particle i to j, $\mathbf{x}_i$ and $\mathbf{x}_j$ are the coordinates of the mass centers of particles i and j, respectively, γ^n is the normal damping coefficient, and $\mathbf{v}_{i,j}^n$ is the normal relative velocity. The damping force is considered when the particles contact each other. In this model, the effects of other forces (e.g., the lubrication interaction) are included in the "damping force" in a lump. The lubrication interaction is not modeled directly. Comparing different models of lubrication interaction would be an interesting topic for future study, but it is outside the scope of the present work. Parameter $\delta_{i,j}$ is the distance between the surface of particle i and that of particle j. The calculation of $F_n(\delta)$ is distinguished on the basis of the following four contact stages: approach, loading, unloading and detachment.

Stage 1: Approach stage. As particles approach, the van der Waals (vdW) force appears. $F_n(\delta)$ is given by

$$F_n(\delta) = -F_{vdw} \tag{8}$$

The vdW force is calculated using the equation for particles with sizes on the order of submicrons or microns [29] because the particles in this work have a diameter of 500 nm. The surface of submicron or micron particles is not completely smooth; thus, the contact between particles is point contact. The attraction force depends on asperity size d_{asp}, which is usually set as 0.1–0.2 μm in the literature. The formula is as follows:

$$F_{vdw} = F_{vdw,0}\left(\frac{H_0}{H_0 + \delta_{i,j}}\right)^2 \tag{9}$$

$$F_{vdw,0} = \frac{A \cdot d_{\mathrm{asp}}}{24 \cdot {H_0}^2} \tag{10}$$

where A is the Hamaker constant, which is set as 5.0×10^{-21} J, H_0 is the minimum distance that the particle surfaces can approach, which is set as 0.4 nm, and $F_{vdw,0}$ is the contact vdW force at H_0; d_{asp} is the asperity size d_{asp}, which is set as 0.1 μm in this work [30].

Stage 2: Loading stage. When two particles contact each other, the interaction force is the combination of the spring force and the vdW force during the loading stage:

$$F_n(\delta) = -k_{load}^n \delta_{i,j} - F_{vdw,0} \tag{11}$$

where k_{load}^n is the loading stiffness determined by the Young's modulus, $k_{load}^n = 0.5\pi d_{\mathrm{p}} E$.