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Prediction of Viscosity of Slurry Suspended Fine Particles Using Coupled DEM-DNS Simulation

Yuichi Yoshida, Taichi Katsumoto, Shun Taniguchi, Atsuko Shimosaka, Yoshiyuki Shirakawa, Jusuke Hidaka

[a] Department of Chemical Engineering and Materials Science, Doshisha University, 1-3, Tatara-Miyakodani, Kyotanabe, Kyoto 610-0321, Japan
jhidaka@mail.doshisha.ac.jp

Prediction of the apparent viscosity of a slurry suspended fine particles is important for developing slurry-treating processes. After Einstein's formulation for the apparent viscosity of a dilute, completely dispersed slurry, several viscosity equations were proposed. However, viscosity depends not only on the volume fraction of solid particles but also on many factors such as particle shape, particle interaction, the aggregation structure of suspended particles and the structure of fluid flow. A hybrid simulation employing the distinct element method (DEM) and the direct numerical simulation method (DNS) was developed to obtain the relation between these factors and the apparent viscosity of slurry suspended fine particles. The apparent viscosity of a completely dispersed slurry obtained by the simulation is in good agreement with the experimental and calculated values obtained by the empirical equations. Shear-thinning behavior due to the collapse of agglomerates with increase in mean shear rate was observed for the case of a slurry with agglomerated particles. This behavior was caused by the existence of so-called immobile water in the agglomerates. The influences of the shape of aggregation and particle interaction on the apparent viscosity are investigated by the simulation.

1. Introduction

Prediction of the rheological properties of slurry is important for slurry treatment technology. In particular, these properties depend on the microscopic structures of suspended fine particles in the slurry, and the dispersion or aggregation state of the fine particles strongly affects the properties of the final products of industrial processes. Therefore, the detection and control of the dispersion state of fine particles in the slurry are required to derive the functional characteristics of the final products. Currently, the apparent viscosity of the slurry is used to detect the dispersion or the aggregation state of the particles. However, the relation between the apparent viscosity and the dispersion or aggregation state of the suspended particles in the slurry is not understood sufficiently well for it to be used in the control of the dispersion state of fine particles in the slurry.

Since the prediction theory of the viscosity for a well-dispersed dilute suspension proposed by Einstein, many modifications of Einstein's equation for application to a higher concentration range have been proposed considering the interaction between particles; for example, Simha's modification (Simha,1952), Mooney's equation (Mooney,1951), and Thomas's equation (Thomas,1965). In real-world slurry treatment processes, however, most slurries show an agglomerative nature, and suspended particles form agglomerated particles. Usui et al. proposed an estimation method for the apparent viscosity of slurry containing agglomerated particles (Usui et al, 2001). However, the shape and size distribution of agglomerated particles in the slurry are not taken into consideration in the model.

In this study, a hybrid simulation employing the distinct element method (DEM) and the direct numerical simulation method (DNS) was developed to predict the apparent viscosity of slurry suspended fine particles. The influences of particle concentration and degree of zeta potential on the apparent viscosity

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are discussed on the basis of the observation of the microscopic behaviours of the particle-fluid multiphase flow obtained by the simulation.

2. Experiments

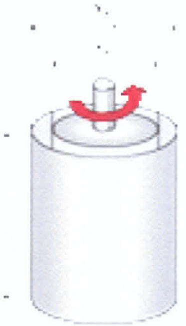


Figure 1: Schematic diagram of coaxial double rotational rheometer

The slurries tested in this study were prepared by using spherical silica particles and glass beads. Spherical silica particles with 2.5, 1.0, 0.45 and 0.1 μm mean particle diameter d_{50} supplied by Nippon Shokubai Co. Ltd. exhibit a sharp mono-modal particle size distribution. Glass beads with 16 and 25 μm mean diameter d_{50} supplied by UNITIKA Co. Ltd. also exhibit a sharp mono-modal particle size distribution. Both a completely dispersed suspension and an agglomerative suspension were prepared. This study used 99% ethylene glycol (NACALAI TESQUE) as the dispersing medium to prepare a completely dispersed suspension (Russel, 1980). Distilled water was used as the dispersing medium for the agglomerative suspension, and PH values of the slurries were adjusted to 1.1 and 6.3 with hydrochloric acid solution and potassium hydroxide solution in order to investigate the effect of the zeta potential of the suspended particles on the apparent viscosity.

The apparent viscosity of the slurries was measured by a commercial coaxial cylindrical rheometer (Rheostress RS75, Haake) as shown in Figure 1. The measurement was taken after a constant shear rate was applied for a sufficiently long time to eliminate the effect of time dependency, and the temperature of the slurry was maintained at 298 K.

3. Hybrid simulation of flow behavior of slurry

A hybrid simulation of the flow behavior of slurry was developed by combining DEM with DNS. DEM developed by Cundall and Strack (Cundall and Strack, 1979) was applied to simulate the movement of particles in slurry, and DNS with a finite difference scheme (Kajishima et al., 1998) was employed to represent complex fluid motion precisely, such as a vortex being shed from moving particles. At the interface between the fluid and the particles, the immersed boundary method (IBM) of body force type proposed by Kajishima (Kajishima et al., 2001) was applied owing to its simplicity.

The distinct element method: The dispersed or agglomerated state of particles in slurry is determined by a superposition of van der Waals attraction forces and the repulsion force due to the electrical double layer around the suspended particle. For simulation of the movement of particles in slurry, these interparticle forces must be considered when adding the mechanical contact forces such as impact and frictional forces between particles. The van der Waals force and the repulsion force given by the DLVO theory are represented as follows:

$$F_{VdW} = -\frac{A_H r_p}{12(L-2r_p)^2 a_{ij}} \tag{1}$$

$$F_{ele} = \frac{64\pi k T_1 r_p \rho_\infty tanh^2\left\{\frac{zc\zeta_0}{4kT_0}\right\} exp\{-K_e(L-2r_p)\}}{K_e a_{ij}} \tag{2}$$

where L is the distance between the particles, ζ_0 is the zeta potential, κ_e is the inverse of the Debye length, z is the ion valence, ρ_∞ is the bulk ion density with z, k is the Boltzmann constant, c is the elementary charge, and A_H is the Hamarker constant. The distance between the particles is limited to 15 nm and Brownian motion is neglected since the small particles move in the high shear flow field.

Direct numerical simulation of fluid dynamics: Since the interaction between fluid and solid phases should alter the microscopic fluid structures, the DNS of multiphase flow, proposed by Kajishima (Kajishima et al., 2001), should be applied to the calculation of the fluid phase. DNS can be used to simulate not only the macroscopic flow dynamics, but also microscopic turbulence structures. The governing equations are the Navier–Stokes equations and the continuity equation for incompressible fluids with constant density and viscosity:

$$\nabla \cdot \boldsymbol{u}_f = 0 \tag{3}$$

$$\frac{\partial u_f}{\partial t} + \boldsymbol{u}_f \cdot \nabla \boldsymbol{u}_f = \frac{1}{\rho_f}\nabla \cdot \tau_f + \boldsymbol{g} \tag{4}$$

$$\tau_f = -pI + \mu_f\left[\nabla u_f + (\nabla u_f)^T\right] \tag{5}$$

where, u_f is the fluid velocity, I is the unit tensor, τ_f is the fluid stress tensor, g is the gravitational acceleration, p is the fluid pressure, ρ_f is the fluid density, and μ_f is the fluid viscosity.

In this study, a second-order central finite difference scheme was applied for spatial derivatives, whereas the second-order Adams-Bashforth method was applied for temporal derivatives of viscous and nonlinear terms. The simplified MAC method was applied for coupling pressure fields with velocity fields. Momentum exchange at the fluid-solid interface was solved by a variety of immersed boundary methods developed by Kajishima. The motion of particles in the fluid flow is generally simulated by solving the equations of linear momentum and angular momentum, which are given by

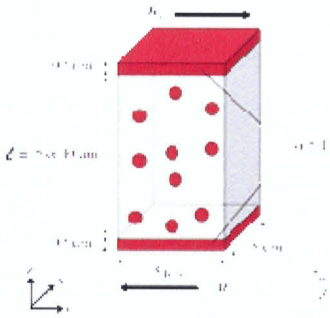


Figure 2: Computational domain in shear field

$$\frac{d(m_p v_p)}{dt} = \int_{S_p} \tau \cdot n dS + G_p \tag{6}$$

$$\frac{d(I_p \omega_p)}{dt} = \int_{S_p} r \times (\tau \cdot n) dS + N_p \tag{7}$$

where v_p and ω_p are the translational and angular velocities respectively, G_p and N_p are the external force and external torque respectively, m_p is the mass of the particle, I_p is the inertia tensor, S_p is the surface area of the particle, n is the outward normal unit vector at the surface, r is the relative position from the center of gravity to a point in the integral region. The computational domain of the simulation is shown in Figure 2. The simulation conditions are listed in Table 1.

Table 1: Simulation conditions

	Dispersion	Aggregation	
System size	5 × 5 × 5	5 × 5 × 10	[μm]
Discrete time	1.0×10^{-10}		[s]
Number of cells for fluid calculation	40×40×40	40×40×80	[-]
Ratio of fluid cell to particle diameter	40×40×40		[-]
Particle	Silica		
Particle density	2500		[kg/m^3]
Elastic coefficient (normal)	2000		[N/m]
(tangential)	1		[N/m]
Restitution coefficient (normal)	1		[-]
(tangential)	0.1		[-]
Friction coefficient	0.1		[-]
Fluid	Ethylene glycol		Water
Density	1113	997	
Viscosity	21×10^{-3}	0.89×10^{-3}	[kg/m^3]
Hamaker constant	1.19×10^{-22}	1.42×10^{-20}	[Pa · s]

4. Results and Discussion

4.1 The apparent viscosity from the hybrid simulation

Figure 3 shows the relation between relative viscosity and shear rate of the slurry. The relative viscosity η_r is the ratio of the apparent viscosity of slurry η_s to that of dispersing medium η. Figure 4 shows the relative viscosity with respect to the relative diameter of the particles defined by d/L, where L is the distance between the inner and outer walls in the coaxial cylindrical rheometer. When $D/L \geq 0.2$, the relative viscosity increases. It tends to deviate from the theoretical value on the order of D/L owing to the existence of the solid boundary. Thus the value of D/L should be less than 0.2 in the measurement of the viscosity of slurry.

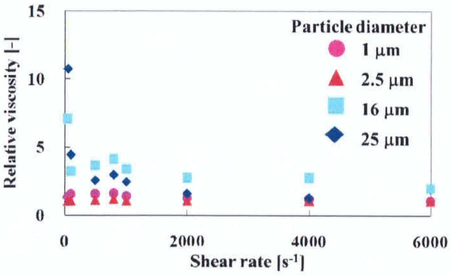


Figure 3: Influence of shear rate on relative viscosity

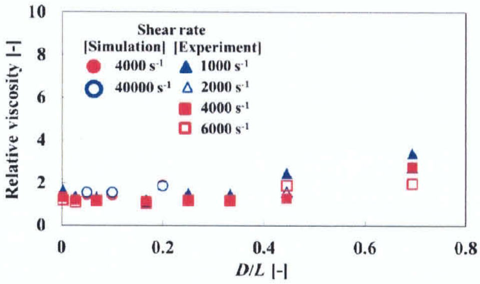


Figure 4: Effect of D/L on relative viscosity

Figure 5 shows the effect of the particle concentration on the relative viscosity of the completely dispersed slurry in which spherical silica particles with 0.54 μm mean diameter are dispersed in ethylene glycol. The simulated values of the relative viscosity agree well with the experimental results. The relative viscosity of slurry increases with the increase in particle concentration ϕ, and is in good agreement with not only Einstein's equation for the apparent viscosity in the case of dilute slurry ($\phi < 0.1$) but also with Mori and Ototake's equation (Mori et al., 1956) for the relative viscosity in the wide range of ϕ specified by

$$\eta_r = 1 + \frac{3\phi}{1 - \dfrac{\phi}{0.52}} \tag{8}$$

This equation is derived under the assumption that the mono-sized spherical particles are oriented in a regular arrangement with the cubic structure in the slurry and are broadening isotropically in the flow of the slurry. The particles in the simulated slurry, however, seem to exhibit a random arrangement in which the particles are irregularly dispersed. To take into consideration the difference in particle arrangement in the particle dispersion, a modified Mori and Ototake's formula whose volume fraction of particles is changed from 0.52 was applied to calculate the relative viscosity and compared with the experimental data. When the volume fraction is 0.57, the estimated viscosity agrees well with the calculated value. Since the relative viscosity obtained by the proposed simulation is in good agreement with both the experimental value and the estimated value from the modified Mori and Ototake's formula, these results show that the proposed hybrid DEM-DNS simulation is very reliable.

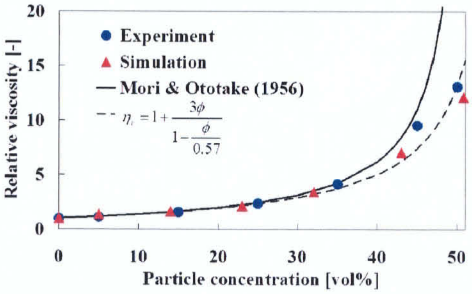


Figure 5: Effect of particle concentration on relative viscosity

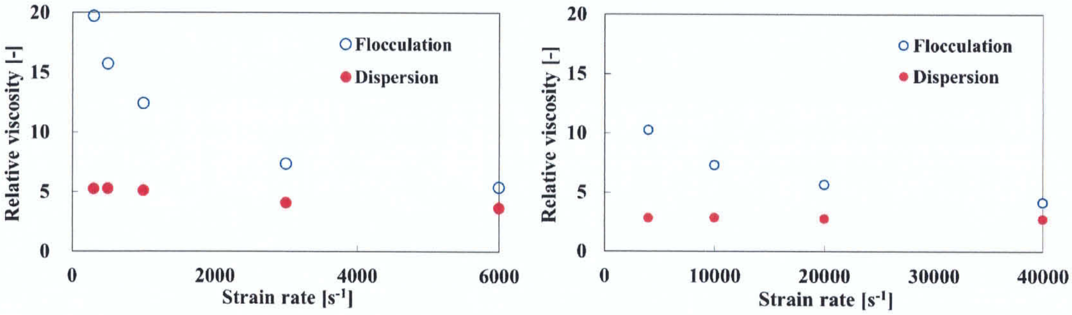
4.2 Influence of dispersed and agglomerated states

To clarify the relation between the dispersion state of particles and slurry viscosity, the relative viscosity of agglomerative slurries is investigated by both experiment and DEM-DNS simulations. In the experiments, 25 vol.% aqueous slurries were prepared and the pH of the slurries was adjusted to either 6.3 for the dispersed slurries or 1.1 for the agglomerative ones. The zeta potentials of the slurries were −58.22 and 0.96 mV, respectively. In the DEM-DNS simulations, the zeta potential of the dispersed slurries was −100 mV, whereas that of agglomerative slurries was −10 mV.

Figure 6 shows the relation between the shear rate and the relative viscosity of the slurries. Figure 6(a) shows the experimental results, and figure 6(b) shows the values obtained by DEM-DNS simulation.

For the dispersed slurries, the relative viscosity is mostly constant similar to a Newtonian fluid, whereas for the agglomerative slurries, the shear thinning of the relative viscosity is observed. These results show that the rheological properties of slurry vary according to the shear rate of slurry.

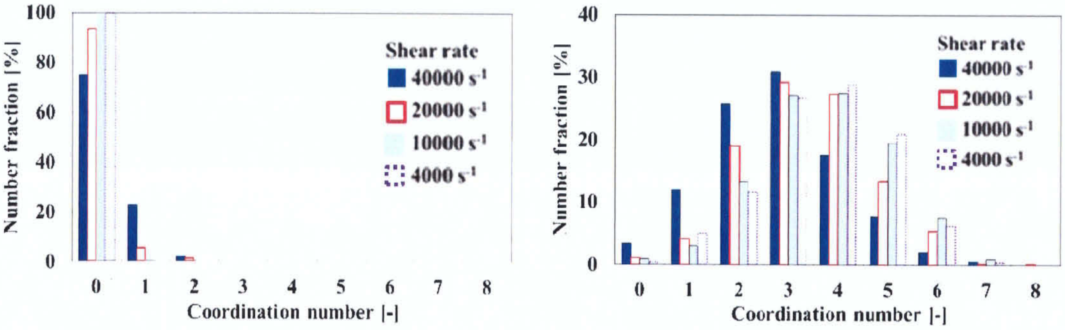
Figure 7 shows the coordination number distribution of suspended particles in shear flow for dispersed and agglomerative slurries, respectively. The coordination number is represented by the number of particles contacting a particle. In the case of the dispersed slurry, the particles in the slurry move with no contacting suspended particles owing to the strong repulsive force between the particles due to the electrostatic interaction. For the agglomerative slurry, the coordination number decreases with an increasing in the shear rate because the agglomerates are broken down owing to the increase in the shear force acting on the agglomerated particles. These results suggest that the breaking up of the agglomerates under the high shear rate causes the shear thinning behavior observed in the case of agglomerated slurries. In other words, the apparent viscosity of the slurry is closely related to the microstructures of particle dispersions or agglomerates.



(a) Experiment

(b) Simulation

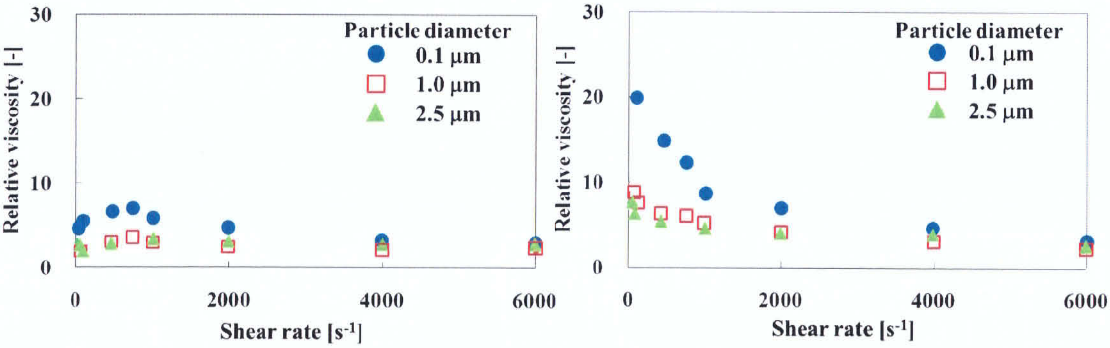
Figure 6: Effect of the dispersion state of suspended particles on relative viscosity



(a) Dispersion

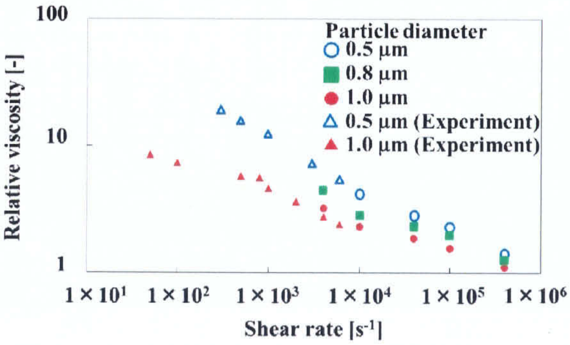
(b) Flocculation

Figure 7: Effect of shear rate on coordition number distribution



(a) Dispersion

(b) Flocculation



(c) Comparison of calculated values with measured values

Figure 8: Influence of particle diameter on relative viscosity

4.3 Influence of particle diameter

Figure 8 shows the influence of particle diameter of suspended particles on the relative viscosity. For well dispersed slurry, the relative velocity is not dependent on the particle size, though the agglomeration of suspended particles is observed in low shear velocity. Figure 8(c) shows the comparison of calculated relative viscosity with the measured ones for agglomerative slurry which the zeta potential is -10 mV and the particle concentration is 25.2 Vol.%. In the region of low shear velocity, the dependence of particle diameter for the relative viscosity is remarkable since the finer particles are easy to form the aggregates.

4.4 Influence of the structure of agglomerated particles on the apparent viscosity

The apparent viscosity of the agglomerative slurry is greater than that of the dispersed slurry, and that of slurry suspended agglomerated particles with a block structure is larger than that of slurry suspended agglomerated particles with a chain structure.

To estimate the effect of immobile water on the apparent viscosity of the agglomerative slurry, we tried to calculate the apparent solid concentration quantitatively. The apparent solid concentration could be calculated by using the threshold value of the velocity gradient. Using the apparent solid concentration, the apparent viscosity of the agglomerative slurry can be estimated by an empirical equation.

5. Conclusions

The DNS-DEM hybrid simulation was developed to predict the apparent viscosity of slurries. The proposed method can be very reliable since the estimated relative viscosity of a completely dispersed slurry obtained by the proposed simulation is in good agreement with the measured and calculated values from the empirical formula. The shear-thinning behaviour resulting from the collapse of agglomerates due to an increase in the mean shear rate is observed in the case of agglomerative slurry. This phenomenon is due to the existence of the so-called immobile water contained in the agglomerates. The apparent solid concentration, which is obtained by regarding the volume of the immobile water as that of solid, is defined. With the apparent solid concentration, the apparent velocity of the agglomerative slurry can be estimated by an empirical equation. The apparent viscosity of the slurry depends on the shape of the aggregates in the slurry. The proposed simulation clarifies the relation between the apparent viscosity of slurry and the characteristics of the suspended particles in the slurry.

References

Cundall P. A., Strack O. D. L., 1979, A discrete numerical model for granular assemblies, Geotechnique, 29, 47–65.

Kajishima T., Ohta T., Okazaki K., Miyake Y.. 1998, High-order finite-difference method for incompressible flows using collocated grid system, JSME,(B), Fluids and thermal engineering, 41, 830–839.

Kajishima T., Takiguchi S., Hamasaki H., Miyake Y., 2001, Turbulence structure of particleladen flow in a vertical plane channel due to vortex shedding, JSME (B), 44, 526–535.

Mooney M., 1951, The viscosity of a concentrated suspension of spherical particles, J. Colloid Sci., 6, 162–170.

Mori Y., Ototake N., 1956, On the viscosity of suspensions, Chemical Engineering, 20,488-493 (in Japanese).

Simha R., 1952, A treatment of the viscosity of concentrated suspensions. J. Appl. Phys., 23, 1020-1024.

Thomas D. G., 1965, Transport characteristics of suspension: Viii. a note on the viscosity of Newtonian suspensions of uniform spherical particles, J. Colloid Sci., 20, 267–277.

Usui H., Kishimoto K., Suzuki H., 2001, Non-newtonian viscosity of dense slurries prepared by spherical particles, Chem. Eng. Sci., 56, 2979–2989.

Russel W. B., 1980, Review of the role of colloidal forces in the rheology of suspensions, J. Rheology, 24, 287-317.