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Impact of Surface Tension and Viscosity on Solids Motion in a Conical High Shear Mixer Granulator

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Surface tension and viscosity are the important properties of liquid binders affecting wet granulation processes. They could be used to control solids flow pattern and relative motion of particles for controlling wetting, granule growth, consolidation, and breakage. This study aims to investigate experimentally the impacts of the two properties with a conical high shear granulator. The results show significant effects of viscosity and surface tension on solids flow pattern and relative motion of particles. The relative importance of the two parameters, the surface tension and the viscosity, are found to vary with the axial and radial positions in the granulator. For example, the viscosity force decreases with an increase in the bed height in the axial direction (vertical plane). The viscosity force between particles coated with PEG4000 solution is in mN order, whereas that between particles coated with ethanol and water is in μN order. © 2009 American Institute of Chemical Engineers *AIChE J,* 55: 3088–3098, 2009
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Introduction

Wet massing granulation is one of the most important unit operations in the chemical, pharmaceutical, detergent, and nuclear industries for enhancing the flowability, appearance, strength, and other physical and mechanical properties of particulate solids.[1–3] It takes place in one of the two types of closed granulating systems: low to medium shear granulators such as fluidized bed granulators and high shear mixers as is used in this study. The main difference between the two types lies in the methods of solids agitation where the former uses an air flow or the vessel wall whereas the latter usually involves the use of impeller to agitate the powder bed. The main mechanisms involved in wet granulation are known to be wetting and nucleation, consolidation and growth, breakage and attrition.[4–6] The properties of the granules such as strength, hardness, friability, morphology, and product size distribution are greatly affected by the uniformity of binder liquid distribution in the bulk powder, the rate of redistribution of the liquid between granules, and the kinetics of granule breakage and size enlargement. These processes are controlled not only by the chemical and physical properties of binder and solids materials,[7] but also by the flow pattern and the relative motion of solids.[8,9] For example, when a binder liquid is poured, sprayed, or dripped onto the surface of a moving powder bed, subsequent spreading

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of the binder occurs through the relative motion of granules and dry powder whereby liquid transfers from coarse wet granules to dry ones and coalescence of dry fine granules with coarse wet granules. The average size and size distribution of the final products are determined by balance between breakage and growth of granules. Such a balance, for a given binder, is a function of particle–particle collisions, which again depends on the particle relative motion and solids flow pattern.[10]

Earlier studies on characterizing flow pattern of an agitated powder bed have been on the effects of solids materials, agitation, and different designs of blades and mixers.[11–24] For example, Bridgwater et al.[25] studied horizontal and vertical displacements of single particles by a single blade pass while Novosad[26] studied particle motion in a vertical cylinder stirred by a long flat blade by dissecting beds of different color layers frozen with paraffin. Ng et al.[3,27] investigated the impacts of particle size, density, fill level, and machine scale on the solids motion in a high shear mixer. Nilpawar et al.[17] measured the average particle velocities on the bed surface and the surface flow pattern in the high shear mixer, and correlated them to the impeller frequency for different binders. Reynolds et al.[23] analyzed the granular temperature in a granulator using a Particle Imaging Velocimetry (PIV). Darelius et al.[28] studied both internal and wall frictional behavior together with velocity data obtained with a high speed CCD camera.

This work is inspired by the fact that the driving force in a high shear mixer is introduced by the agitation blades, and the force is transmitted by particle collisions and liquid bridges. Besides the mixer geometry, the mode of solids agitation and the physical and chemical properties of powder,[8–16,29–33] the impact of liquid binder on the solids motion should also be significant. Liquid binds particles by a combination of capillary pressure, surface tension, and viscous forces. The impacts of these forces on the solids flow patterns and velocity profiles of granular materials in mechanically agitated beds are complex.[7] For example, surface tension and capillary forces always act to pull particles together; their magnitudes depend on the liquid bridge formed between the particles. Viscous forces are dissipative, always acting against interparticle motion. They depend on the solids flow pattern and the relative motion of the particles.

In previous studies, liquid surface tension and viscosity have been seen as creating a force for binding the fine powder to form a granule. These studies focused on the impacts of these forces on the physical properties of granules.[34–40] For example, Simons and coworkers[36,37] identified the impacts of liquid surface tension, viscosity on granule hardness and resistance to deformation, and intended to improve the binder spreading by using wetting agents to reduce the liquid–vapor surface tension. Dreu et al.[38] suggested that the friability, average pore diameter, and porosity of a granule depended on the product of surface tension (γ_L), relative permittivity (ε_R) of the granulation liquid, and cosine of the contact angle (Θ) of granulation liquid on pellets. Tüske et al.[39] studied the effect of surface free energy on the binder spreading and the granule friability. Saleh et al.[5] investigated the size growth mechanisms by using liquids with different viscosities and surface tensions (water, aqueous solutions of polyethylene glycol and an aqueous solution of a polyvinyl alcohol). Thielmann et al.[40] found that particles with a high surface tension resulted in a narrower granule size distribution than hydrophobic ones.

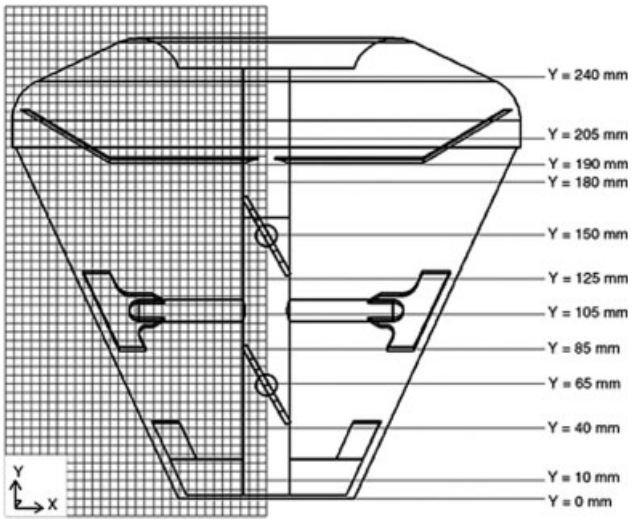

Figure 1. Schematic diagram of the Cyclomix 5 L mixer granulator.

This study is designed to investigate the impacts of viscosity and surface tension on the solids motion in various regions within a high shear mixer granulator, as well as the relative importance of liquid surface tension and viscosity. The aim is to obtain some primary information for understanding the effect of binder properties on the granulation process in a mechanical impeller-type mixer. Positron Emission Particle Tracking (PEPT) technique is used to measure the solids velocity.

Equipment and Materials

A vertical high shear mixer granulator (Model Cyclomix, Hosokawa Micron B.V., Netherlands) was used in this work. The granulator has a nominal capacity of 5 L. As described in the previous communication and shown in Figure 1,[3,27] the mixer has four pairs of flat-bladed impellers, which are 30° to the vertical axis. The first (bottom) and the third pairs of the impellers are opposite to that of the second and fourth (top) pairs in terms of the angle, such that when the shaft is turned clockwise, the first and the third pairs give obtuse motion (upwards agitation) while the second and fourth pairs give acute motion (downwards agitation). The gaps between the impellers and the vessel wall are 5 mm at the side and 2 mm at the bottom. The lid, the impellers, and the shaft are constructed with stainless steel, while the vessel is made of transparent Perspex. All the interior surface in contact with particles is fully polished.

Hollow glass ballotini with an average diameter of 27 μm and a density of 700 kg/m^3 was used in the investigation. Figure 2 shows the particle size distribution. The use of hollow glass beads was based on two considerations: (i) hollow glass beads have a negligible capacity of liquid adsorption, so that liquid volume between particles will not change during the study; (ii) hollow glass beads have a density similar to that of most organic powders.

During the experiments, the granulator vessel was filled to 50% of its volume where the fill covered the shaft of the top pair of impellers. At the beginning of each experiment, the mixer was started at 3 Hz and the binder was poured onto the powder bed surface within seconds. The speed of the mixer was then increased to 12 Hz for 1 min to disperse the

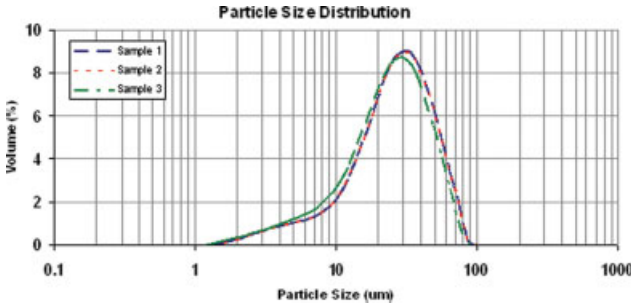

Figure 2. The size distribution of the hollow glass beads.

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binder. After the dispersion phase, the speed was reduced to 6 Hz at which the top impeller tip speed was 4.1 m/s. Particle tracking was then started. Water, ethanol, and 65% v/v aqueous solution of polyethylene glycol 4000 (PEG4000) were used as the binders to investigate the effects of liquid viscosity and liquid surface tension on solids flow pattern and their velocity profiles (Table 1).

In a typical experiment, particles were loaded into the vessel of the mixer granulator which was then started and run for a couple of minutes to ensure that the steady-state was reached before starting the data requisition process. Four hundred gram of 65% v/v aqueous solution of polyethylene glycol 4000 (PEG4000), ethanol, or water were poured into the mixer within seconds, respectively. Particle motion was measured by using the PEPT technique which has been described previously.[41–44] The tracer particles were selected from the bulk powder and radioactively labeled by using techniques reported by Fan et al.[45,46] The data acquisition was performed for 15 min for each run. The time for data collection was determined based on the tracer occupancy, the accumulation of the tracer trajectories, and the kinetics of the process, to ensure that the tracer particle has traversed the full volume of the mixer enough times for sensible averaging. Figure 3 presents the tangential velocity profile at the height 150 mm of experiment using PEG4000 for different data collection time. The results show that the measured velocity profiles are almost the same for the experimental time of 0–7.5 min and 0–15 min. It confirms that 15 min was sufficient for data collection, and the flow field was steady.

Data Analysis

The primary PEPT data will provide the tracer locations against the time in three dimensions (x, y, z) as shown in Figure 4. Typically, a radioactively labeled tracer can be located 100–200 times per second. The data processing involves conversion of the Cartesian coordinates to the cylindrical coordinates, and calculation of the axial velocity (dy/dt), radial velocity (dr/dt), and the angular velocity (ω) by using the

Table 1. Physical Properties of Binders

Binders	Viscosity at 20°C (Pa s)	Surface Tension (N/m)
Water	0.0010	0.0728
Ethanol	0.0012	0.0224
PEG4000 solution	0.343	0.0465

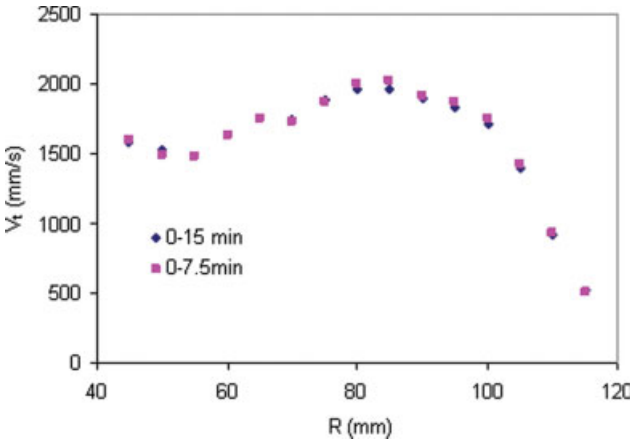

Figure 3. The tangential velocity profile at height 150 mm of the experiment using PEG4000, the measurement time was 0–7.5 min and 0–15 min.

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three-point method. The velocity of a tracer $\mathcal{V}_i = (v_{x_i}, v_{y_i}, v_{z_i})$ is given by three consecutive locations $(x_{i-1}, y_{i-1}, z_{i-1})$, (x_i, y_i, z_i), and $(x_{i+1}, y_{i+1}, z_{i+1})$, for example, the velocity at time t_i in y-direction can be calculated as follows:

$$v_{y_i} = \frac{1}{2}\left(\frac{y_i - y_{i-1}}{t_i - t_{i-1}} + \frac{y_{i+1} - y_i}{t_{i+1} - t_i}\right)$$

In a similar way, the velocity in x- and z-direction can be calculated.

The tracer speed is given by $v = \sqrt{v_{x_i}^2 + v_{y_i}^2 + v_{z_i}^2}$.

The results are further processed by grouping and averaging the data over pixels of 5 × 5 mm in axial (y-axis) and radial (r-axis) directions. This simplified the way of presentation of the data in a 2D axisymmetric system. The results obtained comprise distributions of the axial velocity (dy/dt), radial velocity (dr/dt), and tangential velocity ($r\omega$).

Vertical circulation of tracer particle in the mixer is quantified by a parameter period defined as the time taken by the tracer particle to move from a position higher than an upper boundary in the mixer vessel, reach a position lower than a lower boundary, and return to the upper part of the mixer. The upper boundary is chosen as $Y = 180$ mm, which is about 10 mm above the top impeller. Several lower boundary positions, expressed as the distance from $Y = 180$ mm and denoted as L, are considered to investigate the effect of the boundary positions on the circulation period. The results (Ng et al. 2007, powder tech) shows that selection of the upper boundary at $Y > \sim 180 - 55 = 125$ mm and a lower boundary at $Y < \sim 180 - 140 = 40$ mm should not affect the circulation period much under the conditions of this work.

Theoretical Considerations

Theoretically, the relative motion of the granules/particles can be calculated from Eq. 1.

$$m_i \frac{d\vec{V}_i}{dt_i} = m_i \vec{g} + \vec{F}_s + \vec{F}_a \quad (1)$$

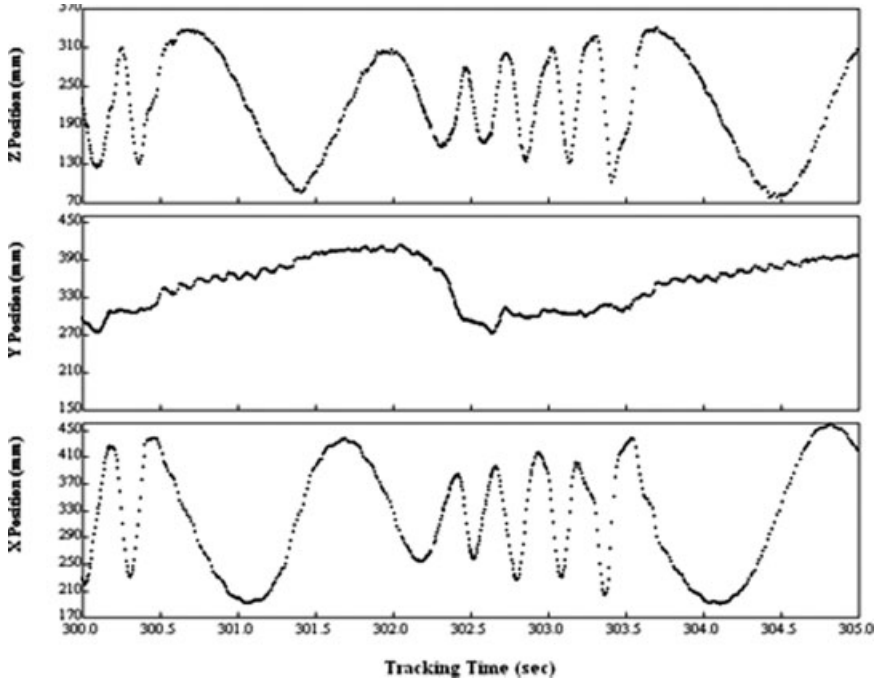

Figure 4. Tracer locations in a 5 s period.

where $\vec{V}_i$ is particle velocity, m_i is the mass of particle/granule number i, $\vec{F}_s$ is a force to separate the granules, $\vec{F}_a$ is a force to hold the partially coalesced granules.

The force $\vec{F}_s$ is determined by the agitation and mixer design. The force $\vec{F}_a$ is known to include at least three components,[4] interparticle frictional force, capillary and surface tension forces, and viscous forces. Van der Waals forces are usually negligible in comparison with liquid bridge forces for granulation processes.[4,47] Electrostatic forces are also negligible in the wet system since the liquid can act as a conductor to neutralize oppositely charged particles, and liquid only takes a small volume fraction in granulation processes. The interparticle friction, capillary and surface tension forces are interrelated, since the liquid bridge acts to pull particles together and this normal force at particle contacts activates interparticle friction. Surface tension and capillary forces always act to pull particles together. Frictional and viscous forces are dissipative, always acting against interparticle motion.

The capillary and surface tension forces have been the subject of many theoretical and experimental studies.[36–39,47–50] Several empirical expressions have been reported in the literature based on individual pendular liquid bridges, such as gorge method and boundary method.[4,34,49] For example, the capillary suction pressure difference ΔP_{cap} across the liquid–vapor interface can be given by the Laplace–Young equation.

$$\Delta P_{cap} = \gamma_{LV}\left(\frac{\ddot{y}}{[1+\dot{y}^2]^{3/2}} - \frac{1}{[y(1+\dot{y}^2)]^{1/2}}\right) \tag{2}$$

where y is the bridge profile as defined by Iveson et al.,[4] $\dot{y}$ and $\ddot{y}$ are the first and second derivative of y.

To solve the above equation, the liquid bridge profile is commonly approximate as a toroid. The surface tension and capillary pressure terms are evaluated at the midpoint of the bridge, called gorge method,

$$F_{gorge} = \pi\Delta P r_2^2 + 2\pi r_2 \gamma \tag{3}$$

or are evaluated at the contact line with one of the spheres, called boundary method,

$$F_{boundary} = \pi\Delta P a^2 \sin^2\phi + 2\pi a \sin(\phi)\sin(\theta+\phi) \tag{4}$$

where a is particle radius (m), θ liquid–solid contact angle, ϕ liquid bridge filling angle.

Willet et al.[4,47] numerically solved the full Laplace–Young equation and obtained the following expression

$$F_{static} = \frac{2\pi a\gamma\cos\theta}{1.0 + 2.1\left(\frac{h^2 a}{V}\right)^{1/2} + 10.0\left(\frac{h^2 a}{V}\right)} \tag{5}$$

where h is half-gap distance between two spheres (m), V the liquid bridge volume (m^3), γ the liquid–vapor interfacial tension (N/m).

However, in practice, the calculation is very complex. Because of the complex interdependence of these different forces, it is often impossible to predict their effects on any particular granule property, unless the relative magnitudes of the forces are known.[4] In this study, we will try to illustrate the impact of liquid surface tension and liquid viscosity on solid motion based on the PEPT data, and discuss the impact based on the theory presented earlier.

The viscous force experienced by a particle moving in a viscous liquid is given by Simons et al.[36] For a dynamic interaction between two spheres, Adams and Perchard[51]

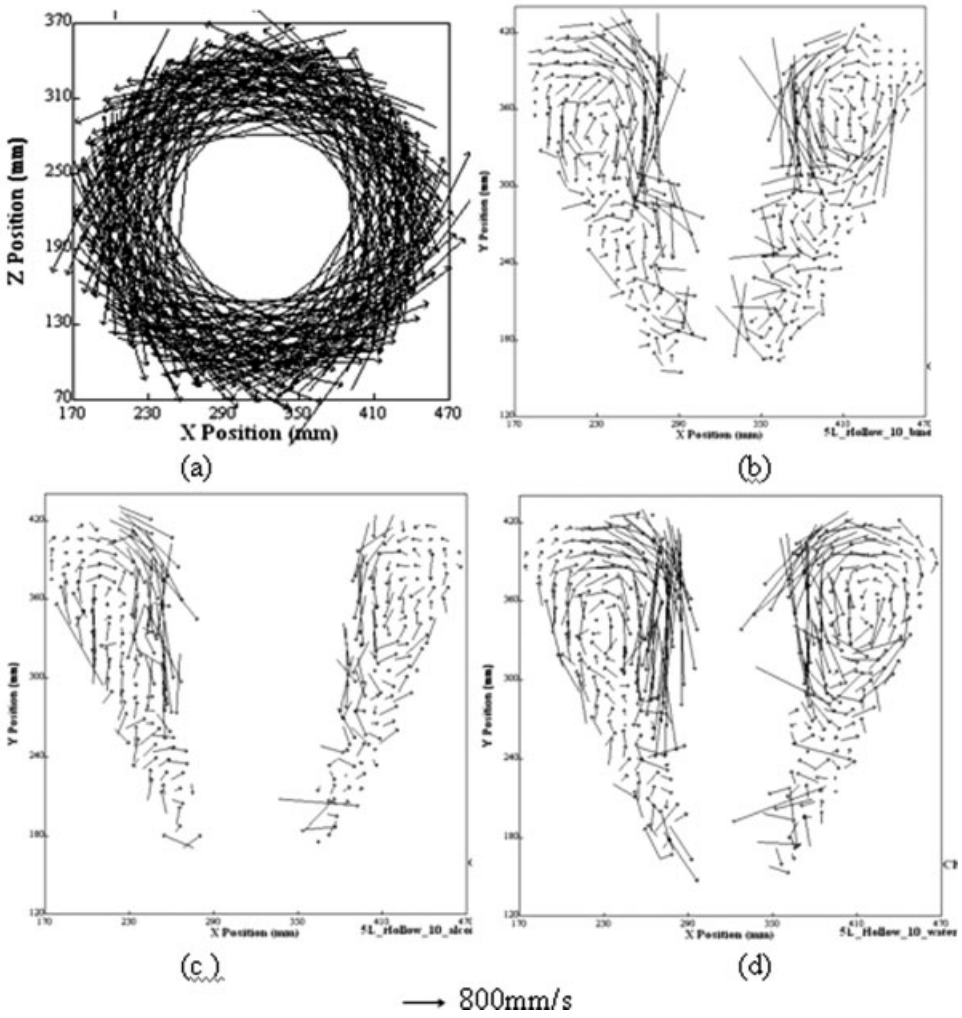

Figure 5. Solids circulation patterns.

(a) Horizontal circulation, (b) vertical motion, PEG4000, (c) vertical motion, ethanol, and (d) vertical motion, water.

applied classical lubrication theory of flow between two rigid surfaces and obtained an expression for the viscous pressure of dynamic pendular liquid bridge. The viscous force of the liquid bridge was given as:

$$F_v = \frac{3}{2}\pi\mu V_{rel}\frac{a^2}{S} \tag{6}$$

where V_{rel} is the relative velocity of the spheres, a the radius of particle, and S the separation between particles.

In fluid dynamics, the capillary number is used to qualify the relative effect of viscous forces vs. surface tension acting across an interface between a liquid and a gas, or between two immiscible liquids. It is defined as:

$$Ca = \frac{\mu v}{\gamma} \tag{7}$$

where μ is the viscosity of the liquid, γ is the surface or interfacial tension between the two fluid phases, v is a characteristic velocity, which is represented by the particle velocity in this study.

Results and Discussion

Effect of added liquids on solids flow patterns in the high shear mixer

In this section, 65% v/v aqueous solution of PEG4000, water, and ethanol are used as binder liquids to investigate the impacts of the surface tension and the liquid viscosity on the solids flow pattern, solids circulation time and solids velocity distribution in the mixer. The results are presented in the Figures 5–8.

As shown in Figure 5, particles circulate in both horizontal and vertical directions of the mixer. The circulation pattern in the horizontal plane is mainly in accordance with the motion of the impellers. In the vertical plane, the particles are pushed upwards at the periphery and fall in the inner annulus closer to the centre. Two vertical circulation cells can be seen, with one located at the top section of the bed and the other at the intermediate section.

Figure 6 shows the solids circulation frequencies in the vertical and horizontal planes. The circulation frequency in the vertical plane is quantified by tracking the tracer particle

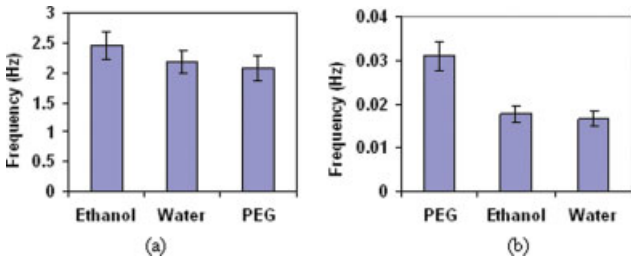

Figure 6. Solids circulation frequency.

(a) Horizontal circulation and (b) vertical circulation. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

movement from a position higher than an upper boundary in the mixer vessel, reach a position lower than a lower boundary, and return to the upper part of the mixer. Our previous work indicates that selection of the upper boundary at bed height $h > 125$ mm and the lower boundary at $h < 40$ mm should not affect the circulation period much under the conditions of this work.[27] The interesting phenomenon is that the circulation frequency, either in vertical or horizontal planes is dependent not only on the impeller motion, but also on the viscosity and surface tension of the added liquids. For the horizontal circulation, the addition of ethanol gives the highest frequency, while the use of PEG4000 gives the lowest frequency. This could be explained by the strength of the liquid bridges. In a granulator, the velocity gradient tends to break the liquid bridge, and part of the solids kinetic energy is consumed to overcome the resistance generated by the liquid bridge. As shown in Eqs. 1–6, the capillary force is dependent on the surface tension and the volume of the liquid bridge. The viscous force is dependent on the relative motion of solids, liquid viscosity, and solid size (Eq. 6). For a given solids and a liquid volume, the strength of the liquid bridge is proportional to the liquid surface tension, the liquid viscosity, and the relative speed of solids. In the horizontal plane, the relative speed is much higher, which can be seen from the data shown in Figures 9 and 10. The viscous force, therefore, dominates the solids motion. A higher viscosity gives a larger resistance, therefore, particles coated with PEG4000 circulate slower in the horizontal plane. For liquids with the same viscosity, a

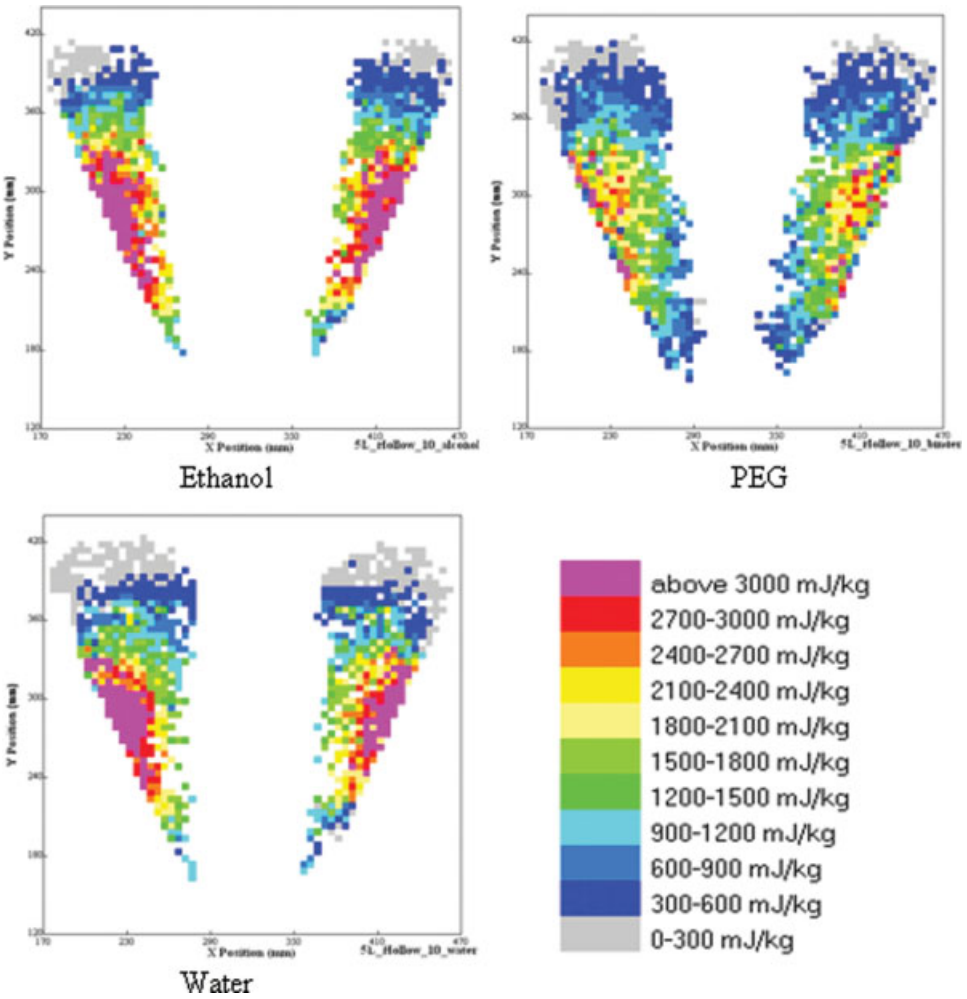

Figure 7. Kinetic energy of particles coated with different liquids.

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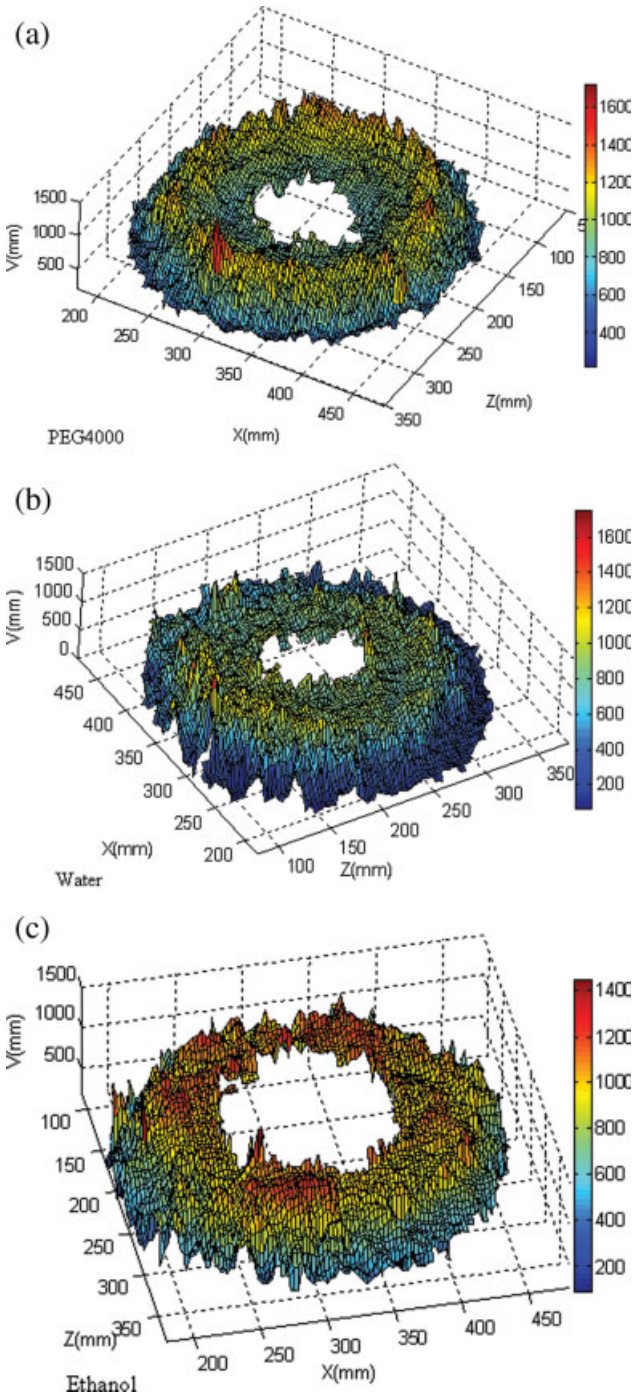

Figure 8. Spatial speed distribution of solids coated with different liquids at a bed level of 200 mm.

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higher surface tension gives a larger resistance. The particle coated with ethanol ($\gamma = 0.0224$ N/m), therefore, circulates faster than the particle coated with water ($\gamma = 0.0728$ N/m).

In the vertical plane, the impacts of liquid viscosity and surface tension on solids circulation are different. Solids particles coated with PEG4000 solution have a higher circulation rate than that coated with water and ethanol, which implies that particles coated with the former binder travel faster than those coated with latter binders in the vertical circulation. For ethanol and water, even though their surface tensions are very different, their vertical circulation rate is very similar. This may indicate that the vertical motion does not make significant contribution to the breakage of the liquid bridge, or granules. The solids are pushed upwards and fall downwards mainly in blocks, i.e., the relative motion between particles in the vertical direction is low in comparison with that in the horizontal direction.

Figure 7 shows the distribution of solids kinetic energy in the vertical plane, whereas Figure 8 illustrates the spatial solids velocity distribution in horizontal plane across a thin layer of 10 mm located at a bed height of 200 mm. One can see that they vary with the added liquids and the bed height level.

Effect of liquid surface tension on solids velocity

The impact of liquid surface tension on solids motion in a granulation process are often analyzed using theoretical models established for individual pendular liquid bridges between pairs of particles.[4,36] As shown in Eqs. 2–5, to calculate the capillary and surface tension force, the shape of pendular liquid bridges between the constituent particles is an important factor. However, in a granulation process, a particle is surrounded by many others. The volume and the shape of a liquid bridge vary with the particle population, the relative motion of the particles, the nature of the particle surfaces, and the wetting behavior of the liquids.[4] There are many uncertainties for the calculation. For example, the particle population in a unit volume varies with solid flow pattern. The number of particles per unit volume is higher in regions adjacent to the mixer wall and the heap just ahead of the blades, but lower in the bed surface and the wake behind the blades. Here, the experiments were designed to directly observe the impacts of the liquid surface tension on the solids motion. Water or ethanol was used because their viscosities are almost the same (0.0010 Pa s and 0.0012 Pa s), but their surface tensions are significantly different. When the same hollow glass beads are used and the agitation speed is fixed, the difference observed in the solids motion should be mainly determined by their surface tension. The van der Waals and the electrostatic forces can be important in some cases. However, in liquid-bound systems, electrostatic forces are negligible since the liquid could act as a conductor if they are conductive to neutralize the charges and also there is a lack of bulk liquid phase to allow for the development of the double layer on particle surfaces. Van der Waals forces are also negligible compared to liquid bridge forces for systems with particle size greater than 10 μm[4].

Figures 9 and 10 present the axial (V_y) and tangential (V_t) particle velocities in different regions of the bed when water or ethanol is used. The results show that the impact of surface tension force on solids motion varies with the bed position and hence the solids flow pattern. At the bed height of 70 mm, the tangential velocity of particles coated with water ($\gamma = 0.0728$ N/m) increases almost linearly at a rate of 20 (mm/s per mm) from the centre to the wall, while the

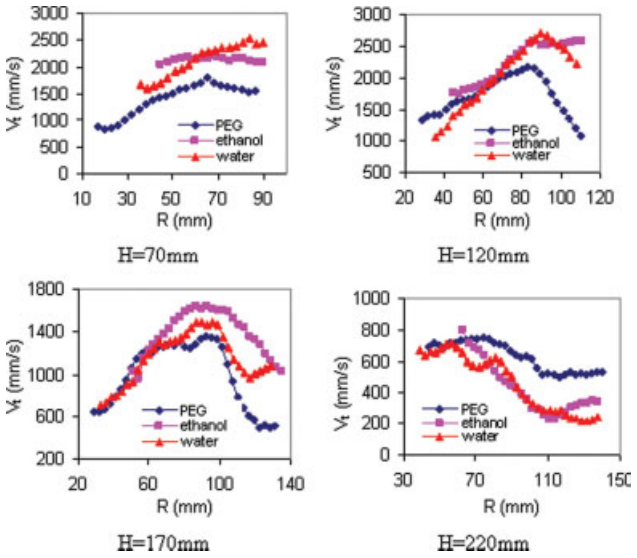

Figure 9. Variation of tangential velocity of solids coated with different liquid in the bed.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

tangential velocity of particles coated with ethanol (γ = 0.0224 N/m) increases only marginally at the rate of 0.37 (mm/s per mm distance). In the inner annulus (R = 30–60 mm), solids coated with ethanol (γ = 0.0224 N/m) move at a velocity higher than the solids with water (γ = 0.0728 N/m); whereas in the region adjacent to the mixer wall (R = 70–90 mm), solids coated with water (γ = 0.0728 N/m) move fast. This means that, at the bottom section of the mixer, the impact of surface tension force on the solids motion is small in the centre part, but is large in the wall region.

As the bed height increases from 110 to 170 mm (Figures 7b, c), the highest value of the tangential velocity is a fifth to a third of the tip velocity and highest velocity is located in the region of R = 80–100 mm. The peak position corresponds to the points where particles change their motion from upwards to downwards as shown in Figures 5 and 7. In comparison with the bottom section, the impact of surface tension on the solids motion is different, particularly, in the region where particles travel upwards, the tangential velocity of solids coated with water is smaller than the solids with ethanol. The drop of the tangential velocity in this region is mainly due to the particle-wall frictional interaction. The water (γ = 0.0728 N/m) has a high adhesion to the steel wall; therefore, a higher wall friction is applied to solids with water than the solids with ethanol (γ = 0.0224 N/m). In the top region of the bed (Figure 7d), the tangential velocity of solids decreases significantly with the increase in the radial position. The wall effect is similar to Figures 7b, c.

In the vertical plane, particles travel upwards at the periphery and downwards in the central region (Figure 5). The magnitude of upward velocity is much smaller than that in the downward motion (Figure 10). The impact of surface tension on the vertical motion varies with solids position within the mixer. In the bottom and top sections (in Figure 10, H = 70 and 220 mm), solids coated with water (γ = 0.0728 N/m) are pushed upwards at a velocity higher than the solids coated with ethanol (γ = 0.0224 N/m). This suggests that the liquid bridge with a high surface tension be more rigid and could transfer the impeller force further away, which leads to particles coated with water to move faster.

In the intermediate section, solids coated with different liquids have a very similar upward velocity; the surface tension seems not to affect the solids upward motion. It is dominated by the impeller rotation.

In the material bed, the relative motion between particles tent to break the liquid bridge. The rupture of the liquid bridge between two particles results in a formation of two fresh interfaces. For the same sized solids, the freshly generated interface area (A) would be the same on the two particles. As the fresh interfaces are created, the surface energy of the particle increases, and could be estimated by

$$\Delta G = 4\pi a h \gamma_{LV} = 6.28 \times 10^{-5} h \gamma_{LV} \tag{8}$$

where h is the cap height of the fresh spherical area which is generated due to particle separation (m), a is the radius of the particle (m).

To break a liquid bridge, the energy (velocity gradient) required to overcome the surface tension force is $4.57 \times 10^{-6}h$ (J) for water, and $1.41 \times 10^{-6}h$ (J) for ethanol. As a consequence, the rupture of water-bridge will consume more solids kinetic energy in comparison to that for rupturing ethanol-bridge. After rupturing the liquid bridge, the change of solids velocity should be greater in water-coated systems than that in the ethanol-coated systems. This explains why particles coated with water have a low velocity in the region where particles travel downwards. When particles avalanche down, they are less compacted, and the liquid bridge could be easily ruptured.

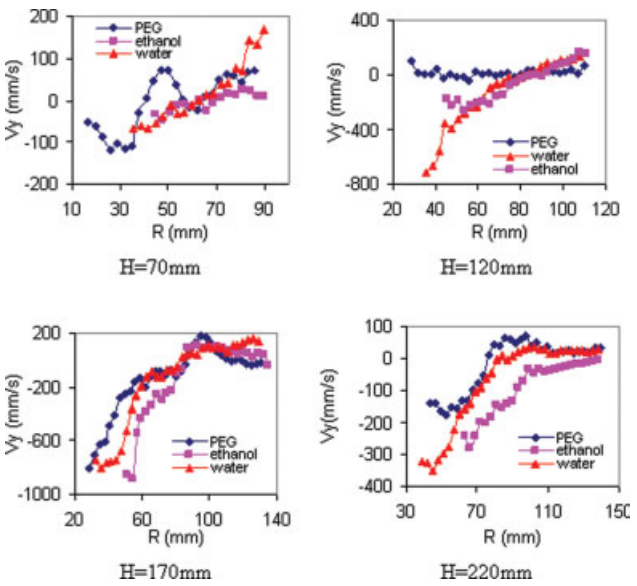

Figure 10. Variation of solids velocity in vertical plane.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

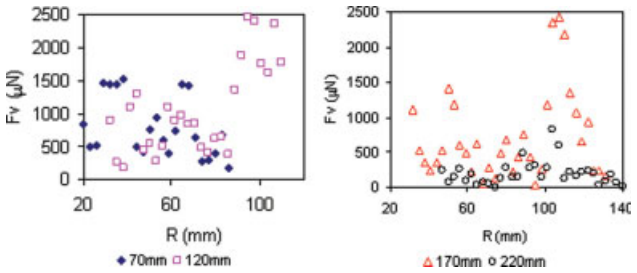

Figure 11. Viscous force between particles at different bed height when PEG4000 is added.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Effect of liquid viscosity on solids motion

Figure 9 presents the tangential solids velocity using different liquids, water, ethanol, and PEG4000. The viscosities of the three liquids are, respectively, 0.001, 0.01, and 0.343. The results indicate that the impact of liquid viscosity on the tangential velocity varies with position in the mixer. In the bottom section of the bed (Figure 9a), the tangential velocity of solids coated with a viscous liquid (PEG4000) is much smaller than the solids coated with less viscous liquids. In the intermediate section of the mixer, both viscous force and surface energy play an important role, particularly, in the upward regime. Solids coated with PEG4000 solution have the lowest velocity, while solids coated with ethanol have the highest velocity. For the three liquids, the viscous forces are in the order: PEG4000 > water and ethanol; the surface energy forces are in the order: water > PEG4000 > ethanol. The strength of the liquid bridge is, therefore, in the order: PEG4000 > water > ethanol. The significant decrease of the tangential velocity in the region adjacent to the wall is due to the effect of wall friction.[19] The solids coated with PEG4000 have the strongest liquid bridge with the mixer wall and between the particles. The rupture of the PEG4000 liquid bridge consumes the largest particle kinetic energy, therefore, creating a large velocity gradient. The solids coated with ethanol have the weakest liquid bridge. The breakage of the ethanol bridge consumes the smallest kinetic energy. The particles, therefore, move fast.

A significant change in the velocity pattern is observed in the upper part of the bed (Figure 9d). The magnitude of solids velocity in the region adjacent to the wall is not in the same order as the strength of the liquid bridge. Solids coated with high viscosity liquid move faster, rather than slower as seen in the intermediate section. The liquid viscosity seems to play a more important role than the surface tension.

In the vertical plane (Figure 10), solids coated with different liquids travel upwards at a relative uniform velocity in the region adjacent to the wall when the bed height is greater than $H = 120$ mm. However, in the region where solids travel downwards, the velocities vary significantly with the bed height and the added liquids. In the lower part of the bed ($H < 120$ mm), solids coated with water or ethanol have a very similar velocity profile. This may indicate that the downward velocity profile is dominated by liquid viscosity, rather than their surface tension. When the bed height is greater than 170 mm, the downward velocity decreases with the bridge strength. Particles coated with PEG4000 have the lowest velocity, while particles coated with ethanol move fast. Both the viscosity and the surface tension are the dominate factors affecting the solids motion.

Figures 11 and 12 demonstrate the distribution of viscous force in the radial direction at different bed heights. It is calculated based on the Eq. 6 and the relative velocity V_{rel} of particles measured by using the PEPT technique. In general, the viscous force of PEG4000 liquid is in mN order, while the viscous force of ethanol and water is in μN order. The pattern of the force distribution varies with bed height and the radial position (R). In vertical plane, it decreases with the increase in the bed height (H). In the lower section of the material bed, viscous force dominates over capillary forces, while in the upper section of the bed, both capillary forces and viscous force are the dominate factors affecting the solids motion. In horizontal plane, the variation is much more complex. For example, the force of PEG4000 liquid is large in the region where particles travel upwards. While for water and ethanol, the radial distribution of the viscous force significantly depends on the bed height (H).

Figure 13 presents the capillary numbers for the experiments where PEG4000, ethanol, and water were used as a binder, respectively. The capillary number represents the relative effect of viscous forces vs. surface tension acting across an interface between a liquid and a gas, or between two immiscible liquids. The calculation is based on Eq. 7 by using particle velocity, liquid viscosity, and liquid surface tension. The results show that, in general, the capillary number (Ca) for the PEG4000 system is much higher than that when water and ethanol were used. The capillary number for the PEG4000 system is in the order of 10^{-3}, while it is in

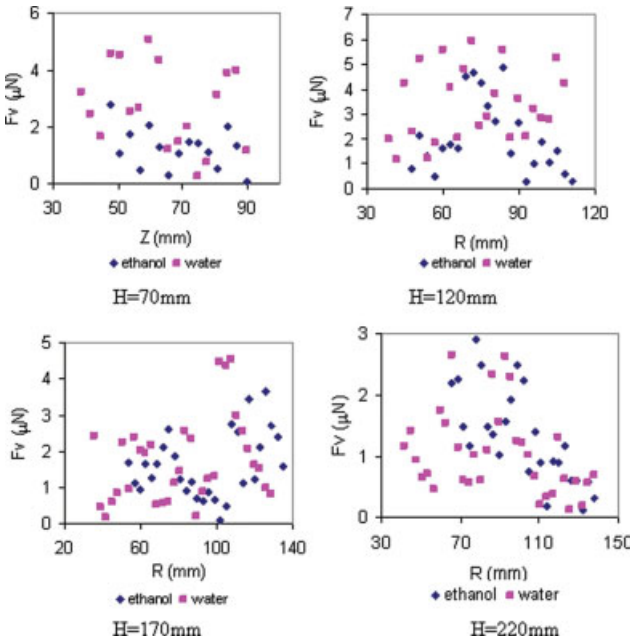

Figure 12. Distribution of viscous force between particles in radial direction when water and ethanol are used.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

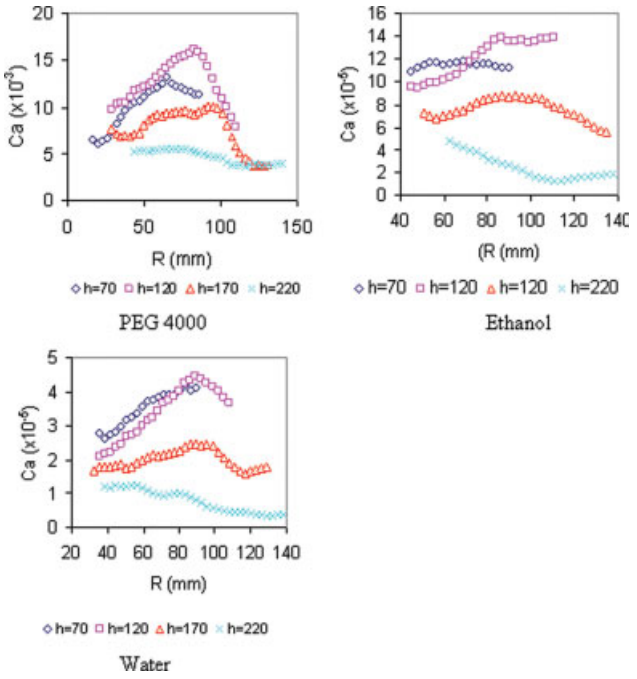

Figure 13. Distribution of capillary number in the mixer when PEG4000, water, and ethanol are used, respectively.

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

the order of 10^{-5} when water and ethanol were used. The capillary number also varies significantly with the geometrical position within the bed. In the axial direction, it decreased greatly with increasing in the bed height. In the radial direction, it is lower in the bed centre and the region near to the bed wall.

Conclusions

Solids motion in a high shear mixer granulator is shown to vary significantly with the addition of liquid binders. In general, solids circulation occurs in both horizontal and vertical planes. The circulation frequency in the horizontal plane is about 100 times higher than that in the vertical plane. The horizontal circulation frequency decreases with increasing the strength of liquid bridge because the rupture of the liquid bridge consumes more particle kinetic energy. The vertical circulation frequency increases with increasing liquid viscosity. This may indicate that the relative motion of the particles in the vertical direction is low as confirmed from the solids velocity profiles.

The impact of liquid surface tension and the liquid viscosity to the solids motion vary with their axial and radial position. In the bottom section of the mixer, the effect of liquid viscosity dominates over that of surface tension. Particles coated with a higher viscosity liquid move slower. For the same viscosity, particles coated with a higher surface tension liquid move faster and generate a larger velocity gradient in the region adjacent to the wall. In the intermediate section of the bed, solids speed depends on the strength of the liquid bridge and is determined by both the effects of liquid surface tension and viscosity. The velocity decreases with an increase in the bridge strength, particularly, in the region adjacent to the mixer wall. The strong liquid bridge corresponds to a high wall friction. In the top section of the bed, the velocity profile is significantly different from that in other sections. The effect of liquid viscosity dominates over that of surface tension. Solids coated with a higher viscosity liquid move faster.

The viscous force between particles is dependent on the liquid viscosity and the relative motion of the particles. In general, the viscous force between particles coated with PEG4000 solution is in mN order, while that for particles coated with ethanol and water is in μN order. In the vertical plane of the mixer, the force decreases with an increase in the bed height. In the lower section of the material bed, viscous force dominate over capillary forces, while in the upper section of the bed, viscous force is much smaller, both capillary forces and viscous force dominate the solids motion at a certain impeller speed. In the horizontal plane, the force with PEG4000 binder is large in the region where particles travel upwards. While for water and ethanol, the radial distribution of the viscous force significantly depends on the bed height.

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