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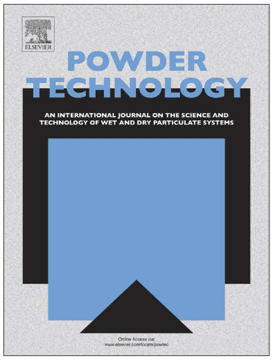

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Experimental Evaluation of the Effect of Particle Properties on the Segregation of Ternary Powder Mixtures

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Abstract

Segregation of components, especially the low content level of a highly active substance, has serious deleterious effects on powder formulation. This study investigates the effect of particle properties, particularly size, shape, density and cohesion, on the segregation of laundry detergent powders. Tetraacetylethylenediamine (TAED) particles, spray-dried synthetic detergent powder referred to as Blown Powder (BP) and Enzyme Placebo Granules (EP granules) are used as a model formulation. The segregation of components is evaluated using image processing of the photographic records taken from the front face of a two-dimensional heap of powders. Coefficient of variation concept is considered as segregation index. It is shown that EP granules, the component used as low-level ingredient in the ternary mixture, are prone to extensive segregation due to their higher density as compared to BP and TAED particles. Desired properties for segregation minimisation of EP granules have been further investigated. It is found that the segregation of EP granules can be reduced noticeably by applying a thin layer of a sticky liquid on them before mixing with the rest of powders with full particle size distribution of the components. Polyethylene glycol has been used for this purpose. Addition of 2.5 wt % gives an optimum level to reduce the segregation of EP granules without compromising the flowability of the mixture as well as EP granules themselves.

Keywords: Segregation Minimisation, Segregation Mechanisms, Laundry Detergent Powders, Low Content Level Ingredient.

1-Introduction

In many industrial sectors involved in manufacturing and handling of granular materials, such as pharmaceutical, food, fertiliser, mining and agriculture, segregation phenomenon is encountered particularly during handling, transportation and storage of the product. For example, variation of active pharmaceutical ingredient content (API) from tablet to tablet in pharmaceutical industry or taste variation in a drink powder mixture could have adverse impact on the product quality as well as costs of the production. Therefore, most powder formulations aim to achieve the most possible homogeneous products [1, 2].

Material properties is an important factor affecting the segregation of granular materials [3, 4, 5]. Among them, the size and density of particles have been widely investigated [6-12]. Liao et al. [13] studied the granule segregation in a rotating drum. They found that increasing the particle density led to an increase in mixing time as dense particles gathered more into the centre of drum. Liu et al. [14] investigated the segregation of granular samples of microcrystalline cellulose and starch during blending in a cylindrical container. They concluded that larger starch granules had more tendency to move to the top of the mixture. On the other hand, the smaller microcrystalline cellulose granules moved to the bottom of the mixture. In another research carried out by Cho et al. [15], segregation of coloured glass beads was investigated in a double cone blender. They reported a better mixing of fines as compared to free-flowing large particles because of their cohesive nature arising from van der Waals forces.

Effects of shape and/or surface roughness of particles were also explored by other researchers [16, 17]. Segregation of binary mixtures containing salt and other food seasoning powders was examined by Shenoy et al. [18] in a paddle mixer. They concluded that particle size, shape and density could influence the segregation of binary powder mixtures. They showed that differences in bulk density and shape had a greater effect on particle segregation than differences in particle size. Remy et al. [19] examined the effect of particle roughness of cohesionless glass beads on their kinematics in a bladed mixer by both experimental and numerical simulation methods, using particle image velocimetry and Discrete Element Method (DEM). Particles of various roughness with different friction coefficients were examined. They found that the amplitude of the velocity fluctuation of components increased as particle surface roughness was increased which led to the formation of less uniform flows inside of the mixer.

Important segregation mechanisms are trajectory, percolation, fluidisation, agglomeration and push-away effect [4, 20]. In inclined chute flow, larger particles could travel further than smaller particles by the trajectory segregation. Based on the percolation mechanism, fines could find their way through the voids of coarse particles during handling, provided the void space between particles is large enough to admit the fine particles to pass. In fact in this mechanism, large particles could act as a screen letting the smaller particles to penetrate. When discharging from a height, dense particles could sink to the bottom while fine particles become airborne by fluidisation segregation. Agglomeration segregation mainly occurs when particles are cohesive (e.g. in the presence of moisture). Based on this mechanism, fine particles could form large clusters and move towards the periphery of the bulk of mixture. Dense particles behave as if they are small and could build up more in the centre of powder mixtures by a mechanism known as push-away effect.

By manipulating particle properties such as changing the particle size distribution and/or optimising the bulk cohesion, it is possible to reduce segregation [4, 21, 22]. Jain et al. [23] showed that the segregation of both binary and ternary mixtures of glass beads with different sizes could be reduced in vertically vibrated cylinders by slightly increasing the size distribution of each species or by increasing the mass fraction of the intermediate size specie in the ternary mixture. In another work done by Chou and Hsiau [24], the effect of wet granular material on the segregation reduction of particles in a rotating drum was investigated. They reported that at a higher liquid content, the segregation index could be reduced, presumably due to the formation of liquid bridge clumping the small particles forming larger particles. The formation of bigger particles could decrease the size ratio in the mixture and therefore could reduce the extent of particles segregation. Optimising the geometrical design of equipment could be another approach for segregation reduction [25, 26]. The effect of geometry design of a mixing process on particle segregation was investigated by Windows-Yule and Parker [27]. In this study, they found that density-driven segregation could be controlled by manipulating the aspect ratio of the equipment without the need of changing particle material properties. In a work done by Vanarase and Muzzio [28], an optimum impeller configuration of a continuous mixer was reported to be when blades could push the powders backward (back mixing) to keep a relatively mixed state of the powders. Nevertheless, optimising the equipment design for the segregation reduction of particles is less favourable in many industries due to the high level of capital investment.

Segregation of minor components has a significant impact on industries dealing with powders. For example, the segregation of low content level enzyme granules in detergent industry should be closely monitored as it adversely impacts the quality of the final product. Despite considerable reported research on particle segregation, there is a lack of in-depth work on the evaluation of segregation reduction of low content level ingredients (less than 2 wt %), particularly in multicomponent powder mixtures. In this paper, the main segregation mechanisms in a multicomponent mixture of laundry detergent powders are first evaluated by investigating the effect of particle properties of components on the segregation of powders. Furthermore, segregation mechanism of low content level enzyme granules is evaluated and the effect of particle surface stickiness on their segregation reduction is studied, where a thin layer of liquid coating is applied to the surfaces of particles. The optimum coating level of the minor ingredient (without compromising the flowability of the powders) to reduce their segregation is then reported.

2-Materials and Experimental Procedure

As model material systems, binary and ternary washing powder mixtures, comprising blue Tetra Acetyl Ethylene Diamine (TAED, used as bleach agent in detergent formulation), white spray-dried synthetic detergent powder referred to as Blown Powder (BP, used as active cleaning agent in detergent formulation), and red Enzyme Placebo Granules (EP granules) are used. The results of particle size distribution of all powders, obtained using British Standard sieves, are provided in Fig. S1, supplementary document, as well as in previous study [29]. To avoid enzyme exposure risk, enzyme placebo granules (EP granules) representing the actual enzyme granules were used. BP and TAED were obtained from Procter and Gamble (P&G), Newcastle Innovation Centre. EP granules were provided by DuPont, USA.

To investigate the effect of particle properties on the segregation mechanisms, components have been characterised separately. Malvern Morphologi G3, Scanning Electron Microscopy (SEM, Hitachi TM3030 Bench Top SEM system) and Nanotom X-ray computed tomography were used for particle shape analysis. Using Malvern Morphologi G3, the average circularity of the projected area of a number of particles was analysed. Also, 3D visualization of particles was obtained by X-ray microtomography (XRT) using the Nanotom X-ray computed tomography instrument (GE Phoenix, Wunstorf, Germany) to measure the sphericity of particles. For the evaluation of the effect of particle stickiness on the segregation of EP granules, Polyethylene glycol (PEG, Pluriol E 600) was used as a coating agent. PEG, a family of long chain polymers, is used in many applications, such as surfactants, ointments, foods, pharmaceutics and cosmetics. In this study, PEG is used as a coating material due to its compatibility and applicability in detergent formulation. PEG is used for many applications in detergent industry, including as a lubricant and surfactant. In some detergent formulations, PEG is used as anti-redeposition agents. It should be noted that the use of PEG with higher molecular weight should be applied with caution as it may not be fully soluble in water [30, 31]. Therefore, a sticky PEG with lower molecular weight, which is fully soluble in water, is used in this study.

A domestic Kenwood mixer was used for coating process and the liquid was added dropwise into the powder bed by means of a syringe. Cross sectional liquid coverage of coated EP granules has then been assessed using confocal laser scanning microscopy (model: Zeiss LSM880). For the flowability analysis, Schulze shear cell and Freeman FT4 Powder Rheometer have been used. The angle of repose of the generated powder piles was measured using image processing tool as shown in Fig. 1.

Fig. 1. Angle of repose measurement using image processing tool.

Segregation mechanisms have been investigated in a two-dimensional (2D) heap test as shown in Fig. 2-a. The mixture of powders is poured through a funnel fixed to the top side of the heap box to form a heap. The funnel has an inclusive half angle of ~40° and the downcomer pipe of 30 mm long with 7 mm ID. When fixed to the top of the rig, the tip of the downcomer is at 150 mm distance from the base of the container. In detergent industry, a tumbler is mainly used to mix the raw materials. Therefore to mimic the real conditions, the mixture of powder was premixed using tumbling method and then was discharged through the funnel. The aim of study is mainly focused on the investigation of the segregation tendency during process of heap formation and vibrated heap which represent conditions encountered during box filling and transportation, respectively.

The Segregation Index (SI) of components in the formed heaps is then evaluated using image processing of the photographic records taken from the front face of the heap. A Nikon camera (D3300, 24 MP) was used for taking the photos of the heap of powders. For measuring SI, the image is first divided into several grids (approximately equal to the size of one scoop required for regular washing) and the component fraction of each grid is estimated using image-processing [29-32, 33, 34]. The scale of scrutiny should be according to the final product specification and is mainly used to determine the ideal size of samples. The grid size chosen for the analysis of the composition in this work represents the required size of scale of scrutiny (based on the regular scoop size in laundry detergent) [35]. For the estimation of component fraction of each grid, the coverage area of particles (Fig. 2-b) is determined, the pixels of the defined area are measured and converted to the fraction data. The step by step image processing for the measurement of the component fractions are explained elsewhere [29].The segregation index (SI) of component is then estimated according to Eq. (1).

$$\mathrm{SI} = \frac{\sigma_{\mathrm{i}}}{\mu_{\mathrm{i}}} \qquad 1$$

where σ_{i} is the standard deviation of the fraction of component i and μ_{i} is the mean fraction of component i in a defined set of grids within the whole area of the mixture. For a perfectly ordered mixture, SI is expected to be equal to zero.

To investigate segregation of coated powders in a dynamic system, the produced heap of powders is also subjected to a vertical vibration by a defined frequency and amplitude using a vibration system as shown in Fig. 2-a, which is controlled by an electrometer, TG315 function generator (AIM-TTI instruments, Cambridgeshire).

Fig. 2. (a) Schematic of segregation set-up and (b) determination of particles coverage area using selection tool of image processing software.

3-Results and Discussion

3-1-Evaluation of the main segregation mechanisms

In this section, the main segregation mechanisms of laundry detergent powder mixtures are presented. Segregation of a binary mixture comprising BP and TAED is first evaluated and then the analysis is extended to a ternary mixture comprising BP, TAED and EP granules. Binary mixtures with different size ratios were first prepared and poured to form heaps as illustrated in Fig. 3-a to Fig. 3-e. The mixture mass was 40 g and had a 50/50 ratio (by mass) of TAED/BP. The sieve cut size of BP was kept constant (425-500 μm) while that of TAED was changed at different size ranges: (425-500), (500-600), (600-710), (710-850) and (850-1000) μm. Figures 3-a to 3-e show how TAED particles segregate towards the corners of the heap as their size is increased. The SI of TAED is presented beside the concentration map of different sizes of TAED in Fig. 3-f, where it can be observed that a better uniformity of the TAED fractions is reached by decreasing the size ratio (the concentration map of TAED for all heaps is provided in Fig. S2, supplementary document). It can be deduced from Fig. 3 that the unity mean sieve size ratio of components results in a better mixing in binary mixture of BP and TAED particles. However, segregation increases as the size ratio is increased.

Fig. 3. Binary mixtures of TAED and BP with different mean sieve cut size ratios of TAED over BP, (a): 1, (b): 1.18, (c): 1.41, (d): 1.68 and (e): 2 and (f) concentration map/SI of TAED particles.

Percolation of smaller particles of BP could take place if the size ratio is such that they can get into the interstices of larger particles of TAED. This process continues to the extent that they can fill the void fraction. According to Savage and Lun [36], Arteaga and Tüzün [37] and Lacey [38], a size ratio typically greater than ~3:1 is needed for the unhindered percolation to happen. Increasing trend of SI as the size ratio is increased could be due to the angle of repose differences between BP (at size range of 425-500 μm) and TAED particles at different sizes (Table 1). Due to the lower angle of repose of larger sizes of TAED particles, they prefer to migrate towards the corners of the heap during the pouring process [39]. Therefore in this case, the effect of percolation of fine BP is negligible compared to the effect of angles of repose of larger particles of TAED.

Table 1. Angle of repose of BP and TAED particles.

Looking at the heaps shown in Fig. 3, some patterns of stratification is also observed, particularly for larger sizes of TAED which is in agreement with the results obtained by Fan et al. [40]. In the work of Fan et al. [40], the stratification for different sizes of spherical particles was studied, whereas no stratification was reported for the small size difference. The authors suggested that stratification was formed due the formation of “kinks” on the slope of the heap for larger size ratios. The stratification pattern could be attributed to the varying angle of repose of ingredients, resulting in “kinks” during filling caused by particle avalanches.

The main segregation mechanisms of ternary mixture are then investigated by adding the EP granules into binary mixtures. For this purpose, some particle characteristics of EP granules are first evaluated and compared to those of BP and TAED particles. It can be inferred from SEM image, G3 morphology and XRT structural analysis of a number of particles that the EP granules are roughly spherical in shape, whilst BP and TAED particles are more of aggregate shape (Fig. 4). Also, the bulk tapped density of EP granules is estimated as 1450 kg.m^{-3} which is nearly three times higher than those measured for TAED (530 kg.m^{-3}) and BP (400 kg.m^{-3}). Bulk tapped density measurement was done by pouring the mixture of powders in a graduated cylinder and tapping the poured powders until the pack reached an equilibrium.

Fig. 4. SEM and XRT photos of a number of (a) EP granules, (b) BP and (C) TAED particles.

40 g ternary mixtures (with equal mass of TAED, BP and EP granules) are then formed by the addition of EP granules into both mixed (with unity size ratio) and segregated binary mixtures of BP and TAED particles (size ratio of TAED over BP=2, Fig. 5). For this purpose, the mode of the narrow size distribution of EP granules (600-710 µm) is used as their representative because it contains majority of the EP granules. In this figure, it can be observed that the EP granules are accumulated towards the centre of both segregated and mixed binary mixtures after pouring process due to presumably their higher density. Although unity size ratio is ideal for a mixed binary mixture, Fig. 5-c reveals that this condition is not favourable to achieve a mixed ternary system in this study, presumably to the higher density of EP granules as compared to the BP and TAED granules.

Fig. 5. Addition of EP granules (mode size: 600-710 µm) into (a) segregated and mixed binary mixtures in size range of (b) 425-500 and (c) 600-710 µm.

To further analyse the segregation mechanisms of EP granules, series of experiments have been carried out using binary mixtures of BP/EP and TAED/EP. For this purpose, EP granules with their mode size have been added to different sieve cut sizes of BP and TAED particles, individually. Figure 6-a shows 50/50 weight percentage ratio binary mixtures (40 g), comprising different sieve cut sizes of BP (sieve cut sizes between 300-500 µm) and the mode size of distribution of EP granules. It can be seen in this Fig. 6-b that the binary mixtures of BP-EP granules with sieve cut sizes of (250-300 µm), (300-355 µm), (425-500 µm) and (500-600 µm) have undergone more segregation as compared to the case of 355-425 µm. It can be inferred that, there is a competition between the segregation of small particles of BP and the segregation of dense particles of EP granules during the heap formation. In general, both small and dense particles are prone to move towards the centre of the pile. Fine BP particles with sieve cut size of less than or equal to 300-355 µm overcame the density segregation of EP granules. On the other hand, density segregation of EP granules (due to push-away effect) could dominate the size segregation of BP particles with sieve cut size of greater than or equal to 425-500 µm. The same behaviour has also been observed for the mixture of TAED/EP granules (Fig. S3, supporting information).

Fig. 6. (a) Binary mixture containing equal weight percentage of BP/EP granules and (b) concentration maps of EP granules.

To verify the segregation mechanisms for EP granules in ternary mixtures (with equal mass of TAED, BP and EP granules), 40 g heaps containing BP and TAED (with different sieve cut sizes of 300-355, 355-425, 425-500 µm) and EP granules (mode size) are then prepared (Fig. 7-a) and the SI of EP particles are measured (Fig. 7-b). It can be observed that a fully-mixed condition of EP granules in the ternary mixture is generated by adding them into the binary mixture of BP and TAED with the narrow sieve cut size of 355-425 *µm*, which is in agreement with the finding of binary mixtures as reported in previous section. The findings suggest that optimising the particle size distribution of components could be a way of reducing the segregation arising from particle size and density variation.

Fig. 7. (a) Ternary mixture and (b) concentration map/SI of EP granules containing equal weight percentage of BP/TAED/EP granules.

It has been mentioned that spherical particles behave like free-flowing particles in mixtures of spherical and irregular particles, preferring to migrate to the surface of the heap. On the other hand, irregular particles build up around the centre of the heap [41, 42]. Angle of repose of particles could be strongly influenced by their shape properties during heap formation. Table 2 shows the angle of repose of different sieve cut sizes of BP and TAED particles. It can be deduced that the angle of repose of EP granules at their mode size, which is obtained equal to 16.01±1.18, is less than those obtained for different sieve cut sizes of BP and TAED particles. Thus in this case, the effect of angle of repose segregation of EP granules could be insignificant for the trends seen in Figs. 6 and 7 compared to segregation arising from other material properties of components, notably size and density.

Table 2. Angle of repose of TAED and BP particles (20 g) at different sieve cut sizes.

In a separate experiment, 40 g ternary mixture heap with equal mass of TAED, BP and EP granules but with full particle size distribution of components was prepared (Fig. 8-a) and compared with that containing the narrow sizes of components proposed for a fully-mixed ternary mixture (Fig. 7). From the SI and concentration map results of components in the case of full particle size distribution (Fig. 8-b), it could be deduced that the segregation is not very intense for the case of BP and TAED particles, presumably due to their wide particle size distribution and the same density. However, SI of EP granules reveals that this component is prone to extensive segregation. Therefore, it is critical to mitigate the segregation of this component to achieve a mixed mixture of laundry detergent powders. This is done by making the EP granules sticky as described in the next section. Another series of experiments were carried out using the exact weight percentage ratio in detergent formulation (92.6 (BP)/ 5.55 (TAED)/ 1.85 (EP granules)) for both full size distributions as well as the optimum sieve cut sizes of components for a mixed ternary system as explained in the previous section. The SI of components in these ternary mixtures were then measured, (heaps are illustrated in Fig. S4, Supporting Information). The SI of EP granules and TAED reduced from 1.36 and 0.55 (for the full particle size distribution) to 0.67 and 0.30 (for the optimum sieve cut sizes of components for mixed ternary system, shown earlier), respectively.

Fig. 8. (a) Ternary mixture containing equal mass of TAED, BP, EP granules with full particle size distribution and (b) concentration map and SI information of the ternary mixtures before and after applying the optimum sieve cut sizes of components.

3-2-Evaluation of the effect of coating on the segregation reduction of EP granules

Using narrow sieve cuts to mitigate segregation is not practical in most industrial manufacturing operations. As a small amount of enzymes is used in detergent formulation, manipulating the surfaces of enzyme granules by making them sticky could be an option for the minimisation of segregation of this component. In this section, the effect of particle coating by a thin liquid layer on the segregation minimisation of EP granules is evaluated. Polyethylene glycol (PEG, Pluriol E 600) is used as coating agent, due to its compatibility with the laundry detergent formulation, as mentioned earlier.

Different amounts of PEG (0.5 to 3.5 wt % with the increment of 1 wt %) were spread over EP granules surfaces to evaluate its effect on their segregation. The coated EP granules were added to the mixture of BP and TAED particles (with full particle size distribution) and mixed to make a ternary mixture (92.6/5.55/1.85 wt % ratio of BP, TAED and EP granules, respectively). Finally, the SI of EP granules was measured after generating the heap of ternary mixtures using image processing and Eq. (1). It has been shown that the segregation of EP granules decreases as coating level is increased (Fig. 9-a, the concentration map of EP granules at different coating levels is provided in supplementary document, Fig. S5 along with the ternary mixture heaps before and after coating.

Liu et al., [43] investigated the effect of cohesion on the radial segregation of granular materials in a rotating drum using DEM. In the case of size-induced segregation, the segregation of granular material was decreased at higher cohesion levels. However, it was demonstrated that at lower cohesion levels, mixing process was even more suppressed than the non-cohesive system. In density-induced segregation, the segregation decreased as cohesion was increased due to the change of flow regime as suggested by the authors. For the case of combined size and density-induced segregation, the segregation pattern could be enhanced or reduced bu cohesion depending on the collaboration or competition of percolation and "buoyancy" mechanisms [43]. In current study, all coating levels seem to be influential for the segregation reduction of EP granules, presumably due to significant change of their flow regimes. It should also be noted that conclusions from the study of Liu et al. [43] may not be extended to this study as they simulated fully spherical particles. Other mechanisms may be involved in the segregation reduction of EP granules in the current study as BP and TEAD particles in the systems are non-spherical. This will be further explored in this work.

Increasing the coating level could adversely influence the flowability of the powders and therefore the optimum coating level for an acceptable flowability of the particles is also required to be explored. For this purpose, the flowability of both coated EP granules and the bulk of mixture was analysed using the Schulze shear cell instrument and FT4 device (Fig. 9-b and c). In the Schulze shear cell, the yield locus of the bulk of powders is determined and expressed in term of the flow function, the ratio of consolidation stress to the unconfined yield strength. In FT4 device, due to gentle upward motion of blade, a low stress environment could be generated in powder bed and hence cohesion becomes a very significant factor. The specific energy (SE) of the bed of powders, defined as the energy required to withdraw a rotating impeller out of a powder bed, could then be estimated. Using both instruments, it is possible to evaluate the flow behavior of coated materials according to the regimes shown in Table 3.

Table 3. General flow regimes in Schulze shear cell and FT4 instrument [44, 45].

It can be observed from Fig. 9-b that the mixture’s flow property does not change significantly with the coating levels of 1.5 to 3.5 wt % (remains in easy-flowing regime and low cohesion regions, respectively for Schulze shear cell and FT4). However, the behaviour of coated EP granules on their own shifts notably to the moderate cohesion in FT4 and cohesive regime in Schulze shear cell, particularly at lower pre-shear, with 3.5 wt % coating level (Fig. 9-c). Based on the values of SI for different coating percentages in Fig. 9-a, 2.5 wt % coating level could be adequate to reduce the segregation of EP granules without compromising the flowability of both mixture and EP granules themselves.

Fig. 9. (a) Concentration map and SI of EP granules versus coating percentage of PEG, (b) flowability of the ternary mixture, and (c) EP granules alone after coating.

Using confocal imaging, it is possible to obtain the cross-sectional view of tiny liquid layers. In this study, 1.5 to 3.5 wt % coating levels of PEG on the EP granules have been analysed using a Zeiss LSM880 confocal laser scanning microscope. For this purpose, a small amount of fluorescent pigment was added to PEG which enabled differentiation between EP granule and the coating liquid. The liquid coating is shown by green, red and blue colour for 1.5, 2.5 and 3.5 wt % coating levels of PEG, respectively (Fig. 10). It can be qualitatively observed that the liquid bridge volume is increased by increasing the coating percentage. Also, the number of bridges could increase as the coating level is increased. The top view of the 3D liquid coverage of EP granules are further analysed using image processing, as shown in Fig. 11. It is observed that the percentage of surface liquid coverage is increased from 17 to 24 and 28 % by increasing the coating level from 1.5 to 2.5 and 3.5 wt %, respectively. The percentage of liquid coverage for the optimum coating percentage (2.5%) is estimated as 24 %.

Fig. 10. Cross sectional views image of coating layer obtained by confocal laser scanning microscope, (a) 1.5, (b) 2.5 and (c) 3.5 wt %.

Fig. 11. Top view picture of 3D particle coverage by PEG obtained by confocal laser scanning microscope.

The SEM image of 2.5 wt % coated EP granules after mixing with TAED and BP is shown in Fig. 12. Attachment of fine BP particles onto the surfaces of the coated EP granules could modify their shape and produce rougher surfaces. This could reduce their free movement in the mixture by the process of interlocking leading to a reduction of the segregation of EP granules as compared to the case of free-flowing round granules. In fact, the process of interlocking of some rough EP granules could mitigate segregation, retarding their movement to the center of the pile. Attachment of BP granules on the surfaces of EP granules could even change the restitution of the EP granules on impact. For this purpose, the impact of a single EP granule (before and after BP coverage) to the surface covered by BP particles, was filmed by a high-speed camera (videos of particle impact are provided in the supplementary document). No bouncing of the EP granule (when it was covered by BP particles) could be observed after the impact (Video entitled Particle 1, probably due to the interlocking between the rough EP granules and the bed of BP particles), whilst the round EP granules bounced off the surface (Video entitled Particle 2). It should be noted here that due to the size range and the structural properties of particles as well as the exerted stress during discharging process, the surfaces of EP granules have not been fully covered by BP particles, enabling their approximate SI determination using image processing after filling process. Nevertheless, the conclusions drawn in this study are in a good agreement with the DEM simulation of this powder system [46]. In the study of Alizadeh et al. [46], JKR theory was applied to simulate the cohesion of coated EP granules in the ternary mixture containing BP, TAED and EP granules.

Fig. 12. SEM photo of coated EP granules with PEG after mixing with TAED and BP.

Mobility of EP granules (uncoated and coated by PEG) in the mixture has also been tested by exposing the ternary mixture to the vibration. Segregation of components could become intense during vibration, a condition occurring particularly during transportation in trucks caused by the road conditions, left or right turns and/or difference in road levels [47]. An intense vibration has been applied (frequency= 50 Hz, amplitude=15 mm) for the bulk of powder mixtures (containing 92.6/5.55/1.85 wt % ratio of BP, TAED, EP granules) using the system shown in Fig. 2-a. The EP granules moved upward from the center towards the apex of the heap after vibration and move towards the corners of the heap (Fig. 13-a).

From the comparison of segregation of EP granules during filling and vibration, it could be deduced that unlike the filling case where density-induced segregation due to push-away effect could drive the EP granules towards the centre of the heap, up-thrusting or Brazil nut effect phenomenon may dominate during vibration. Different factors could influence the Brazil nut effects of particles during vibration, such as particle size ratio and percolation [48], vibration frequency and amplitude [49], internal friction and elasticity [50, 51] and the particle density [52]. Therefore, both material characteristics and vibration conditions are contributing to the Brazil nut effect of EP granules during vibration. For instance, the Brazil nut effect of EP granules was negligible when the ternary mixture was exposed to the moderated vibration as compared to the intense vibration (frequency= 50 Hz, amplitude=15 mm), Fig. S6, supplementary document. Thus, the segregation of coated and uncoated EP granules in the ternary mixtures during vibration has been investigated when they are exposed to the intense vibration condition (frequency= 50 Hz, amplitude=15 mm), where Brazil nut effect was significant.

As shown in Figs. 13-a and 13-b, the coated EP granules are less segregated in the corners of the heap after 900 s vibration as compared to the case of uncoated ones. This might be due to the reduced mobility of the rough EP granules (SEM photo is shown in Fig. 12) by the interlocking effect during vibration. SI of EP granules versus time of vibration before and after coating is illustrated in Fig. 13-c. It can be deduced from Fig. 13-c that the SI of coated EP granules in a mixture is less than that of uncoated one at different times of vibration, particularly after achieving the equilibrium (at around 300 s).

Fig. 13. Vibrated heap of ternary mixture (a) without coating and (b) with coating of EP granules (time of vibration= 900 s), (c) SI versus time of vibration obtained for the ternary mixtures with and without coating (frequency= 50 Hz, amplitude=15 mm).

Overall, the combination of the interlocking effect between particles and particle size/restitution property changes after the coating of EP granules could be influential on their segregation reduction during filling and vibration. The main underlying mechanisms of segregation reduction of EP granules after coating is recommended to be further investigated by quantifying the interparticle forces and tracking each particle at desired time using DEM simulation, which is the scope of further research.

4. Conclusions

The effect of physical properties of particles, notably size, shape and density, on the segregation behaviour of components in formulated detergent powder mixture has been investigated. For a ternary mixture, sieve cut size of BP and TAED particles in the range of 355-425 μm has been found as optimum criteria for fully-mixed EP granules (with the mode particle size distribution). The SI of EP granules could be reduced from 1.36 to 0.67 by applying this size rather than using their full particle size distribution of BP and TAED in the ternary mixture. The effect of coating of EP granules on their segregation reduction is studied. From flowability test results, it is shown that 2.5 wt % PEG could be enough to minimise the segregation of the low content level ingredient EP granules in the ternary mixture without compromising the flowability of the mixture as well as EP granules themselves. It has been observed that fine particles of BP and TAED components adhere to the coated EP granules and produce rough surfaces which could reduce the mobility of EP granules by enhancing the process of interlocking in the ternary mixture. The SI of EP granules reduces from 1.36 to 0.69 using the proposed optimum coating percentage in the ternary mixture with full particle size distribution of the components.

5. Acknowledgments

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Table 1. Angle of repose of BP and TAED particles.

	BP	**TAED**				
sieve cut size (*μm*)	**425-500**	**425-500**	**500-600**	**600-710**	**710-850**	**850-1000**
Angle of repose, °	36.06±1.63	33.43±1.13	32.91±2.91	31.93±1.17	28.80±0.75	26.28±1.90

Table 2. Angle of repose of TAED and BP particles (20 g) at different sieve cut sizes.

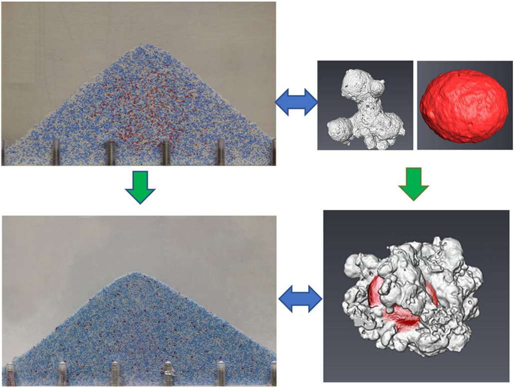
		Sieve cut size (*μ*m)				
		250-300	**300-355**	**355-425**	**425-500**	**500-600**
Angle of repose, °	**BP**	40.01±1.07	39.50±1.78	39.38±1.37	36.06±1.63	34.48±1.68
	TAED	39.7±1.55	37.11±1.27	34.71±1.02	33.43±1.13	32.91±1.91

Table 3. General flow regimes in Schulze shear cell and FT4 instrument [39,40].

Schulze shear cell, FFC		**FT4, SE (mJ/g)**	
FFC<1	Not flowing	SE<5	Low cohesion
1<FFC<2	Very cohesive	5<SE<10	Moderate cohesion
2<FFC<4	Cohesive	SE>10	High cohesive material
4<FFC<10	Easy flowing		
FFC>10	Free flowing		

Highlights

- Segregation mechanisms of particles in ternary washing powder mixtures are investigated.
- Mitigation of segregation of low content level enzyme granules in detergent powders is evaluated.
- An optimum cohesion level to reduce the segregation of low content level enzyme granules is reported.



Graphics Abstract

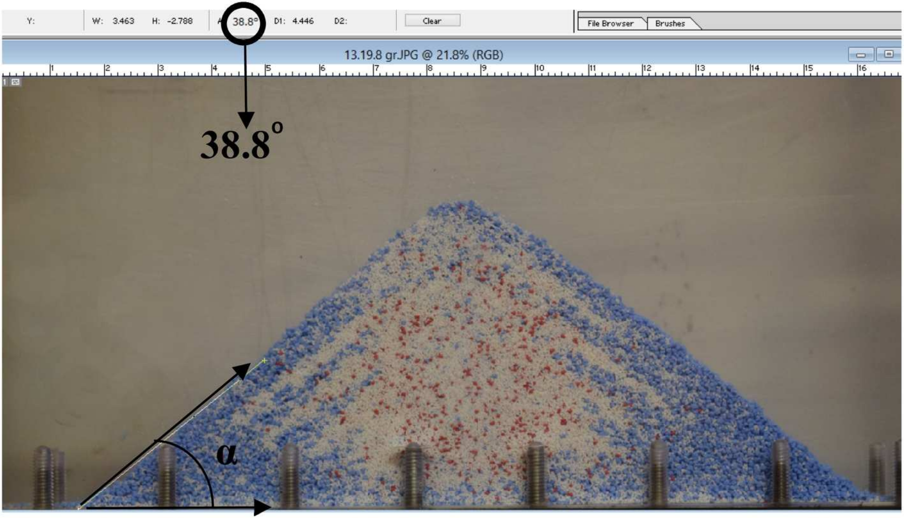


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Figure 1

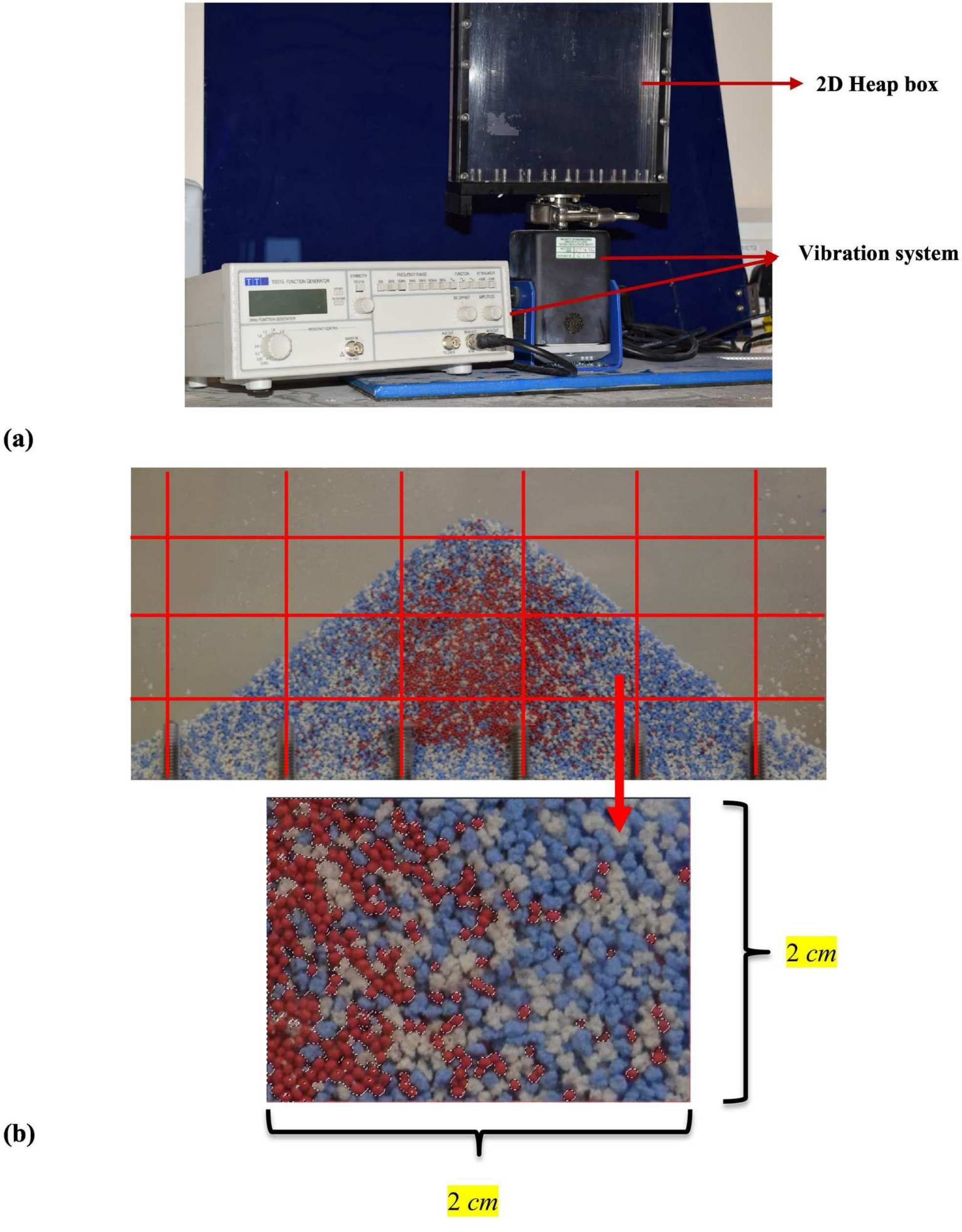


Figure 2

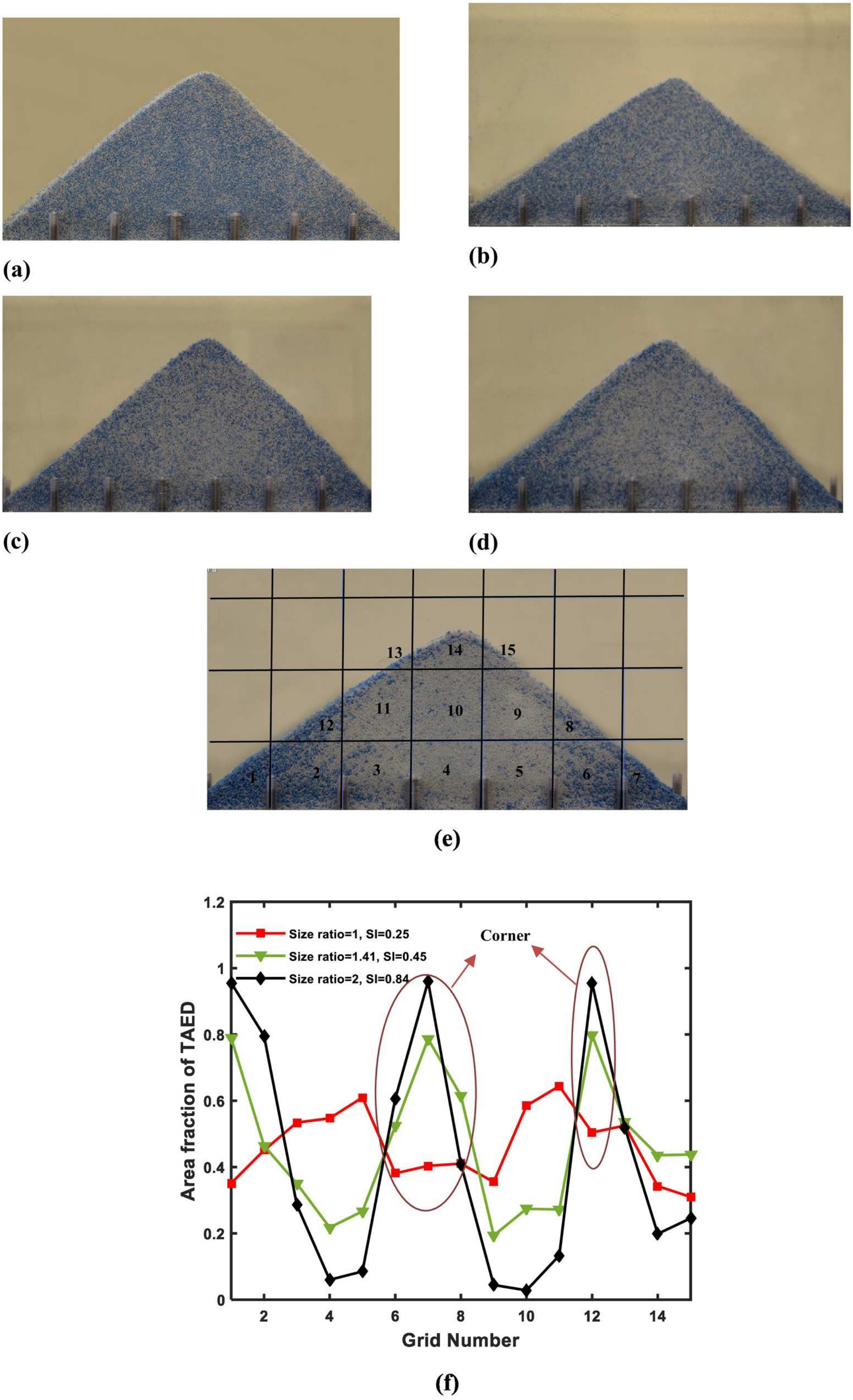

(a)
(b)
(c)
(d)
13
14
15
11
10
9
12
8
1
2
3
4
5
6
7
(e)
Size ratio=1, SI=0.25
Size ratio=1.41, SI=0.45
Size ratio=2, SI=0.84
Corner
Area fraction of TAED
Grid Number
0
0.2
0.4
0.6
0.8
1
1.2
2
4
6
8
10
12
14
(f)

Figure 3

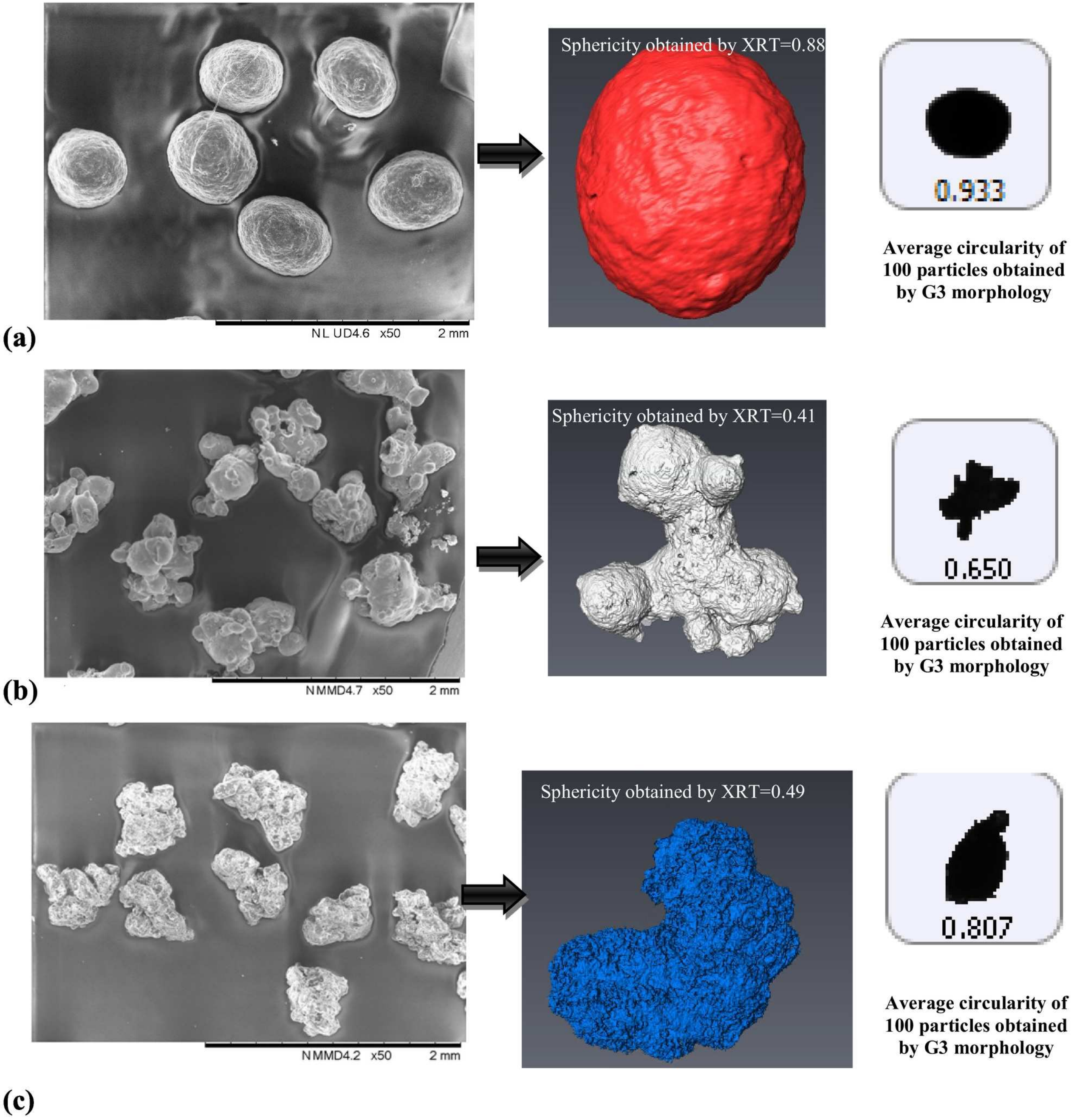
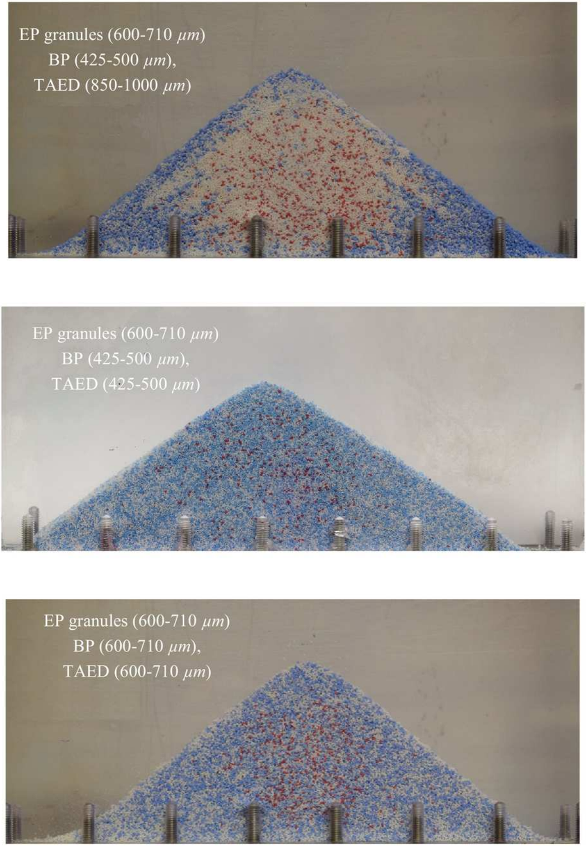

NL UD4.6 x50 2 mm
Sphericity obtained by XRT=0.88
0.933
Average circularity of 100 particles obtained by G3 morphology
(a)
NMMD4.7 x50 2 mm
Sphericity obtained by XRT=0.41
0.650
Average circularity of 100 particles obtained by G3 morphology
(b)
NMMD4.2 x50 2 mm
Sphericity obtained by XRT=0.49
0.807
Average circularity of 100 particles obtained by G3 morphology
(c)

Figure 4


EP granules (600-710 μm)
BP (425-500 μm),
TAED (850-1000 μm)
EP granules (600-710 μm)
BP (425-500 μm),
TAED (425-500 μm)
EP granules (600-710 μm)
BP (600-710 μm),
TAED (600-710 μm)

(a)

(b)

(c)

Figure 5

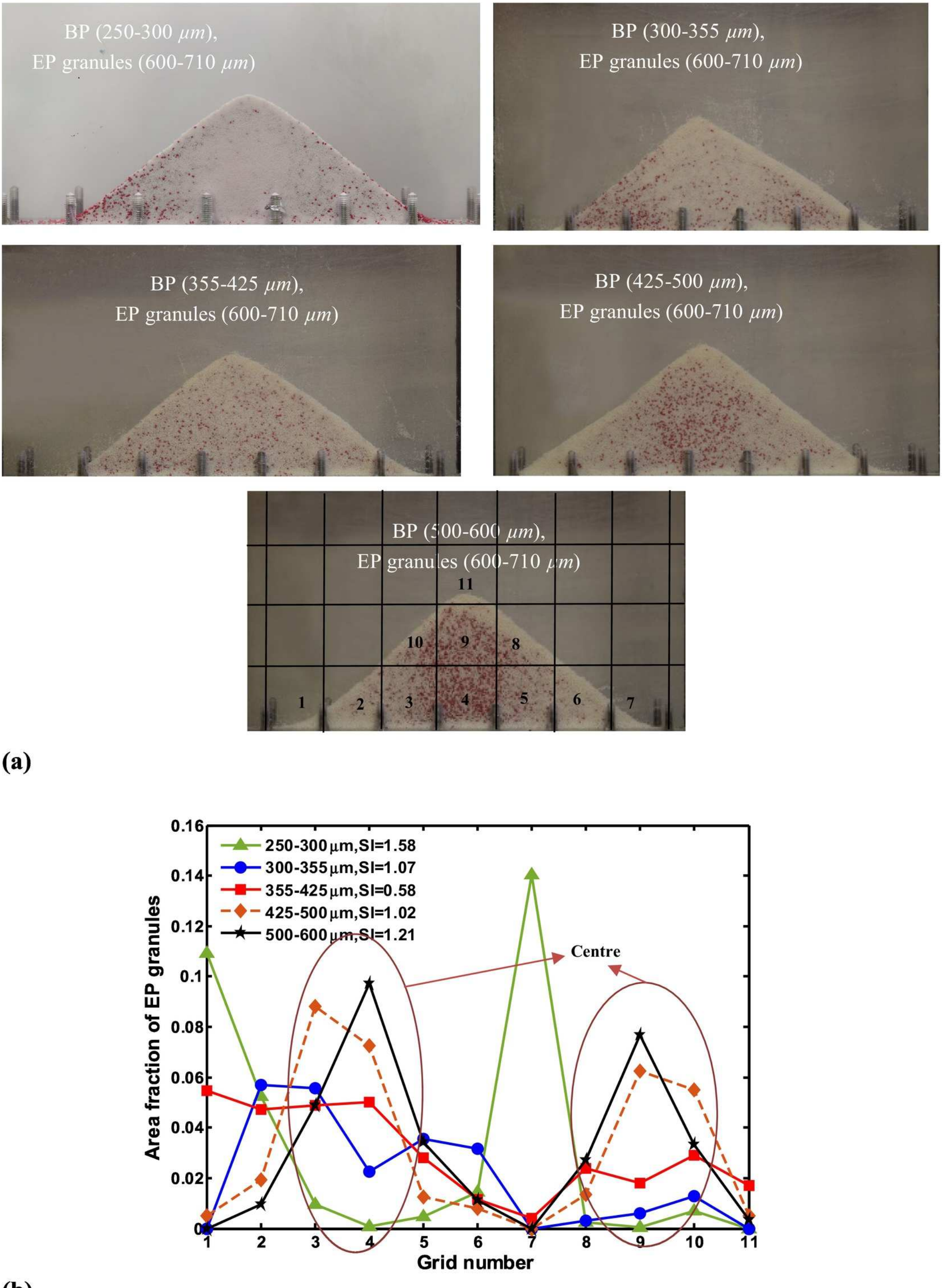
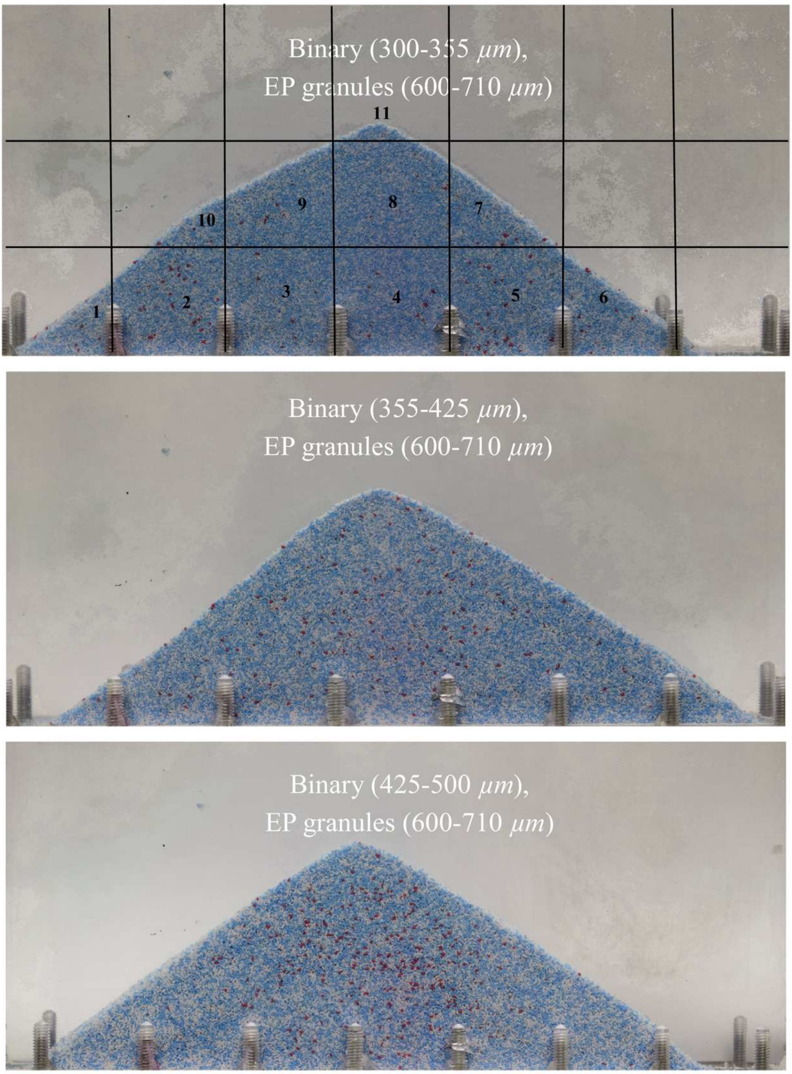
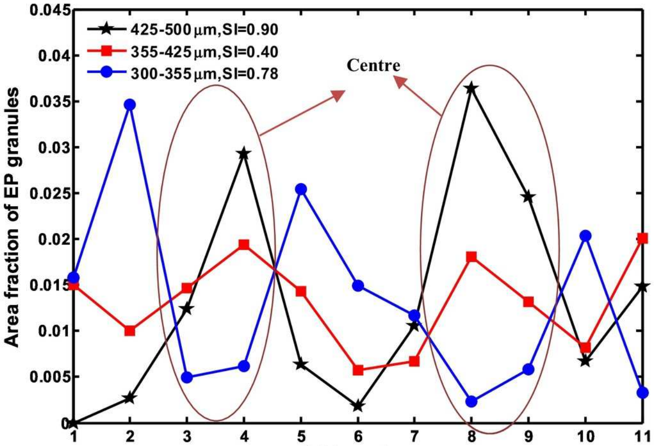

BP (250-300 μm),
EP granules (600-710 μm)
BP (300-355 μm),
EP granules (600-710 μm)
BP (355-425 μm),
EP granules (600-710 μm)
BP (425-500 μm),
EP granules (600-710 μm)
BP (500-600 μm),
EP granules (600-710 μm)
11
10
9
8
1
2
3
4
5
6
7
(a)
0.16
0.14
0.12
0.1
0.08
0.06
0.04
0.02
0
Area fraction of EP granules
250-300μm,SI=1.58
300-355μm,SI=1.07
355-425μm,SI=0.58
425-500μm,SI=1.02
500-600μm,SI=1.21
Centre
1
2
3
4
5
6
7
8
9
10
11
Grid number
(b)

Figure 6


Binary (300-355 μm),
EP granules (600-710 μm)
11
10
9
8
7
1
2
3
4
5
6
Binary (355-425 μm),
EP granules (600-710 μm)
Binary (425-500 μm),
EP granules (600-710 μm)

(a)


425-500 μm,SI=0.90
355-425 μm,SI=0.40
300-355 μm,SI=0.78
Centre
Area fraction of EP granules
Grid number
0
0.005
0.01
0.015
0.02
0.025
0.03
0.035
0.04
0.045
1
2
3
4
5
6
7
8
9
10
11

(b)

Figure 7

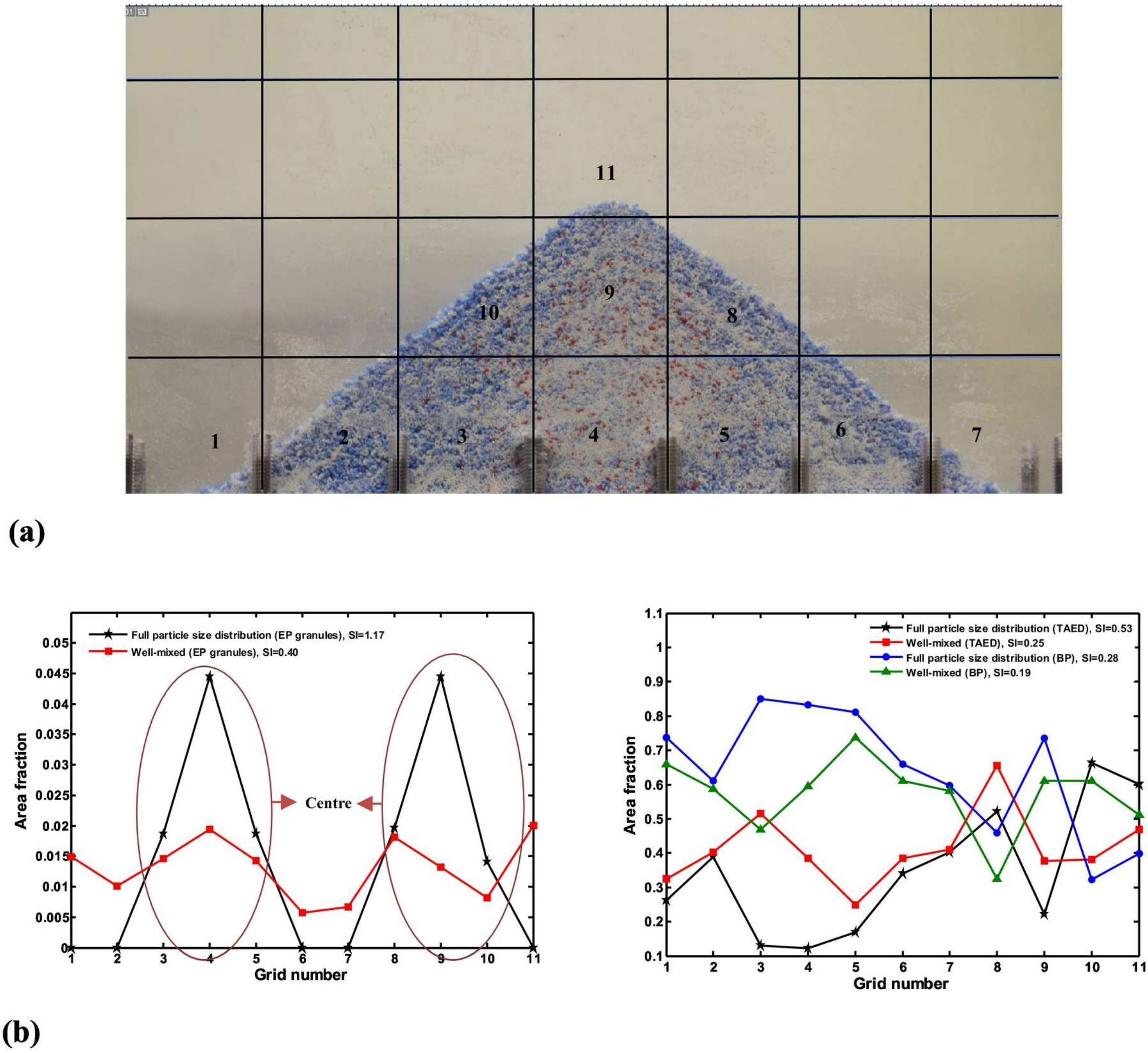
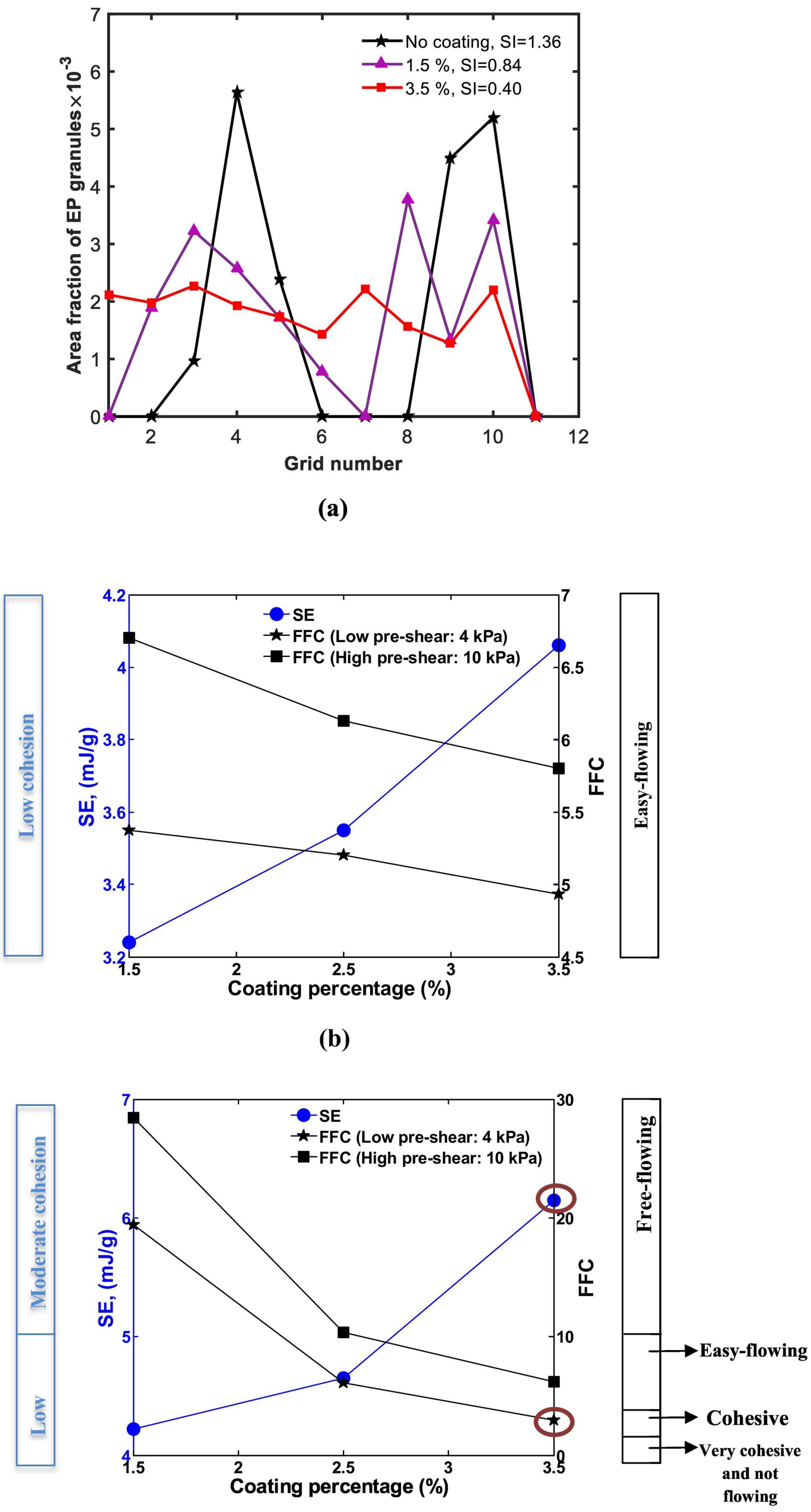

11
10
9
8
1
2
3
4
5
6
7
(a)
Full particle size distribution (EP granules), SI=1.17
Well-mixed (EP granules), SI=0.40
Centre
Area fraction
Grid number
0
0.005
0.01
0.015
0.02
0.025
0.03
0.035
0.04
0.045
0.05
1
2
3
4
5
6
7
8
9
10
11
Full particle size distribution (TAED), SI=0.53
Well-mixed (TAED), SI=0.25
Full particle size distribution (BP), SI=0.28
Well-mixed (BP), SI=0.19
Area fraction
Grid number
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
1.1
(b)

Figure 8


Area fraction of EP granules×10^-3
Grid number
0
1
2
3
4
5
6
7
2
4
6
8
10
12
No coating, SI=1.36
1.5 %, SI=0.84
3.5 %, SI=0.40
Low cohesion
SE, (mJ/g)
FFC
Coating percentage (%)
3.2
3.4
3.6
3.8
4
4.2
4.5
5
5.5
6
6.5
7
1.5
2
2.5
3
3.5
SE
FFC (Low pre-shear: 4 kPa)
FFC (High pre-shear: 10 kPa)
Easy-flowing
Moderate cohesion
Low
SE, (mJ/g)
FFC
Coating percentage (%)
4
5
6
7
0
10
20
30
1.5
2
2.5
3
3.5
SE
FFC (Low pre-shear: 4 kPa)
FFC (High pre-shear: 10 kPa)
Free-flowing
Easy-flowing
Cohesive
Very cohesive and not flowing

(a)

(b)

(c)

Figure 9

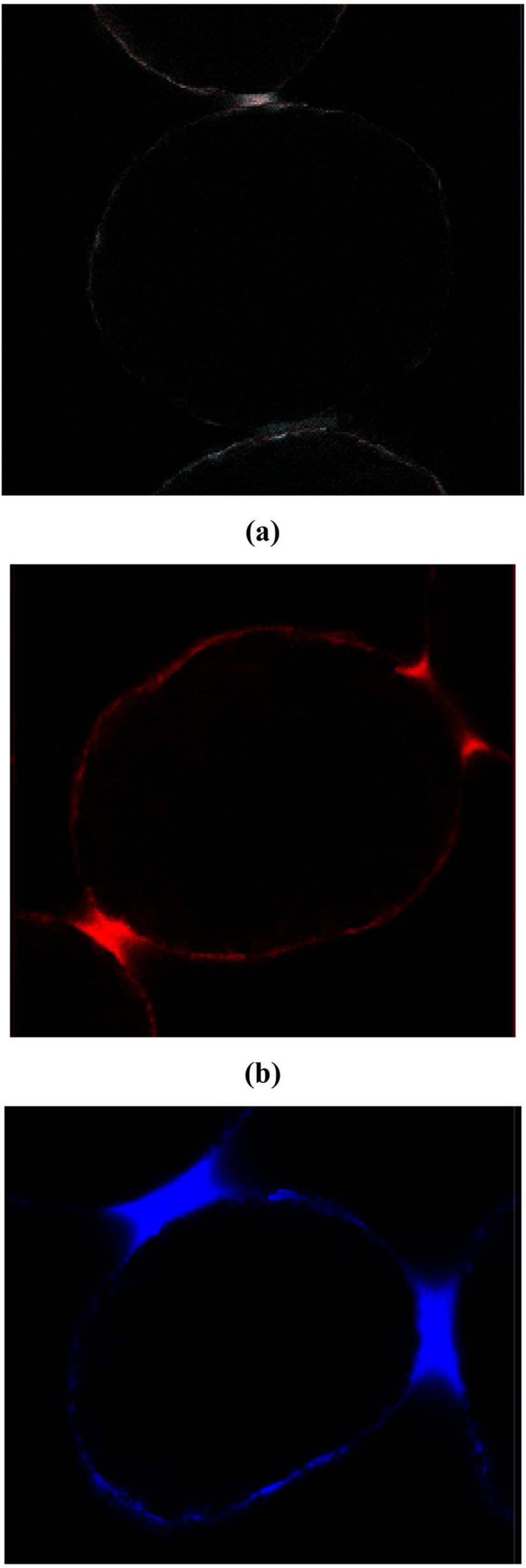


Figure 10

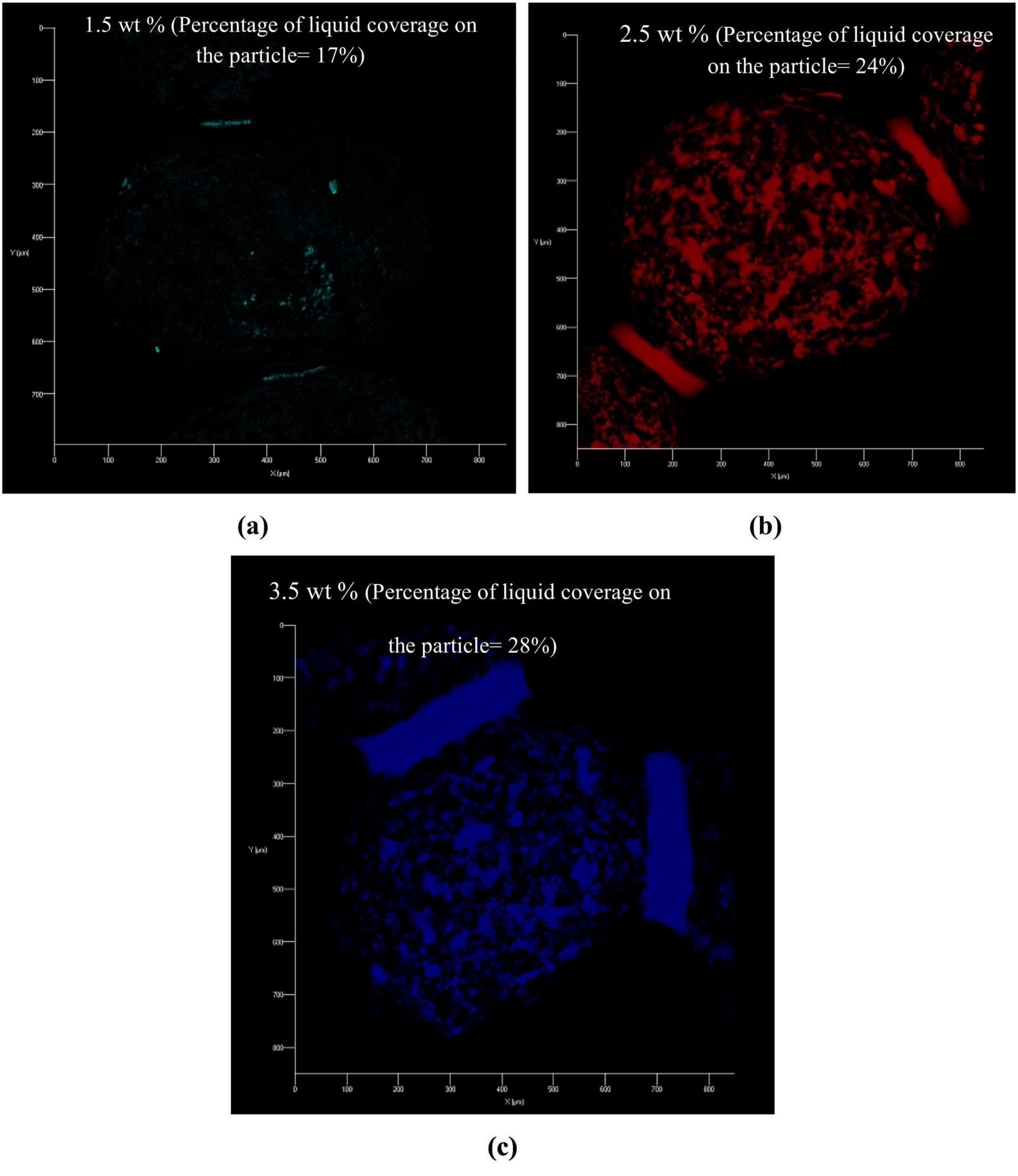

1.5 wt % (Percentage of liquid coverage on the particle= 17%)
2.5 wt % (Percentage of liquid coverage on the particle= 24%)
3.5 wt % (Percentage of liquid coverage on the particle= 28%)
Y [µm]
X [µm]
0
100
200
300
400
500
600
700
800
(a)
(b)
(c)

Figure 11

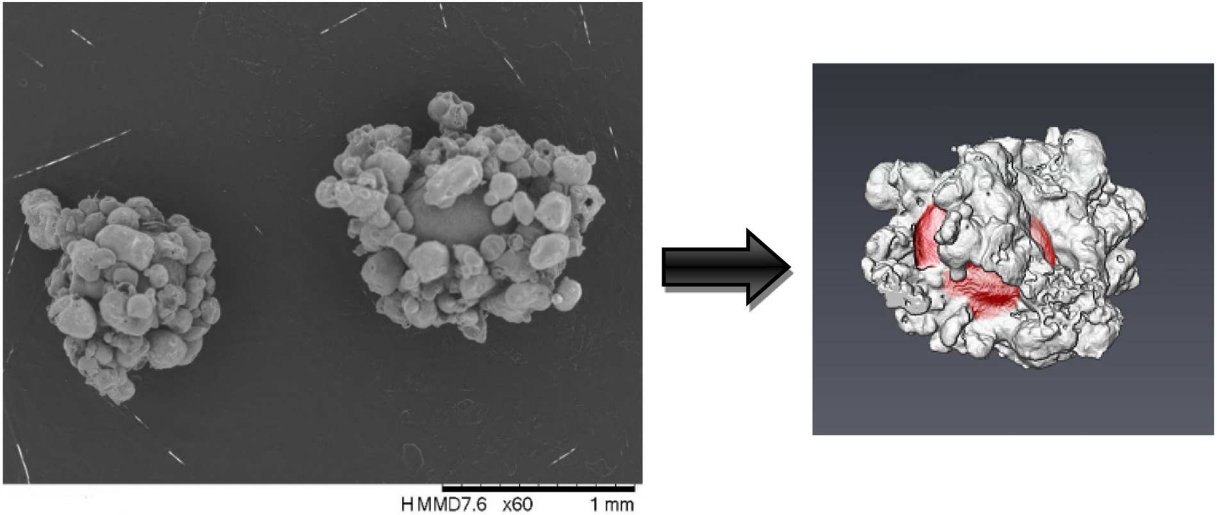


Figure 12

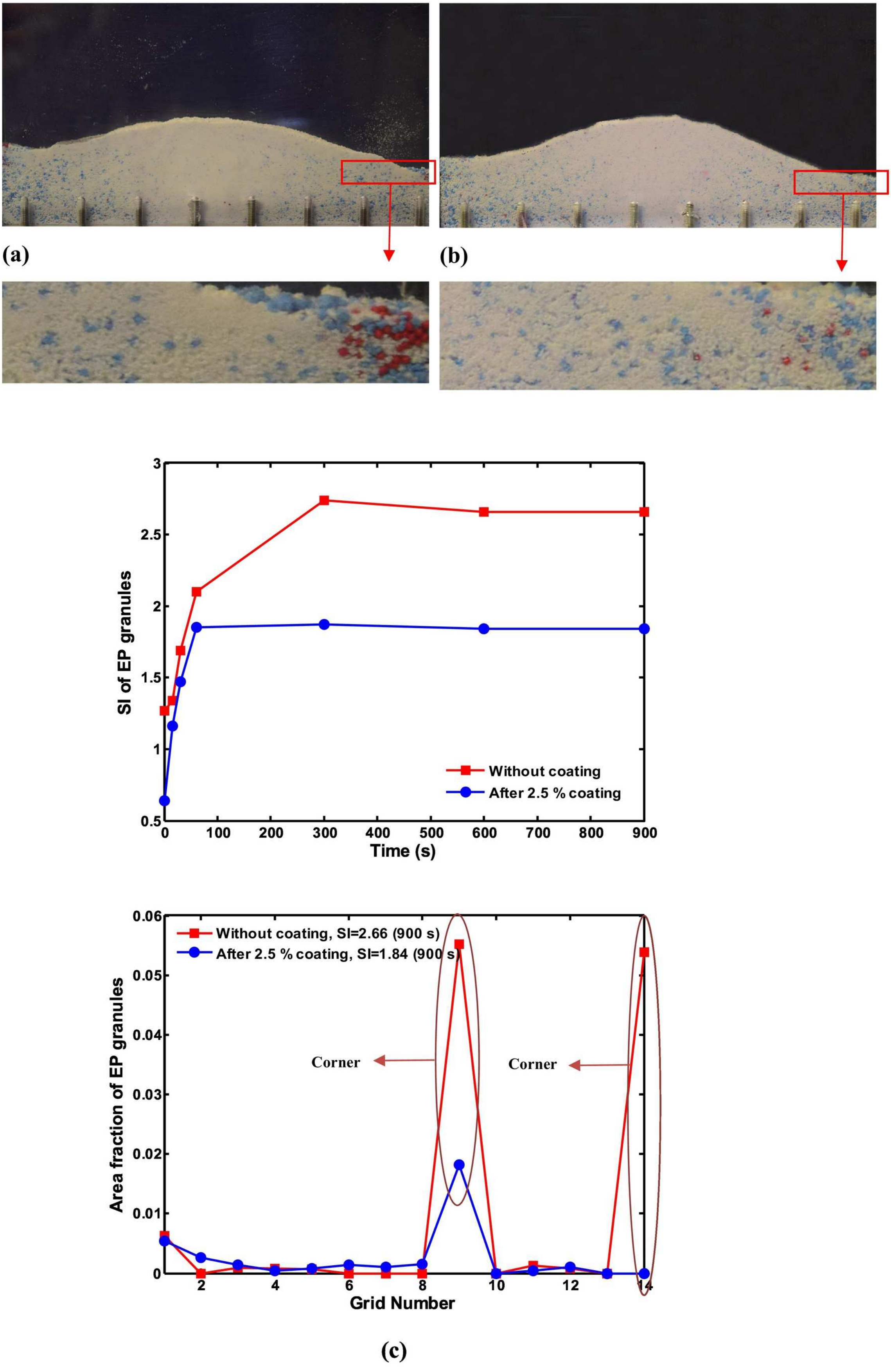

(a)
(b)
SI of EP granules
Time (s)
0
100
200
300
400
500
600
700
800
900
0.5
1
1.5
2
2.5
3
Without coating
After 2.5 % coating
Area fraction of EP granules
Grid Number
Without coating, SI=2.66 (900 s)
After 2.5 % coating, SI=1.84 (900 s)
Corner
Corner
0
0.01
0.02
0.03
0.04
0.05
0.06
2
4
6
8
10
12
14
(c)

Figure 13