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# Tailoring particle shape for enhancing the homogeneity of powder mixtures: Experimental study and DEM modelling

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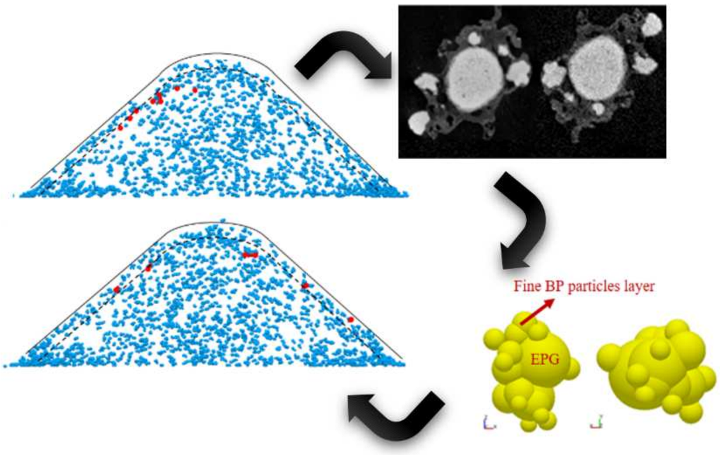
**Abstract:** The effect of particle shape modification on the segregation reduction of enzyme granules in laundry detergent powder mixtures was investigated, both experimentally and computationally using Deseret Element Method (DEM). The shape of modified enzyme particles was in such a way that the large and dense enzyme particles were layered by other fine particles in the detergent powder, by means of a process known in the literature as "seeded granulation". It is found that the homogeneity of modified enzyme particles could be improved significantly comparing to the original spherical enzyme particles in powder mixtures. Overall, the results of this research demonstrated that the segregation-induced properties of the dense/spherical enzyme particles could be lowered by altering their shape, which could enable the enzyme particles to behave almost similar to other ingredients during the pile formation process.

**Keywords:** Segregation Minimisation, Particle Shape Modification, Seeded Granulation, Near-Infrared Spectroscopy, Laundry Detergent Powders, Discrete Element Method

## Graphical abstract



## Highlights:

- Segregation of enzyme particles was evaluated experimentally and computationally.
- Structural shape modification was applied to mitigate the segregation of enzymes.
- Seeded granulation process was used to alter the surface properties of enzymes.

## 1. Introduction

Segregation of particles (i.e. powder inhomogeneity) during the build-up of discharging granular materials is ubiquitous in different powder industries including detergent, food, pharmaceutical, and mining [1-5]. Segregation reduces the process efficiency by producing a low-quality product. Filling the silos, bins, and hoppers with granular materials of different properties are typical examples of the processes where segregation could occur. In detergent powder formulations, inhomogeneity of components particularly minor ingredients could lead to significant economic and health issues [6, 7]. A typical laundry detergent powder product is a complex formulation containing the surfactants, bleaching agents, and auxiliaries such as enzymes. Asachi et al. [8, 9] demonstrated that the minor ingredient of enzyme granules tends to segregate towards the centre of the granular pile due to its higher density as compared to other ingredients, as a result of the push-away effect. Laundry detergent powder products are consumed in daily life and their optimum quality is demanded by consumers. Furthermore, their quality must be strictly monitored as they could adversely impact human health in case of presence of excess auxiliary components such as enzymes.

The effect of various operational parameters [10-12] including the geometry and design [13, 14], as well as material properties such as cohesion [15, 16, 17], particle size and density [10, 18] on the homogeneities improvement of powder mixtures has been widely investigated in the past literature. In a work performed by Hajra et al. [19], a method based on modifying the geometry design by locating an axially-located baffle has been proposed to hinder the density-driven segregation in a binary mixture containing cellulose acetate beads and glass beads. According to their results, the best mixing performance was achieved when the baffles were located within the shear layers during mixing process. In another research, the effect of cohesion on the homogeneity improvement of glass beads was studied by adding liquid binder into the mixture [15]. For this purpose, two types of beads were investigated having the same size but different densities. It was shown that by adding more liquid binder in the mixture, the cohesive force between particles was strengthened and therefore, the extent of segregation was hindered. In a separate study, Liao [20] found out that the density-induced segregation of glass beads could be diminished upon the addition of a high-viscosity liquid due to the formation and rupture of the liquid bridges. The increased energy dissipation due to the implementation of a high-viscosity liquid during the mixing process was reported to mitigate the density-driven segregation of heavy beads.

All the aforementioned approaches could be utilized for segregation reduction of dense particulates in laundry detergent power products; nevertheless, diminishing of segregation by modification of process equipment may not be a reasonable option due to the high level of demanded cost. On the other hand, adding liquids into the powder mixtures may promote the risk of compromising the flowability of the final product and particularly the coated material itself [9]. Furthermore, segregation mitigation of granular materials by means of an optimized size/density range, which has been the subject of a large number of studies reported in the literature [10, 18, 9], is not advised due to the costly, laborious, and time-consuming nature of the process. It should be noted that the experimental investigation of the segregation reduction of granular materials using a convenient method based on particle shape modification has received much less attention in the literature. Following the research of Asachi et al. [9], adherence of fine particles onto the surfaces of sticky dense enzyme granules (which produced particles with extremely rough surfaces) was hypothesized to decrease the segregation of enzymes by restricting the particulate free motions during the pile build-up process as a result of interlocking effect between particles. In the current study, which is continuous to our previous research [9], a simple methodology based on seeded granulation technique is utilized to modify the shape properties of spherical/dense enzyme granules. The effect of shape modification on the density-driven segregation of granules has been further evaluated both experimentally and computationally using Discrete Element Method (DEM).

DEM has great potential in modelling particulate systems. Researchers try to incorporate the effect of shape into DEM simulations in various ways, such as using spherical particles with an artificial rolling friction coefficient, clumped spheres, polyhedral shapes, and digitisation techniques [21]. Recently, Alizadeh et al. [22] showed that using spherical particles with calibrated rolling friction leads to underestimation of the segregation tendency of particles, whereas, the clumped sphere approach predicts the segregation with reasonable accuracy. This is mainly due to the fact that irregular particles experience interlocking and, therefore, have less tendency for segregation as compared to their spherical counterparts. In the current study, the effect of shape is therefore evaluated by implementing the clumped sphere approach due to its superiority over the rolling friction approach. Although the numerical modelling of the effect of shape on granular segregation is investigated by some researchers [23-26], the idea of designing and manufacturing new particle shapes which resist segregation is scant in the literature. This study tries to benefit from both experimental and DEM techniques to evaluate the potential of the shape modification approach for the reduction of density-induced segregation in laundry detergent powder products.

## 2. Methodology

### 2.1. Materials

Blown Powder (BP) as active cleaning agent in detergent formulations and Tetra Acetyl Ethylene Diamine (TAED) as bleach agent were supplied by Procter and Gamble (P&G) Ltd. Enzyme Placebo Granules (EPG), representing the actual enzyme granules in laundry detergent powder formulation, was supplied by DuPont, USA. It should be noted that the particles have different colours, which allow particle position detecting in the ternary powder mixture using image processing analysis. Detailed information about the properties of the aforementioned materials is available elsewhere [9]. The enzyme is responsible for accelerating the biochemical reaction to improve the cleaning efficiency in detergent powders [27]. The images of Scanning Electron Microscopy (SEM) of a number of particles (Fig. 1) as well as the bulk tapped density measurements demonstrated that the EPG is denser and more spheroid in shape than other two ingredients (BP and TAED). An aqueous solution of Polyethylene glycol (PEG 4000) was used as a binder during the shape modification process of EPG. Shape modification of particles was performed using the seeded granulation process which is described in the next section.

## 2.2. Seeded granulation process

Seeded granulation methodology was introduced by Rahmanian et al. [28] for producing the granules with large particles at their core surrounded by fine particles. Similar approach was utilized in the current research to eliminate the density-driven segregation of EPG in detergent powders. Layering of fine BP particles on the surfaces of EPG, as larger nuclei/seeds, was performed to modify their shape. A high shear wet granulator (MiPro, ProCepT, Zelzate, Belgium) was used for the process (Fig. 2). Seeded granulation effectiveness is mainly a function of the feed particle size distribution and the operating conditions of the granulator [28]. It should be noted that an optimum condition for obtaining maximum seeded granules can be achieved when the feed powder contains adequate number of large particles so as they could be entirely covered by fine particles. Rahmanian et al. [28], tested different powders with different primary particle size distributions (Calcium carbonate of different commercial grades, including the coarse Durcal 65 ($d_{50}$ = 63 μm) and the finer Durcal 40 ($d_{50}$ = 43 μm) and Durcal 15 ($d_{50}$ = 23 μm)). They showed that the formation of seeded granules was heavily dependent on the particle size distribution of feed powder. It was demonstrated that there must be enough number of large particles to act as seeds for the seeded granules to form. Durcal 65 granules showed a seeded structure owing to the fact that 18 wt.% of particles were larger than 125 μm, but this was not the case for the medium and fine Durcal powders with only 4.2 and 3 wt.% of particles larger than this size, respectively. Ultimately, efficient seeded granules were produced by mixing fine calcium carbonate powder (Durcal 15) and a sieved fraction of Durcal 65 containing particles larger than 150 μm. In the current study, the mode size of particle size distribution was used for seeded granulation. It should be noted that the size distribution of the enzyme particles was really narrow with most of the particles in the range of 600–710 μm (detailed information about the particle size distribution is available elsewhere [9]). On the other hand, BP particles were smaller in the size than the enzyme particles, with most of the particles in a size range between (212–250 μm). For the granulation test, the bowl of granulator (250 mL) was filled with 100 g of EPG (600–710 μm) and BP (212–250 μm) and then 5% of binder was sprayed over the powder mixture. The optimum ratio of EPG to BP was calculated equal to 0.80 by assuming that the surfaces of EPG completely were covered by the BP particles. Polyethylene glycol (70-wt% aqueous solution, molecular weight: 4000), as a well-known binder [28-31], was used to adhere BP particles to the surfaces of EPG.

The granulation process was performed at three different speed levels including low (300 rpm), medium (500 rpm) and high (700 rpm) and several samples were taken at time intervals equal to 30, 60 and 90 s.

Particle size distribution and the shape of extracted samples (around 20 g) were analysed after drying in an oven, by means of standard sieve analysis and optical microscopy, respectively.

### 2.3. Powder homogeneity evaluation techniques

In a recent research, different methods for the analysis of powder blend uniformity were described and compared (Asachi et al. [32]), where they were categorized into two main groups of wet-based and dry-based techniques. In the current research, dry-based techniques were selected for the homogeneity analysis of enzyme particles which are advantageous over the wet-based techniques due to their fast analysis time. Furthermore, the material does not need to be dissolved in a solution by adopting these techniques, thereby eliminating the production of liquid waste [32]. A heap box similar to detergent powder package (Fig. 3) was implemented to evaluate the segregation of detergent powder. For this purpose, the mixture of powders was poured through a fixed funnel on top of the heap box. For the measurement of the segregation index, the image taken from the surface of the heap was first divided into several segments and then the component fraction of all sections was estimated using image-processing technique to estimate the coefficient of variation (CoV) of the component fractions. Detailed information of the image processing technique for the segregation measurement of particles is provided in our previous research [9]. Extraction of samples from the formed heap was further performed by inserting the horizontal and vertical blades into the generated pile for subsection division. The extracted sub-samples were then scanned using Near-Infrared spectroscopy for the segregation analysis (following the optimised approach presented in our previous work [8]). In fact, adopting the latter technique for the segregation analysis could be a better representation of the component homogeneity within the entire powder mixture. Finally, the experimental process of the heap formation was simulated in DEM with the same geometrical specifications as the experiments to verify the experimental results. Physical parameters similar to the experimental conditions were used in DEM simulation (detailed information of the calibration of this model system using DEM is described elsewhere, [33]). All the aforementioned techniques used for the segregation evaluation of EPG, before and after shape modification, are summarized in Fig. 3.

## 3. Results and discussions

### 3.1. Seeded granulation technique for the EPG shape modification

There are several mechanisms including wetting and nucleation, coalescence and growth, consolidation and breakage which contribute to the formation of granules [34-36]. In our case, the mechanisms of wetting and nucleation have been controlled to produce EPG covered by fine BP without significant particle growth. This means that the surface properties of EPG were modified in such a way that minimum amount of granule growth and self-aggregation of EPG or BP particles could be yielded. The ideal size of the seeded granules could be obtained after a layer surface coverage of BP particles over EPG, referred in this work as primary seeded granules. With attention to the particle sizes of EPG and BP, the ideal size of primary seeded granules must be in a range of $850<D<1180$ μm after the termination of granulation process. Fraction of particles with a size smaller than 850 μm, mainly represented unseeded EPG (size range between 600 to 850 μm) and fine BP particles (size smaller than 600 μm*)*, while fraction of particles with size larger than 1180 $\mu m$ showed overgrown particles (Fig. 4).

The influence of the granulation time and impeller speed, as the main operating conditions, was studied on the trends of particle size distribution (Fig. 5) [28].

It can be observed in Fig. 5 that increasing the granulation time from 60 to 90 s, has negative effect on the quality of produced seeded granules due to the decreased generation of primary seeded granules and the production of lumps. Therefore, granulation time equal to 60 s was chosen as an optimum time for modification of the EPG. In addition, the fraction of products within size range of 850–1180 μm increased when impeller speed was increased. The main reason for that is probably due to generation of higher kinetic energy at higher impeller speeds which could promote the collision and attachment of particles [28]. Similarly, Rahmanian et al. [28] observation showed that the lower impeller speeds lead to the less seeded granules formation. On the other hand, larger impeller speeds could promote the seeded granular formation [37]. However, dissipation of energy could be enhanced at a very high impeller speed, and therefore effective speed, which produce maximum fraction of seeded EP granules must be explored in the current study. Around 200 particles of the final products in a size range between 850-1180 μm (granulation time of 60 s) were analysed by optical microscopy for more clarification about the effect of impeller speed on EPG modification. The amount of BP granulates, EP granulates and EP granules covered by BP (referred as seeded EP granules) were determined in the final seeded granules (Fig. 6). Fig. 7 illustrates the related results based on a Pie chart format after the evaluation of optical microscope images. According to Fig. 7, impeller speed of 500 rpm produced a larger amount of seeded EP granules as compared to 300 rpm and 700 rpm. At a low speed of 300 rpm, impeller's energy would not be sufficient to disperse the binder over the surfaces of EP granules to produce seeded EP granules. On the other hand, generation of BP granulates at 700 rpm could decrease the amount of produced seeded EP granules due to the high shear rate and energy dissipation.

The results of this investigation demonstrated the significance of performing process optimisation for increasing the number of seeded granules. In the current investigation, the produced granulated product containing around 70% of seeded EP granules (60 s and 500 rpm), was used for the homogeneity analysis of the EPG as it contained the most seeded EP granules. Overall, by considering the proper size distribution for powders, the optimum condition for the seeded granulation can be obtained by employing a design of experiment approach and testing different ranges of operating conditions. As predicting the effect of operating conditions for different powder systems is difficult, design of experiment approach could be used to optimize the system and understand the effect of different parameters on response (here the number of seeded granules) by utilizing the minimum number of test runs. Other parameters such as mixing ratio of powders, amount of binder can also be elucidated using this approach. A seeded granulation regime map can also be prepared to specify optimum operating conditions under which seeded structure is formed (further detail is provided elsewhere [28]).

## 3.2. Homogeneity of the ternary mixture containing EPG (before and after shape modification)

Powder heaps containing 40 g of ternary mixture and with the weight percentage ratio of 92.6 (BP)/5.55 (TAED)/1.85 (EPG) were prepared to analyse the effect of shape modification on the segregation reduction of detergent powders (Fig. 8(a) and (b)). One of the heaps was formed using the original rounded EPG (0.74 g, with the mode size of particle size distribution) and the other by applying the seeded EP granules (2 g which is approximately equivalent to the same number of rounded EP particles used in the ternary mixture). The total weight difference between the two types of enzyme granules is due to the changed bulk densities and the additional weight of binder and BP for the case of seeded EP granules. It should be noted that the granulated BP and granulated EP were separated from the seeded EP granules by gentle vibration in a vibratory system and then the remaining seeded EP granules were used as modified EPG for weighing. The Segregation Index (SI, i.e. CoV of enzyme particles) was estimated by image processing of the taken image from the front face of the heaps. The SI was decreased from 1.25 to 0.68 after the modification of the particle's shape structure. Due to the presence of the red pigment on the surfaces of EPG, the fine white BP particles went pink in colour during the granulation process, enabling their differentiation from the rest of materials and therefore the SI determination using image processing. The conclusions drawn in this study have been further verified using NIR spectroscopy as well as DEM simulation as described in the section 3.3 [8]. The homogeneity of EPG was analysed for some of the extracted powder samples (Grid number: 2, 3, 4, 5, 6, 8, 9, 10 and 11) by means of NIR spectroscopy, showing that the SI decreased from 0.50 to 0.15 after shape modification. The magnified segregation extent of EPG measured using surface analysis (2D) versus sample extraction strategy (3D) is probably because of the wall effects on packing of powder mixtures inside the heap [33]; this will be fully evaluated in the DEM analysis as described in the section 3.3.

Fig. 9 illustrates the cross-sectional of EPG obtained by X-ray microtomography, (XRT-NanotomX-ray instrument of GE Phoenix-Germany), before and after shape modification. 3D visualization of particles was produced by the reconstruction of XRT images using VGStudio software and the post-processing analysis by Avizo Fire. The rougher surfaces for the modified EPG could be observed in the cross-sectional XRT images after granulation process which is different from the original round particles (Fig. 9). Therefore, the physical interlocking phenomena between particles could be promoted when the modified EPG was used in the ternary mixture, thereby mitigating the free movement of dense EPG in the mixture and their accumulation into the centre of the piles. To gain a better understanding of the trajectory of EPG (before and after the shape modification) in the generated ternary pile, the analysis using a DEM simulation has been carried out. Therefore, the movement patterns of the original and seeded EP granules in the studied ternary mixture were further compared computationally.

## 3.3. Computational methodology and validation of the experimental results using DEM

Heap formation of the ternary mixture of laundry detergent powders containing BP, TAED and EPG has been successfully simulated previously in the work of Alizadeh et al. [33]. The same calibration methodology was implemented in the current study to simulate the segregation of the ternary powder mixture containing BP, TAED, and EPG (before and after shape modification using seeded granulation technique). EDEM2018 software was applied to simulate the heap formation process. The powder mixture used in the current study is not cohesive and the particles are relatively large; therefore, the adhesive forces are negligible compared to their weights (low granular Bond number and low Cohesion number) [38, 39]. On this basis, only the Hertz-Mindlin (no slip) [40-42] contact model was applied. Here, the no-slip condition means that two contacting particles will slip on each other only if their relative tangential force exceeds their static friction force. In such a condition, they are prone to both rolling and sliding. The details of these models are available elsewhere [43-45].

In order to mimic the structure of seeded EP granules in DEM, one particle was selected randomly from the seeded granules as presented in Fig. 9. Using the Avizo Fire software, the mesh files of the real shapes have been generated as .stl file by which the clumped spheres were modelled as presented in Fig. 10.

After characterising the properties of seeded EP granules, the heap formation process was simulated by DEM modelling using the normal spherical EPG (case 1) and seeded EP granules (case 2). The properties of BP, TAED and raw EPG are similar to those used in our previous work [33], and are given in Table 1. To simplify the DEM analysis, it is assumed that the outer layer of the granules with the modified shape (case 2) are covered with the BP particles; therefore, similar surface properties as of the BP particles are considered for the seeded EP granules (Table 1). It should be noted that the seeded EP granules have a dense core of EPG and lighter shell of the BP particles, giving a non-uniform structure of the agglomerates. The equivalent density of the seeded EP granules was measured by dividing the total mass of an agglomerate by its total volume, knowing the densities and volume fractions of the BP and EPG in an agglomerate, which has been already modelled as clumped spheres. The specifications of the modelling and the physical and mechanical properties of the particles used in simulations for the case 2 are detailed in Table 1.

In the next stage, the same experimental process of heap formation has been mimicked computationally using DEM (based on the material properties presented in Table 1) and then the image from the heap surface was divided into 2 rows and 5 columns giving 10 bins in total (Fig. 11(a)), to obtain the concentration map of EPG using image processing software. Similar to the experimental analysis, CoV concept has been used as SI to evaluate the segregation tendency of the species. The bin size was selected according to the experimental procedure so that the numerical results could be compared with those of the experiments. It can be observed from Fig. 11(a) that when the spherical EPG are used (case 1) for the process of heap formation of the ternary mixture, the granules are inclined to segregate into the centre of the pile. On the other hand, case 2 shows that seeded EP granules could spread all over the heap which is in a good agreement with the experimental results. The SI values of the fraction distribution of EPG (before and after shape modification) for the front face of the heap were then calculated and the results are shown in Fig. 11(b). A good agreement between the experimental and DEM simulation results is obtained as illustrated in Fig. 11(b). It is observed that replacing the round EPG (case 1) with seeded EP granules (case 2) could halve the segregation extent (50% reduction in CoV) in the studied model formulation.

To have a more in-depth analysis of the mixture quality, the mass concentration of the EPG (before and after shape modification) was calculated for the DEM results to obtain the related SI for the entire heap (3D analysis). The results were then compared with those obtained from the NIR spectroscopy presented earlier, (Fig. 11(c)). It should be noted that conversely to the NIR technique in which a very thin layer of the particles surface was scanned and analysed, the SI was obtained based on the mass fractions of the particles in the DEM modelling. Although the methods of analysing the segregation extent are not the same for the two cases, the change in the segregation tendency of the EPG before and after altering their shapes could be compared for the experimental and DEM modelling cases. The results obtained from the experimental and numerical approaches are presented and compared in Fig. 11(c), both of which show significant decrease in the segregation tendency of the seeded EP granules compared to the round EPG. It is observed that the value of SI obtained from the NIR technique (experiment) decreased by 70% after using the seeded EP granules. However, the SI value reduced by 40% when DEM modelling was used. Although the SI values in the two described methods do not show similar figures, they corroborate each other by showing similar decreasing trends.

In the next series of the analysis, the heap width was divided into 5 layers across which the particle distribution and SI were calculated and compared (Fig. 12(a)). It can be observed in Fig. 12(a) that spherical EPG particles have poor distribution at the front and back sides of the heap (case 1). However, the distribution patterns become more uniform for the case of seeded EP granules (case 2) and, therefore, a better homogeneity of enzyme particles through the depth of the heap could be obtained. Simulation outputs in Fig. 12(b) additionally confirmed the experimental observation, where applying the seeded EP granules resulted in less variation in SI values and considerable reduction of the average value of the SI for the entire heap. It can be seen from Fig. 12(b) that the SI index values for the middle layers (layers 2, 3, and 4) are lower for both cases, showing that the segregation extent is exaggerated on the visible side walls of heaps.

## 3.4. Segregation mechanisms before and after shape modification

The results of Fig. 8 and Fig. 11 show that when the spherical EPG are used for the process of heap formation of the ternary mixture, the granules tend to segregate more into the centre of the pile, which is due to the higher density of EPG as compared to other ingredients (push-away mechanism). Full investigation of the segregation mechanisms of the studied ternary mixture are described in our previous researches [8, 9, 33]. Several snapshots of the impact of the modified EP granules to the surface of BP particles at different time frames were obtained using a high-speed camera, as demonstrated in Fig. 13. Clearly, surface modification of the EP particles changed their restitution properties which results in a no/very little bounce for the seeded granules after the impact onto the bed of BP particles. This is different than the original rounded EPG, which obviously was bounced after impacting the bed of BP particles.

Movement pattern of some selected EP granules obtained from DEM analysis are presented in Fig. 14. As presented in the Fig. 14, the spherical EPG segregate to the left hand side of the heap due to a lack of interlocking with other particles; whereas, the seeded EP granules (case 2) distribute over the heap surface more evenly. In addition, round EPG (case 1) pass the top moving layer of the heap more easily than their agglomerated counterparts and get buried in the stagnant part of the mixture more quickly. On the other hand, the seeded EP granules stay in the moving layer for a longer time and, due to their larger and irregular shapes, cannot penetrate the bed as easily. As a result, the seeded EP granules travel further down the heap and reach the corners as well. The highly irregular shape of the seeded EP granules leads to the considerable level of interlocking with BP and TAED particles which helps the agglomerates to maintain their initial homogeneity for a longer time.

The balance between size and density of particles is also an important factor for the mitigation of segregation when the modified enzyme granules are used. As reported by Arntz et al. [46], if a parameter like density is driving particles to segregate, increasing their size can neutralise the adverse effect of density, and vice versa. In the present case, where the EPG have a density nearly three times higher than other species, formation of the seeded EP granules has reduced the segregation via increasing the granules size as well as reducing their effective density in the system. Obviously, if the proportion of BP to EPG granules increases in agglomerates, too large lumps with low packing density will form. These lumps will segregate to the heap corners. Therefore, a careful balance between the size and density of agglomerates should be considered. The same concept is also studied and discussed at different cases by Asachi et al. [9].

## 4. Conclusions

In this study, a new method based on shape modification of dense particles by surface coverage of other fine ingredient has been investigated to enhance the homogeneity of granular materials which could have a great potential for powder process applications. For this purpose, a simple methodology based on the "seeded granulation process" was applied and optimised for two parameters of the mixing rate and time in such a way that a high amount of modified EPG (known in this study as seeded EP granules) was yielded. In particular, the segregation of original enzyme particles (EPG) in a model powder product system containing the BP, TAED and enzyme granules (representing a typical laundry detergent powder mixture) has been evaluated and compared to the case where modified enzyme granules have been utilized in the mixture. More specific points can be drawn as follows:

- The significance of performing the granulation process optimisation for increasing the number of seeded EP granules was demonstrated experimentally. Around 70% seeded EP granules was produced after 60 s and impeller speed of 500 rpm.
- The effect of particle shape modification on the segregation of EPG was first assessed experimentally using 2D analysis, where it was found that the SI decreased after particle shape modification. This has been further verified using experimental analysis of entire heap using NIR spectroscopy as well as DEM simulation.
- The same experimental process of heap formation has been mimicked computationally using DEM and it was shown that when the spherical EPG was used in the process, the granules were inclined to segregate into the centre of the pile. On the other hand, seeded EP granules could spread all over the heap, resulting in a reduced segregation.
- Using DEM approach, the mass concentration of the EPG (before and after shape modification) was measured and the related SI for the entire heap was estimated (3D analysis). The results were then compared with those experimentally obtained from the NIR spectroscopy. A significant decrease in the segregation tendency of the seeded EP granules compared to the round EPG has been shown by both techniques.
- Based on both experimental and DEM analyses, the main reason for the reduction of segregation of modified granules in the ternary mixture could be attributed to the changed surface properties of particles and therefore the enhanced interlocking effect which could change the movement patterns of granules during the heap formation.

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# Figures

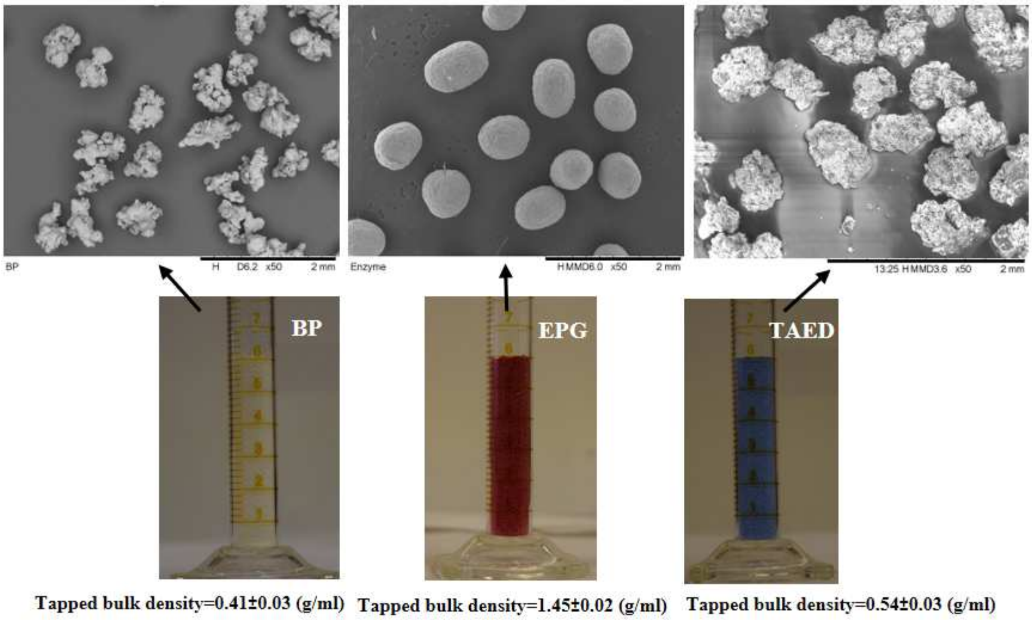


**Fig. 1.** SEM images of a number of BP, EPG and TAED particles along with the measured bulk tapped densities.

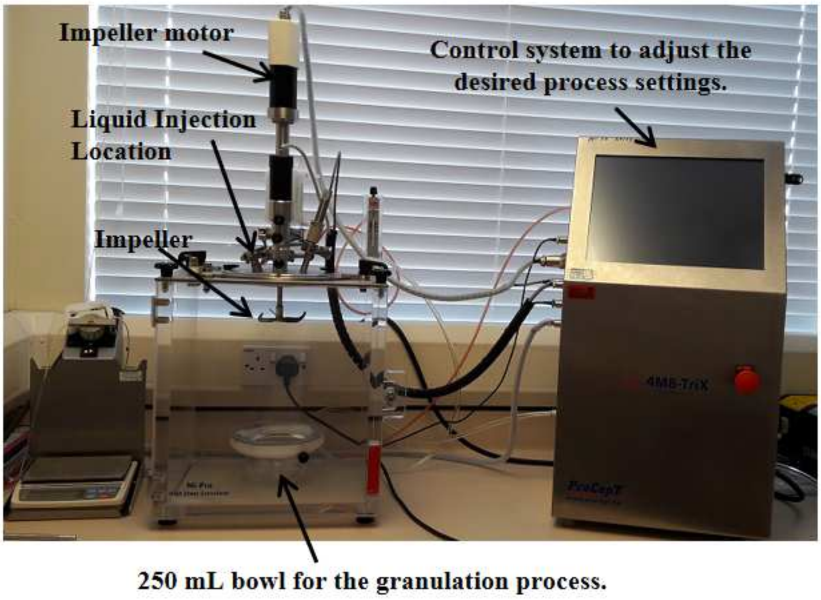
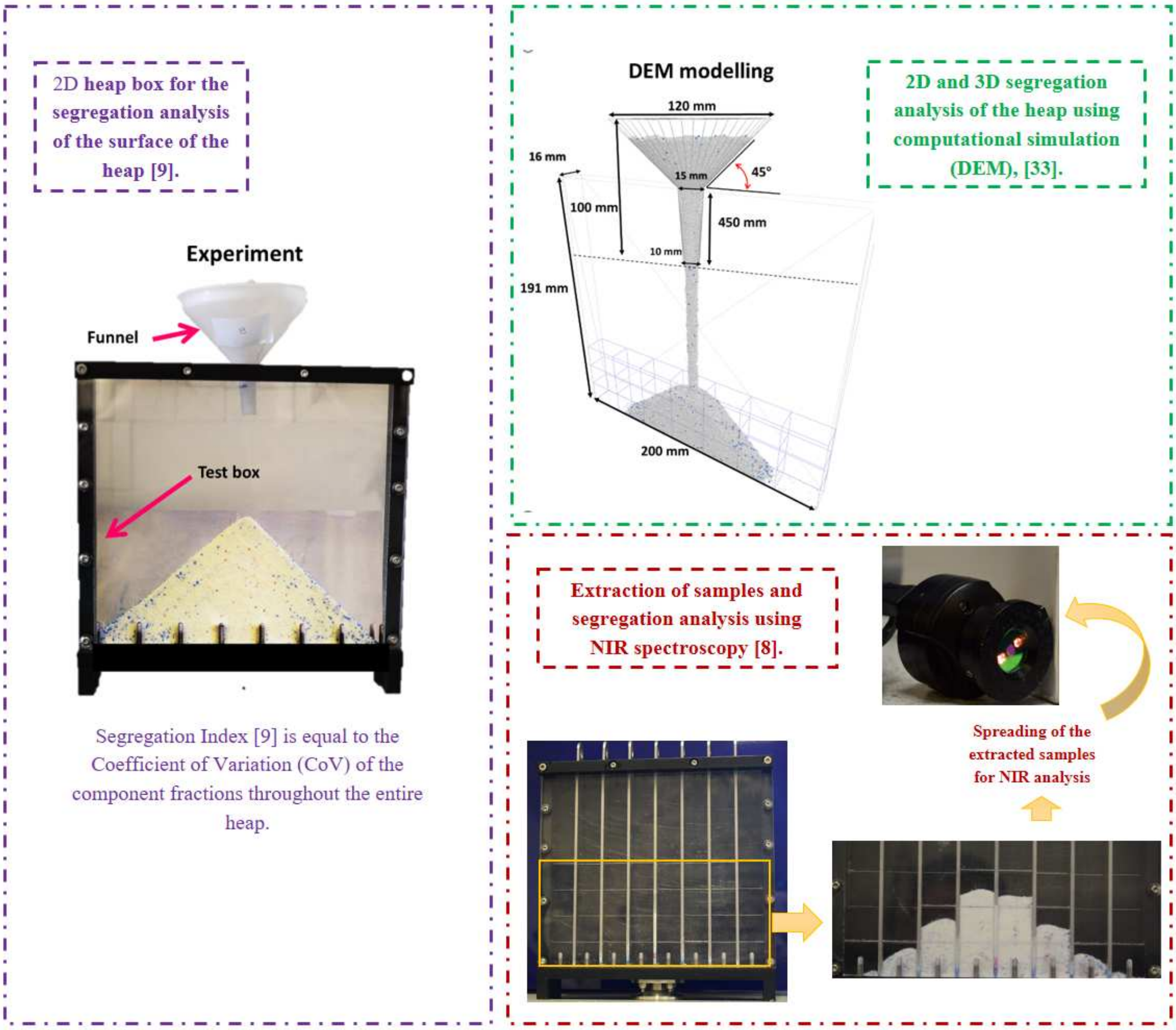


**Fig. 2.** High shear wet granulator system for the granulation of particles.



Segregation Index [9] is equal to the Coefficient of Variation (CoV) of the component fractions throughout the entire heap.

**Fig. 3.** Different techniques used for the 2D and 3D segregation evaluation of particles in a heap of laundry detergent powder mixture.

D>1180 $\mu m$ 850<D<1180 $\mu m$ 600<D<850 $\mu m$ D<600 $\mu m$

**Fig. 4.** The image of the materials extracted from different sieve cut sizes of a generated granulated product.

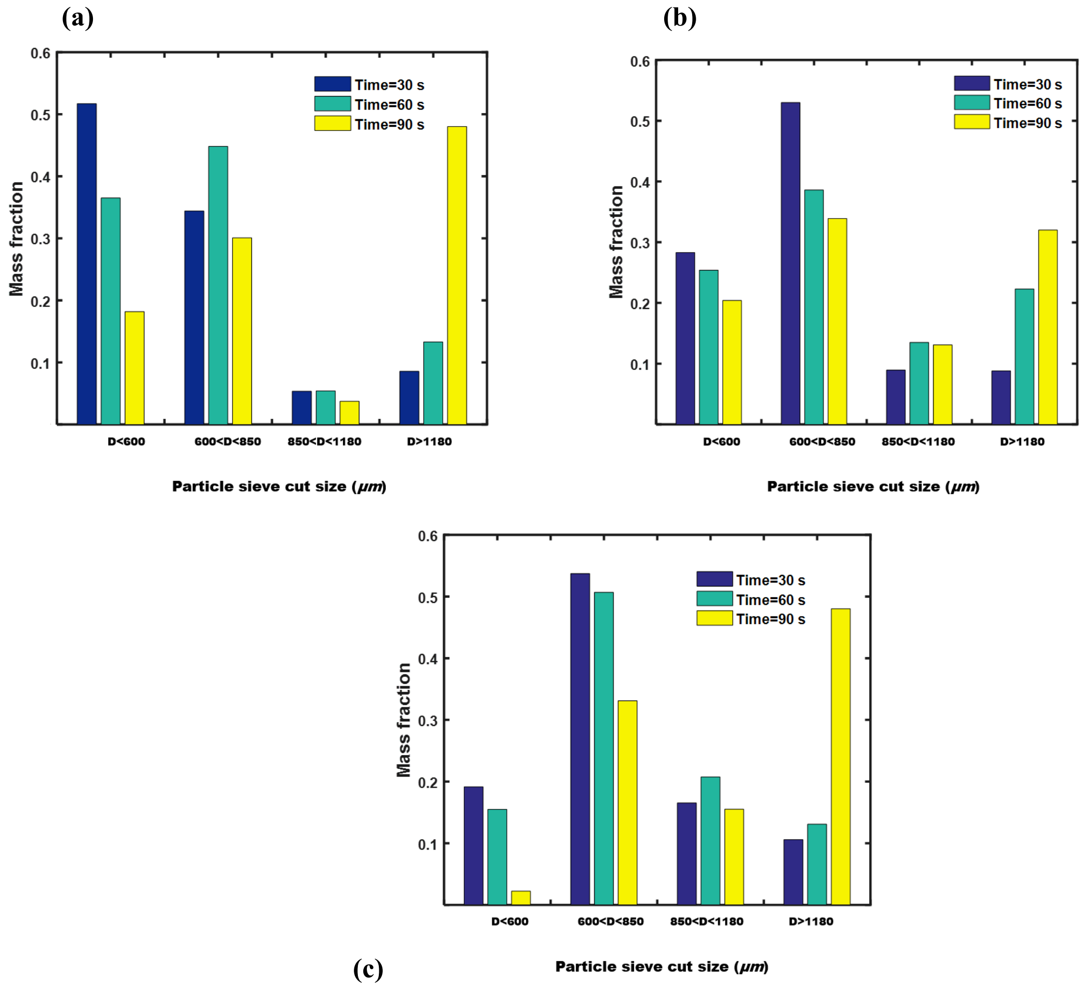


**Fig. 5.** The effect of impeller speed and granulation time on the particle size distribution of granular products: (a) 300, (b) 500 and (c) 700 rpm.

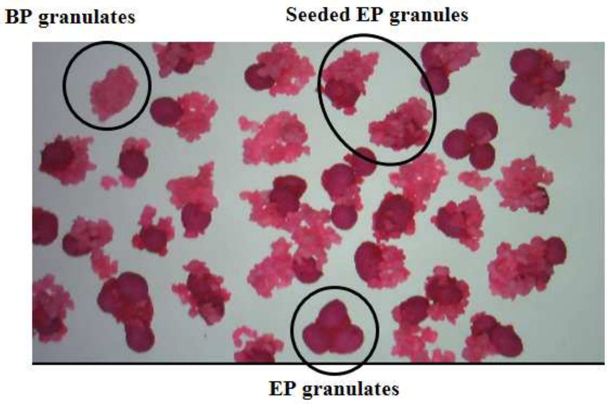
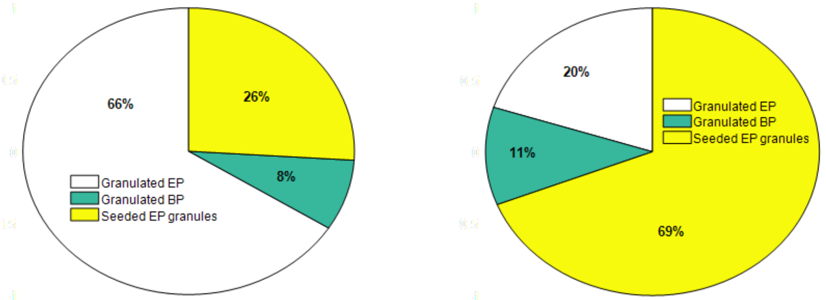
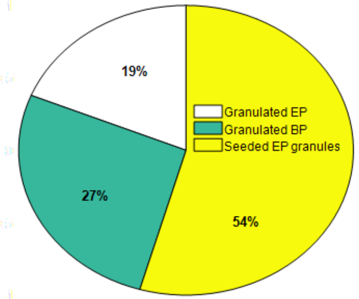


**Fig. 6.** Different types of granulates in the optimum granulated size range.



**Impeller speed=300 *rpm*** **Impeller speed=500 *rpm***



**Impeller speed=700 *rpm***

**Fig. 7.** Pie charts of the scanned granulates in a size range between 850<*D*<1180 µm, obtained from granulator at the time of 60 s and the impeller speeds of 300, 500 and 700 rpm).

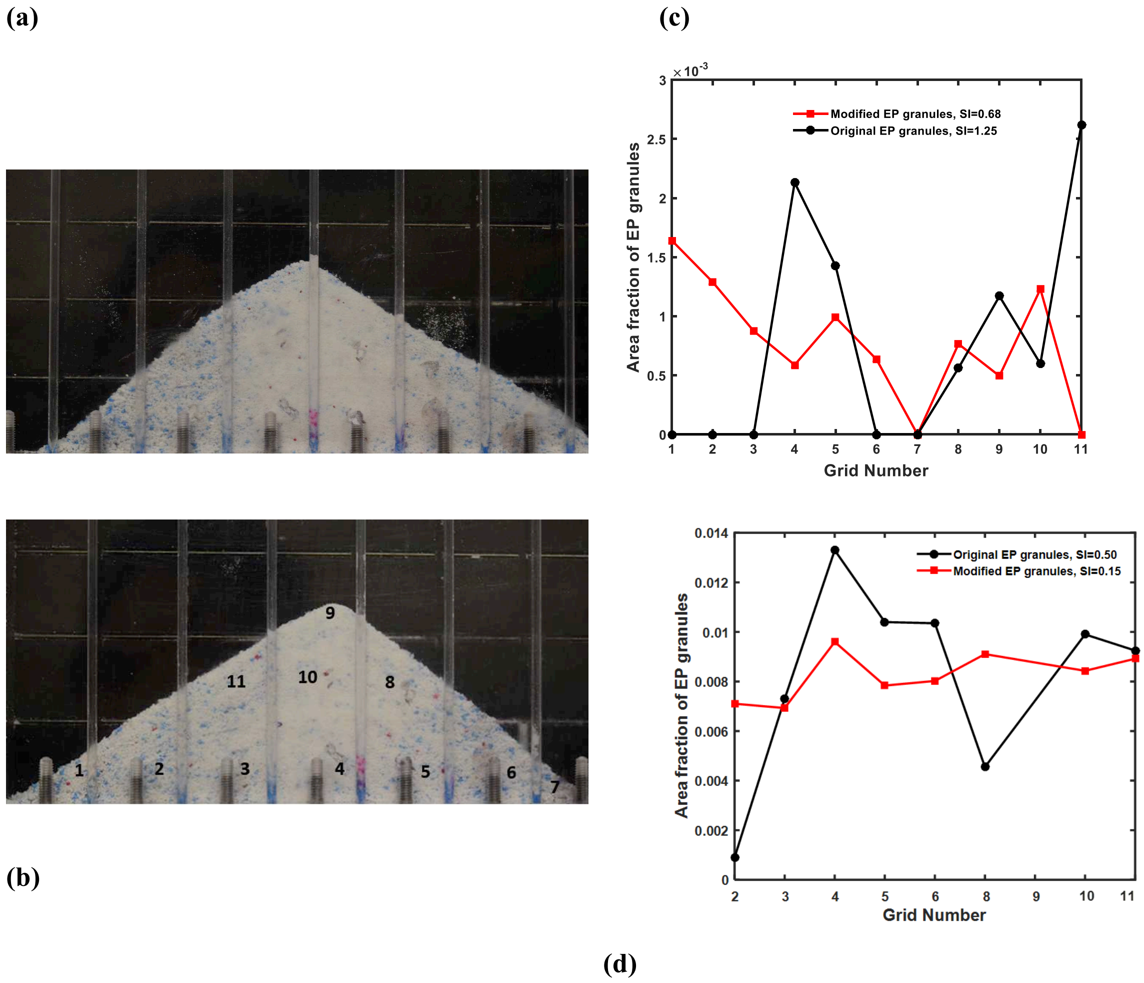
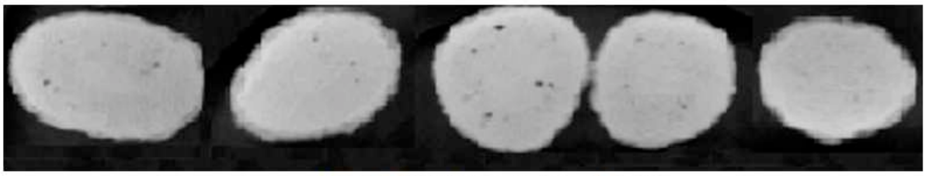
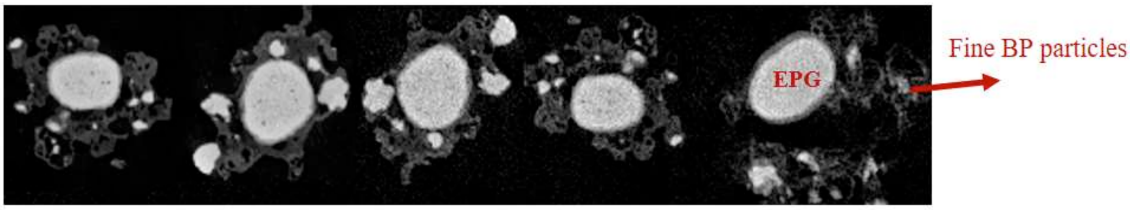


**Fig. 8.** Heap formation using (a) original EPG and (b) modified EP granules, concentration map and the SI of EPG in the ternary mixture before and after particle shape modification (c) obtained from image processing of the heap surface and (d) spectra analysis of the extracted samples.

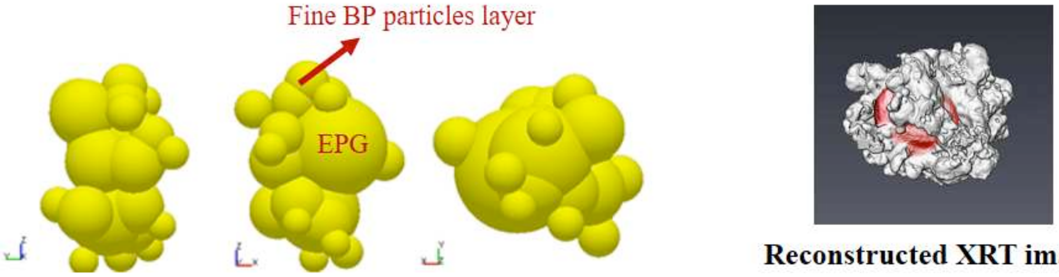


Round EPG (resolution=2 $\mu m$)



Seeded EP granules (resolution=4 $\mu m$)

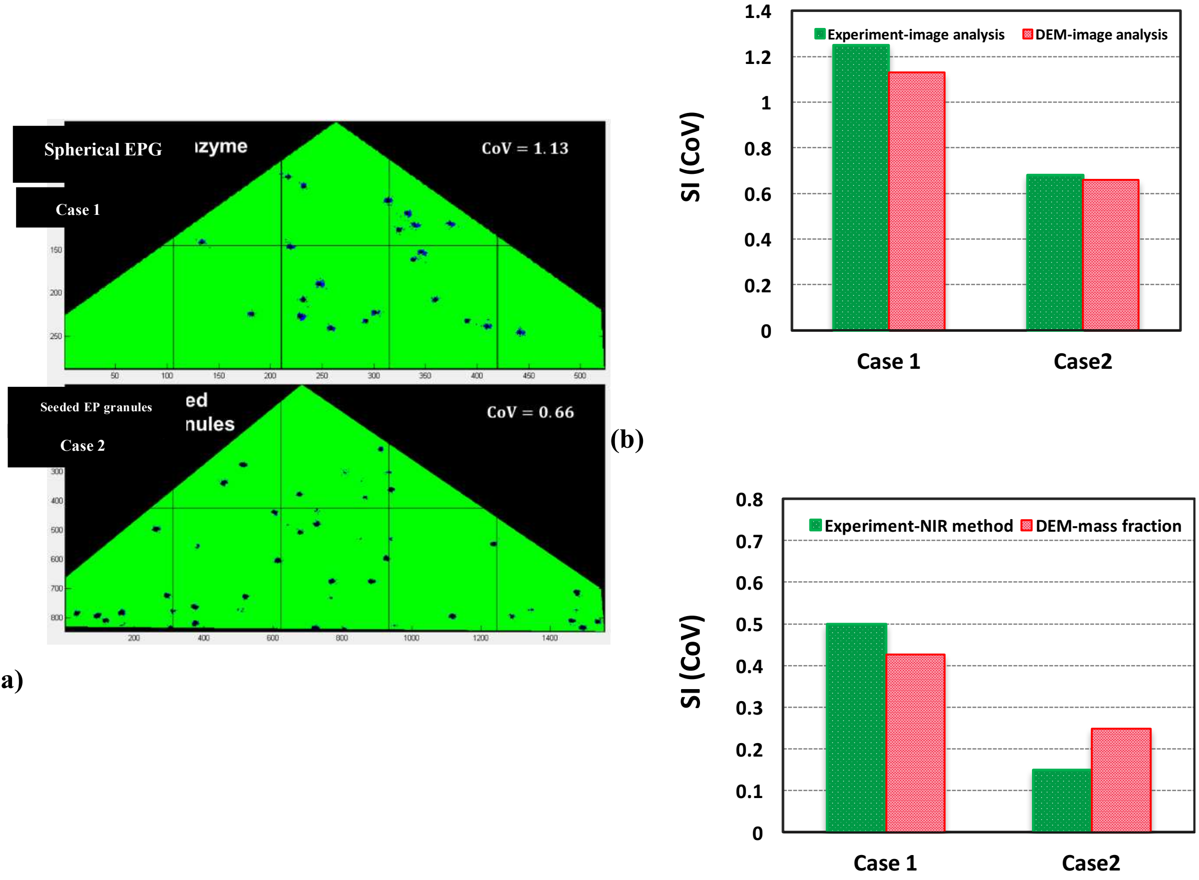
**Fig. 9.** Cross sectional view of EPG before and after structural shape modification obtained by XRT analysis.



Reconstructed XRT image

**Clumped sphere image used in DEM**

**Fig. 10.** Clumped sphere images of one of the seeded EP granules used in the DEM simulations (images presented from different angles).



(a)

(b)

(c)

**Fig. 11.** (a) The front image taken from the simulated heaps for the simulation analysis of cases 1 and case 2. The green colour shows the BP and TAED particles and the black spot indicates the EPG before and after shape modification, comparison of the SI of the spherical EPG (case 1) and seeded EP granules (case 2) obtained from the experimental and DEM data analysis based on (b) surface image processing (2D) and (c) depth analysis (3D).

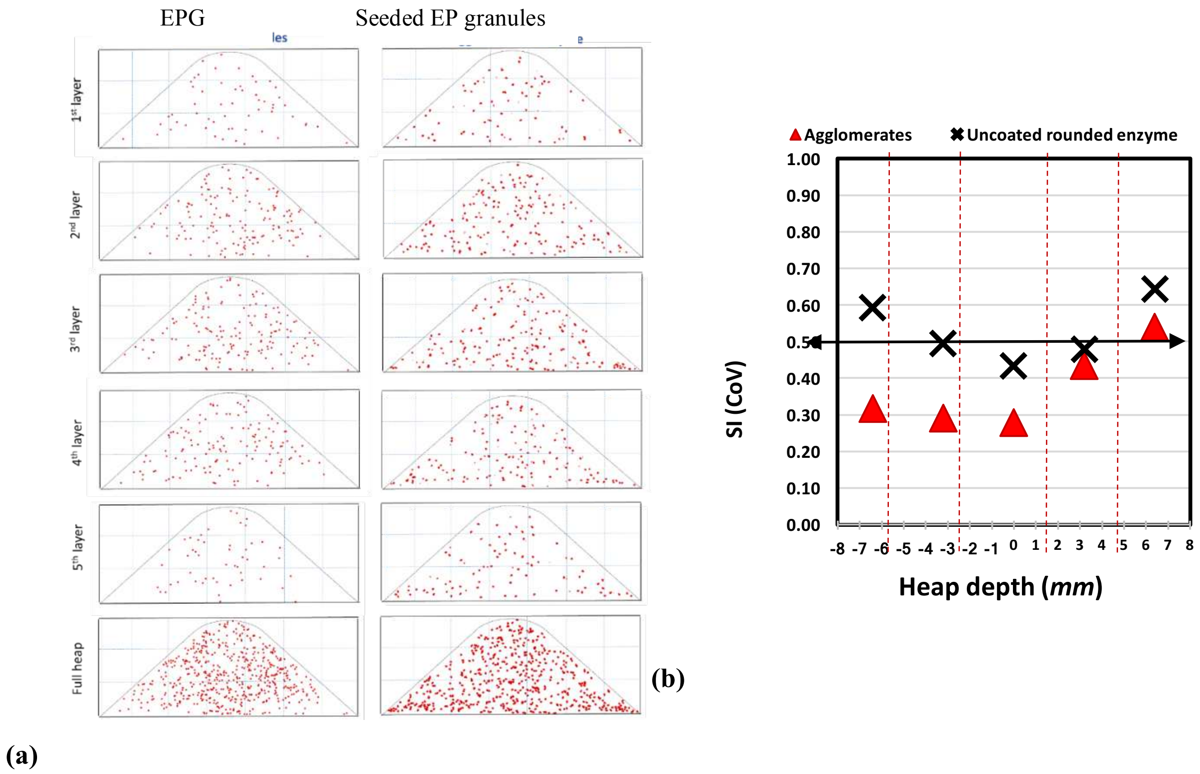


**Fig. 12.** (a) Comparison between the distribution patterns of the EPG at different layers of the heap and (b) the measured SI of the EPG (before and after shape modification) at different depth layers obtained from the particle mass fraction results.

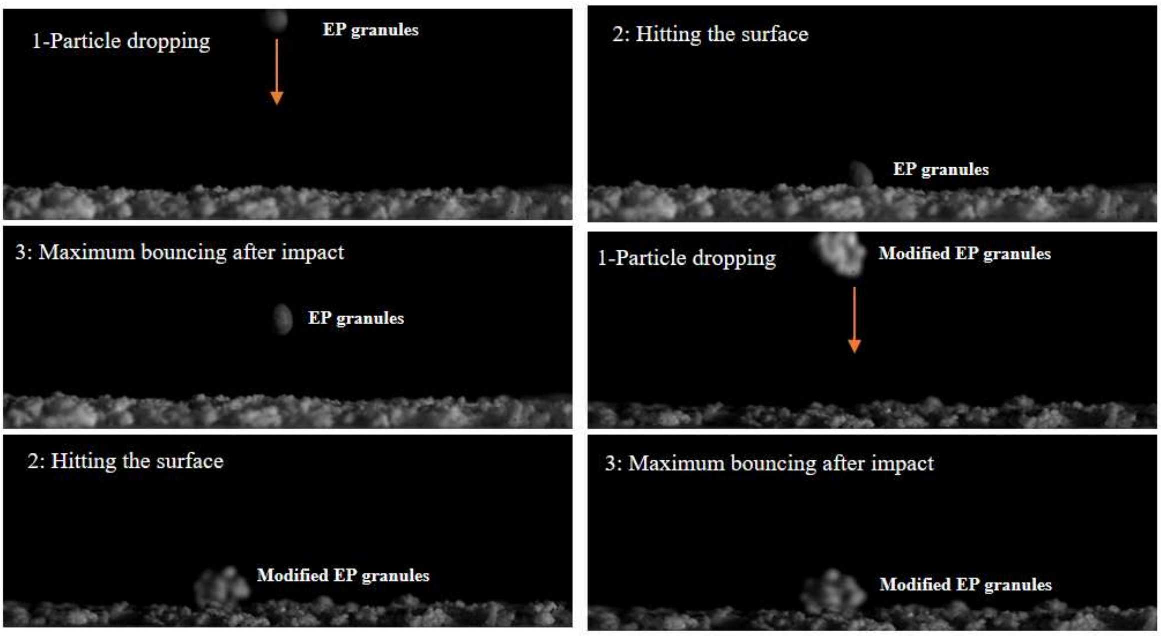


**Fig. 13.** Impact properties of enzyme granules to the bed of BP particles.

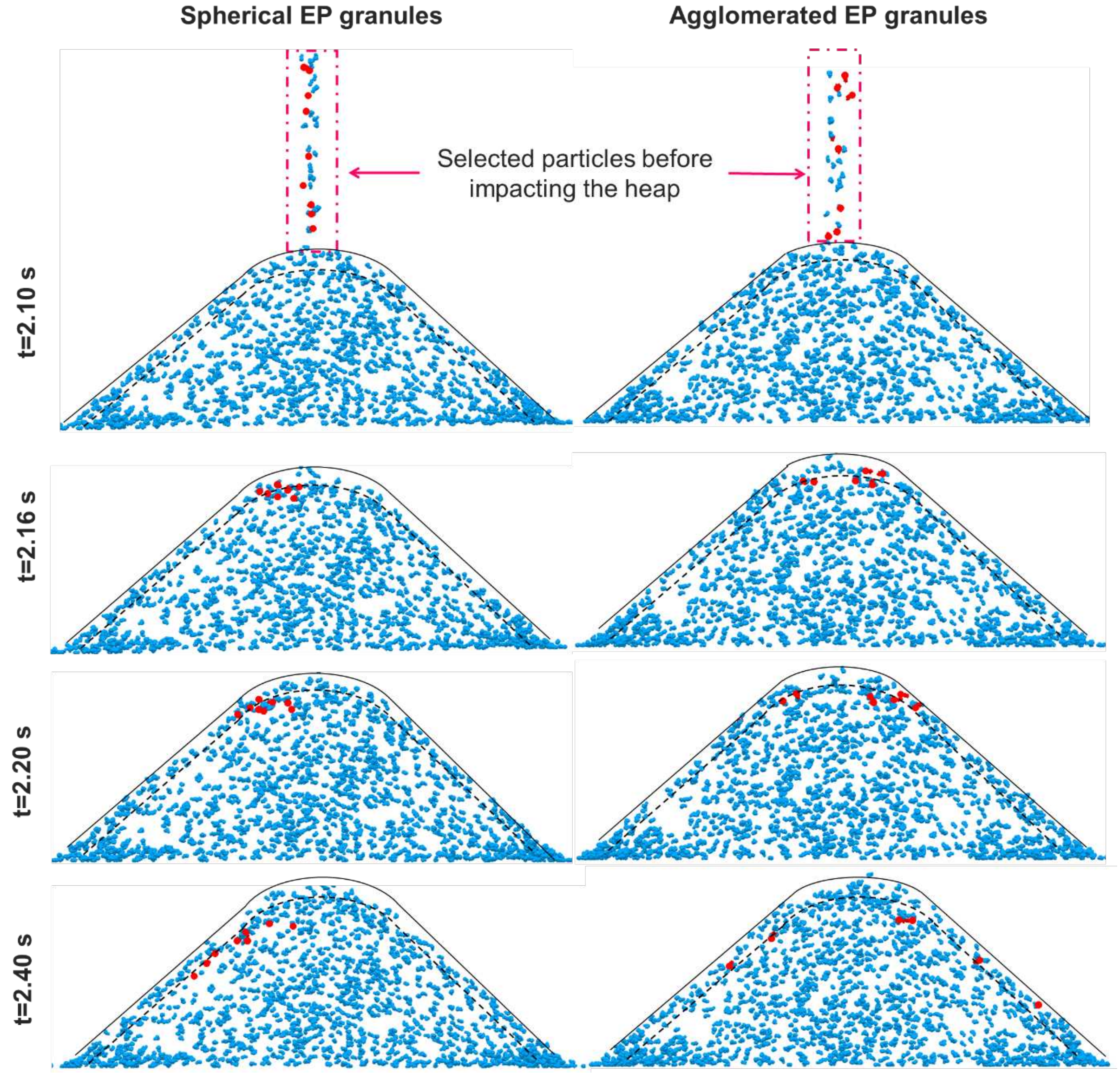


**Fig. 14.** The movement pattern of selected enzyme granules (red particles) before and after impacting the heap surface.

# Table:

**Table 1.** Specifications of the modelling and the material properties for simulations with spherical EPG (case 1) and seeded EP granules (case 2).

| Material type | BP | TAED | Case 1 | Case 2 | Perspex |
|---|---|---|---|---|---|
| | | | EPG | Seeded EPG | |
| Size (μm) | 425–500 | 850–1000 | 600–700 | 850–1000 | |
| Particles number | 205774 | 1517 | 554 | 537 | |
| Total mass (g) | 37.03 | 2.22 | 0.74 | 0.95 | |
| Weight Percentage | 92.59 | 5.56 | 1.85 | 2.4 | |
| Particle shape | 5-sphere | 5-sphere | 1-sphere | 21-sphere | |
| Shear modulus (MPa) | 10 | 10 | 10 | 10 | 1000 |
| Envelope density ($kg/m^3$) | 780 | 850 | 2320 | 1704 | 1180 |
| Coefficient of rolling friction | 0.10 | 0.01 | 0.05 | 0.10 | 0.01 |
| Poisson's ratio | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 |
| CoF (BP particle) | 0.62 | 0.69 | 0.70 | 0.62 | 0.42 |
| CoF (TAED particle) | 0.69 | 0.75 | 0.75 | 0.69 | 0.36 |
| CoF (Placebo particle) | 0.70 | 0.75 | 0.75 | 0.70 | 0.75 (EPG),<br>0.42 (Seeded EPG) |
| CoR (BP particle) | 0.20 | 0.30 | 0.20 | 0.20 | 0.28 |
| CoR (TAED particle) | 0.30 | 0.32 | 0.20 | 0.30 | 0.32 |
| CoR (Placebo particle) | 0.20 | 0.20 | 0.10 | 0.20 | 0.20 (EPG),<br>0.28 (Seeded EPG) |