Experimental Studies on the Effects of Process Parameters on Granule Properties in a Conical High Shear Granulator

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ABSTRACT: *Experimental work was carried out to investigate the influence of impeller speed, granulation time, binder mass and their interactions on granule size distribution, mean size and binder content distribution in a conical high shear granulator. It was observed that the response of high shear granulation to changes in process parameters varies significantly from one operating condition to another. For all quantities of binder, the granule mean size increased with time at lower impeller speeds, while the opposite trend was observed at higher impeller velocities. For long mixing times, increasing impeller speed decreased the granule average size constantly; whereas for other durations, an increase in the mean size followed by a decrease was observed across ascending impeller speeds. It was also found out that variations of the span of granule size distribution can be utilized to indicate the dominant rate process. Coalescence which was prevailing at lower impeller speeds, was limited by attrition, breakage or shattering at higher impeller velocities. Binder-solid ratio of granules showed a continuous increase across size at lower impeller speeds and a maximum at middle size range for higher impeller velocities. Binder content distribution showed different trends with time at different operating conditions. Furthermore, increasing impeller speed limited the domain of binder content variations and made the granule binder-solid ratio to be much closer to theoretical value.*

KEY WORDS: *High shear granulation, Impeller speed, Binder distribution, Span.*

INTRODUCTION

High shear granulation converts fine powders to larger agglomerates by mixing powder with suitable binder at impeller speeds greater than ~100 rpm. Complex material flow patterns, wide range of shear rates inside the granulator and simultaneous influence of all granulation rate processes, make control of product attributes very difficult in this type of granulation [1]. Furthermore, formulation, process parameters and equipment design and scale affect final product quality. Therefore, considerable efforts have been made to investigate the effects of different parameters on mechanisms involved in high shear granulation and properties of produced granules. Some of these attempts have been focused on the influence of operating

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parameters, especially in industries where feed materials and granulator are not altered for long periods of time.

Among several operating parameters, some parameters seem to attract more attention in related researches. For instance, many authors have examined the influence of binder mass on granulation behavior. *Iveson et al.* [2] showed that maximum saturation of granules is one of the key factors determining the regime of granule growth. Others [3-5] observed that increasing binder to solid ratio results in greater extent of granulation, increased consolidation and growth rate and larger granules. The effect of granulation time has been also investigated. *Hoornaert et al.* [5] observed different behaviors of granulation with time according to binder-solid ratio of the system. They found out that at low binder-solid ratios, the size of granules does not change with time, while increasing binder-solid ratio induces granule growth and accelerates its rate. *Knight et al.* [6] and *Schaefer & Mathiesen* [7] reported that extended granulation increases granule size, makes size distribution narrower and reduces granule porosity.

The other noticeable parameter is the rotational speed of impeller. *Knight et al.* [8] produced high shear granules at three impeller velocities. They observed an initial increase in granule mean size, followed by a decrease across ascending impeller speeds. The decrease in granule size by increasing impeller speed was also reported by *Ramaker et al.* [9]. Thies and *Kleinbudde* [10] studied the effects of process variables on melt high shear granulation and noted that increasing impeller speed and binder concentration increase growth rate and granule size. *Mangwandi et al.* [11] showed that how the granulation system responds to changes in impeller speed depends on the viscosity of binder. For low viscosity binders, the granule size and strength decrease by increasing impeller speed, while for high viscosity binders the opposite trend is observed. *Oulahna et al.* [12] examined the effect of impeller velocity on granule strength, size and binder content. They found out that increasing impeller speed reduces granule porosity and friability, narrows granule size distribution and makes granule binder content to be much closer to theoretical average value. The increase of granule strength to a fixed value by increasing impeller speed was also reported by *Rahmanian et al.* [13]

In the most reported studies, the interactions of process parameters have not been considered. Therefore, different granulation behaviors have been reported for a similar change in a specific operating factor. Furthermore, the influence of process parameters on binder content distribution of granules has been noticed in a limited number of works. The present study intends to investigate the effects of granulation time, impeller speed, binder mass and their interactions on granule size distribution, mean size and binder content distribution. The paramount objective of this paper is to obtain fundamental understanding of dominant rate process from the bulk behavior of granules at different operating conditions.

EXPERIMENTAL SECTION

Materials

Calcium carbonate (supplied by Poudrsazan Co., Iran) was used as primary powder. The particle size distribution of calcium carbonate was measured by HELOS light scattering particle size analyzer (Sympatec, Germany) and the following data were obtained: d_{10} =1.8 µm, d_{50} =54.4 µm and d_{90} =100.0 µm. The skeletal density of calcium carbonate was 2700 kg/m^3. The solution of polyethylene glycol (PEG 4000, Merck) in deionized water (50% concentration by mass) was used as liquid binder. The density and viscosity of the liquid binder at room temperature were 1078 kg/m^3 and ~0.088 Pa.s, respectively.

Granulator

Experiments were carried out in a 1 L conical high shear granulator made from polyamide (Fig. 1), which was outfitted with an electrical motor of 0.5 hp. The granulator vessel was made by depleting the interior part of a full cylinder in the form of a truncated cone. The agitator included a central shaft and five sets of blades, made from stainless steel. Central shaft was supported by two sealed ball bearings located inside the granulator top lid and bottom plate. The blades angle with horizon was 50°. The distance of blades to wall, to top lid and to bottom of the granulator was minimized to prevent paste formation. The conical shape of the vessel and special design of impeller allowed the wet mass to circulate thoroughly inside the granulator.

Granule production

In each experiment, 300 g of the primary powder was premixed for 2 min at 250 rpm. Then the impeller

Table 1: Values of operating parameters.

Binder mass (g)	36	33	30	
Impeller speed (rpm)	200	300	400[a]	500[b]
Granulation time (min)	4	8	12	16

a) Runs for 16 min were not conducted.

b) Runs were performed only for 4 min.

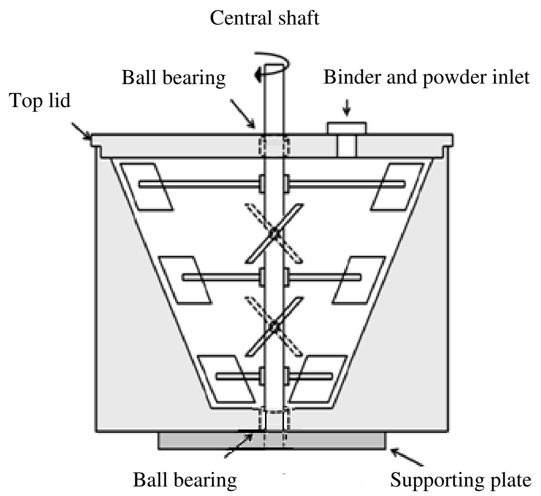


Fig. 1: Schematic of the conical high shear granulator.

was stopped and a desired quantity of the binder was poured on the powder. Granulation commenced as the impeller was started and was proceed up to definite times. The values of applied impeller speed, binder mass and granulation time are presented in Table 1. The experiments were designed in such manners that only one factor was changed in each run, called one at a time method. To avoid the unintended effect of sampling on granulation process, for each granulation time the experiment was conducted from the beginning without any stop and at the end of each run, total granules were analyzed. Some experiments at high impeller speeds and long mixing times were not carried out because of motor power limitation. To investigate the results reproducibility, most of the runs were performed twice and mean values were reported.

Produced granules were tray dried at ambient temperature for at least 24 h. Granule size distribution was measured by 16 ASTM-standard sieves in the range of 212 μm to 6.3 mm. Granules above 6.3 mm and clumps stuck to the granulator wall and to impeller's blades were considered as paste. A portion of each sieve cut was then analyzed for binder content using thermo-gravimetric method [6]. According to this method, granules were heated in a furnace at ~600°C for 2 h to remove their PEG.

RESULTS AND DISCUSSION

Effects of operating parameters on granule mean size

Fig. 2 shows the variations of granule mass mean size with time for three quantities of liquid binder and impeller speed. Mass mean size (d_m) of granules is calculated by Eq. (1):

$$d_m = \frac{\sum_{i=1}^{n} m_i d_i}{\sum_{i=1}^{n} m_i} \quad (1)$$

where m is the mass of granules retained on each sieve, d is the arithmetic average size of collecting sieve and the one above and n is the total number of sieves. According to Fig. 2, to increase binder mass by ~ 9% (i.e. the percentage of binder mass increase from 30 g to 33 g or form 33 g to 36 g) increases the mass mean size from at least 15% to a maximum of 44%. Increasing granule size by increasing binder-powder ratio is logically expected, because granulation is induced by binder.

Fig. 2 also represents that prolonged granulation increases granule size at lower impeller speeds (Fig. 2a), while the opposite trend is observed for higher impeller velocities (Figs. 2b and 2c), irrespective of the mass of binder. So it is concluded that the coalescence of granules which is dominant at lower impeller speeds is limited at higher velocities.

The influence of impeller speed on granule mean size is depicted in Fig. 3. From this figure, it is quite evident that except for 16 min, increasing impeller speed from 200 rpm to 300 rpm leads to an increase in the mean size, while further increasing to 400 rpm or 500 rpm reduces the average size. The effect of impeller velocity

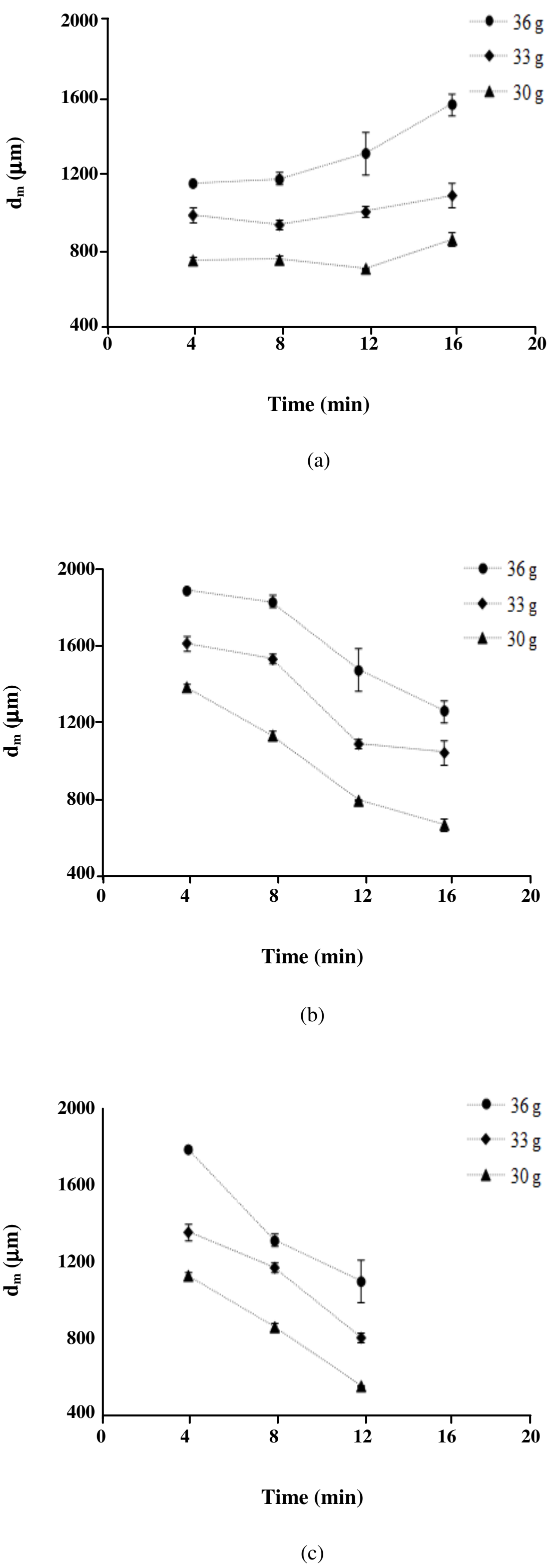


Fig. 2: The effect of granulation time on granule mass mean size at different quantities of binder for (a) 200 rpm, (b) 300 rpm and (c) 400 rpm.

on granule mean size depends on how granules respond to energy brought to the system by impeller. In this experimental work, increased input energy from 200 rpm to 300 rpm during 4-12 min, increases granule deformation, which in turn increases the probability of successful coalescence and size enlargement. Conversely, at higher input energy due to more granulation time (16 min) and/or higher impeller speed (400 rpm and 500 rpm), granules are not able to absorb the total impact energy via deformation. So, coalescence is limited and breakage (attrition) occurs and granule mean size decreases ultimately.

Statistical evaluation of the effects of operating parameters on granule mean size has been analyzed using response surface methodology with results published elsewhere [14].

Effects of operating parameters on the span of granule size distribution

The span of granule size distribution, which is a criterion for its broadness, is defined as Eq. (2):

$$\text{span} = \frac{d_{90} - d_{10}}{d_{50}} \qquad (2)$$

The variations of calculated spans (based on mass distribution) are shown in Fig. 4 as a function of granulation time, binder mass and impeller speed. Although a glimpse at Fig. 4 reveals that there is no specific relationship between span and the mentioned operating parameters, the results contain valuable information regarding rate processes and the trends are explainable by attention to the values of d_{10}, d_{50} and d_{90} presented in Table 2, where only the results of 200 rpm are shown for brevity.

At 200 rpm and for all quantities of binder (Fig. 4a), the span of Granule Size Distribution (GSD) decreases initially with time (from 4 min to 8 min) and then increases up to the end of the experiments. The span decreases initially due to the fact that at 200 rpm, low intensity of agitation inside the granulator leads to poor binder dispersion, broad nuclei size distribution and consequently broad granule size distribution at earlier times. By extending the granulation to 8 min, granule impacts continue, very loose and large granules break and their binder is re-dispersed in the granulator. The growth

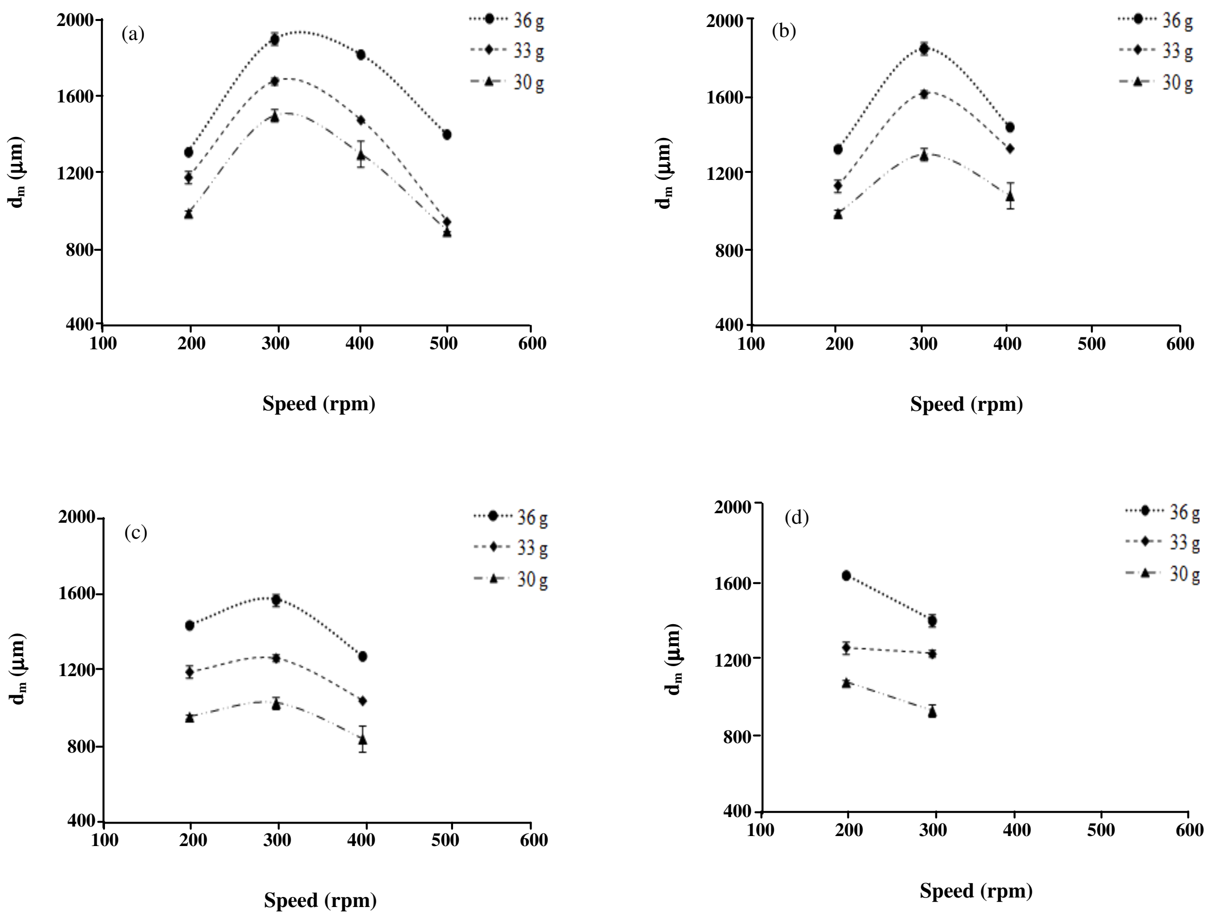
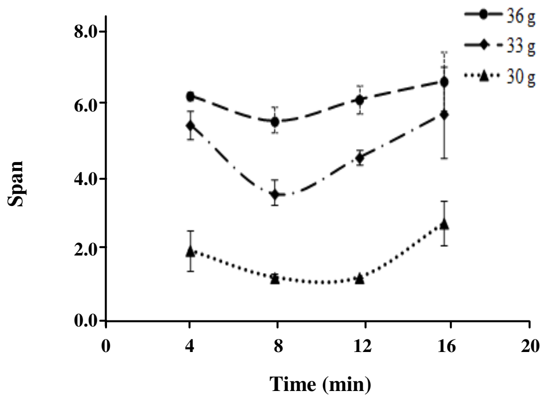


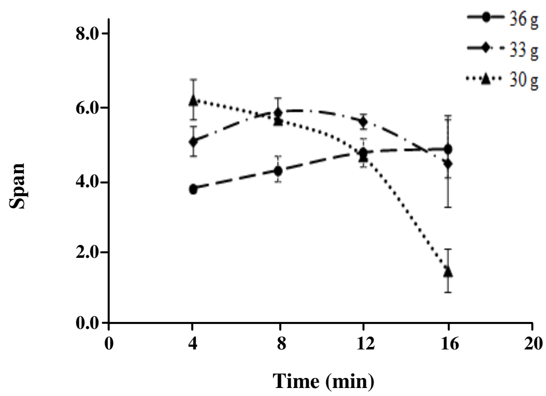
Fig. 3: The effect of impeller speed on granule mass mean size at different quantities of binder for (a) 4 min, (b) 8 min, (c) 12 min and (d) 16 min.

of very small and dry granules by re-dispersed binder as well as the breakage of very large granules makes the GSD narrower. The values of GSD characteristics in Table 2 confirm this elucidation. After this time, the breakage of large granules is limited by greater extent of consolidation and most of the granules will grow. Since the growth rate is directly proportional to granule binder content and the binder content is greater for larger granules at this impeller speed, the right-hand tail of GSD expands with time more rapidly to involve larger sizes. Hence the span of GSD increases with time. This explanation is verified by considering the variations of GSD characteristics. Table 2 shows that from 8 min to 16 min the corresponding values of d_{90} and d_{50} increase with time, whilst the values of d_{10} show small variations. In other words at 200 rpm, coalescence - which is the prevailing rate process - is in favor of larger granules.

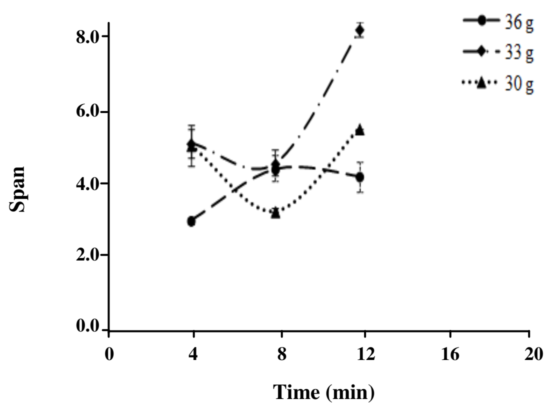
At 300 rpm (Fig. 4b) different trends of span variation are observed for different binder masses. Fig. 2b already presented the reduction of granule mean size with time at 300 rpm, and now calculations show that at this impeller speed and for all quantities of binder, the values of d_{10}, d_{50} and d_{90} decline with time (few exceptions exist for 16 min). These observations show that prolongation of granulation at 300 rpm shifts the whole granule size distribution to smaller sizes. This phenomenon can merely occur as a result of either attrition or breakage or a combination of them. The parameter that determines which rate process dominates the others is the binder content of granules. For 36 g of binder, granules are more deformable due to more binder content. So, the majority of granules inside the granulator can absorb increased impact energy at 300 rpm and resist breakage. In this case, the enhanced impact energy causes some



(a)



(b)



(c)

Fig. 4: The effect of granulation time on the span of granule size distribution at different quantities of binder for (a) 200 rpm, (b) 300 rpm and (c) 400 rpm.

extent of attrition in larger granules (with more impact energy). So, these granules loose very small parts without considerable change in their own size. Therefore, the amount of small granules increases, while the mass of large granules is not changed considerably. In other words, the left-hand tail of GSD shifts to smaller sizes more swiftly than the right-hand tail and the span of GSD increases with time. On the contrary for 30 g of binder, granules are less deformable due to less binder content. So, in this case the enhanced impact energy at 300 rpm causes considerable extent of breakage. Larger granules with more impact energy are more prone to break and convert to smaller granules. Hence, noticeable change in the mass of large granules occurs, whereas small granules experience little extent of breakage. Therefore, the right-hand tail of GSD shifts to smaller sizes more rapidly than the left-hand tail and the span of GSD decreases with time. For 33 g of binder, moderate binder content leads to moderate deformation of granules. Hence, a combination of mentioned variations is observed. For short granulation durations (4 min to 8 min) attrition is dominant and the span of GSD increases with time, while for longer times (8 min to 16 min), breakage overcomes because of prolongation of energy input, and the span decreases with time.

At 400 rpm (Fig. 4c) the variation of span for 36 g of binder shows good resemblance to that of 33 g of binder at 300 rpm. According to the mentioned argument, the prevailing rate process switches from attrition (for 4-8 min) to breakage (for 8-12 min). This observation is expected because of increased input energy at 400 rpm. By the same reasons presented for 300 rpm, it is anticipated that for 33 g and 30 g of binder, the span decreases constantly with time. But it is observed that in these cases, the span decreases initially and then increases considerably. The initial decrease occurs as a result of breakage, but calculations show for these binder contents, corresponding values of d_{10} decreases from 8 min to 12 min by one order of magnitude (e.g. from 212 μm to 75 μm for 33 g of binder). This behavior can be solely occurred by shattering of granules. Therefore, at 400 rpm and for longer than 8 min times, the influence of shattering on granule size reduction dominates other rate processes for medium to low binder contents. Further tracer studies can help verifying these inferences.

Table 2: Characteristic values of GSD at 200 rpm.

	36 g binder			33 g binder			30 g binder		
Time (min)	d_{10} (μm)	d_{50} (μm)	d_{90} (μm)	d_{10} (μm)	d_{50} (μm)	d_{90} (μm)	d_{10} (μm)	d_{50} (μm)	d_{90} (μm)
4	229	573	3779	218	528	3066	200	469	1125
8	234	589	3431	224	557	2193	214	503	841
12	229	619	4018	225	566	2773	221	532	843
16	228	676	4622	229	568	3481	219	534	1656

Effects of operating parameters on granule binder distribution

The amount of binder in each granule plays a major role on its growth rate. In other words, the binder to solid ratio of a granule is a key parameter indicating the probability of successful coalescence of the granule with other granules [15]. In present work, the binder distribution has been obtained as the ratio of polyethylene glycol (PEG) to solid (calcium carbonate) for different sizes of granules produced at several operating conditions. This ratio is defined as Eq. (3):

$$\frac{\text{PEG}}{\text{Solid}} = \frac{m_b}{2m_s} \qquad (3)$$

Where m is the mass and subscripts b and s denote liquid binder and solid, respectively. The coefficient of 1/2 involves the concentration of PEG in binder solution (50% w/w). Fig. 5 to Fig. 7 depicts the results at different impeller speeds.

At 200 rpm (Fig. 5) it can be seen that in spite of small fluctuations, the general trend is the increase of PEG-solid ratio with size for all quantities of added binder and granulation time. This trend has been already observed in high shear granulation [15, 16]. The increase of binder-solid ratio across ascending granule size is an intrinsic behavior of every poor mixing granulation. At 200 rpm, low mixing intensity results in poor wetting and broad nuclei size distribution which contains small dry to large over-wetted nuclei. According to previous sections, at this impeller speed coalescence is the prevailing rate process. It is well known that the rate of coalescence depends directly on binder content, i.e. the more binder content, the greater rate of coalescence. Therefore, the growth rate of larger granules is greater than that of smaller ones, so larger resultant granules come from nuclei with higher binder content. This observation is another proof that at 200 rpm coalescence is more probable for larger granules. Fig. 5 also depicts that at 200 rpm, the ratio of PEG-solid decreases from 4 min to 8 min, irrespective of the mass of added binder. However, this trend is less pronounced for other times.

At 300 rpm (Fig. 6) for all quantities of added binder, the trend of PEG-solid ratio shows an increase (up to ~1 mm) followed by a gradually decrease (to 5 mm). In this case, the successful coalescence of larger granules is limited by increased impact energy and the growth of small granules is constrained by low binder content. So, the greatest growth rate is allocated to medium granules. Therefore, the maximum PEG-solid ratio is observed for middle size range. At this impeller speed, the ratio of PEG-solid increases with time, especially for 16 min. This observation is in contradiction with that of 200 rpm and can be explained by the competition of rate processes inside the granulator. For medium to large granules (~1 mm to 5 mm) attrition increases the binder content, because small particles are removed from the granule surface by attrition and the outer part of granules is drier than the inner part [17]. Breakage does not affect the binder content of the mentioned range of granules, since the broken granules exit from their determined size range and the remained unbroken granules are not influenced by breakage. Finally, coalescence reduces the binder content of medium to large granules, because of descending trend of binder distribution across granule size in the specified size range. As a result, the binder-solid ratio of medium to large granules increases with time. The binder content of small granules (<1 mm) increases due to the breakage of larger granules. In this case, some extent of binder belonging to larger granules is re-dispersed by breakage. This binder can be accessible for small granules and increase their binder-solid ratio.

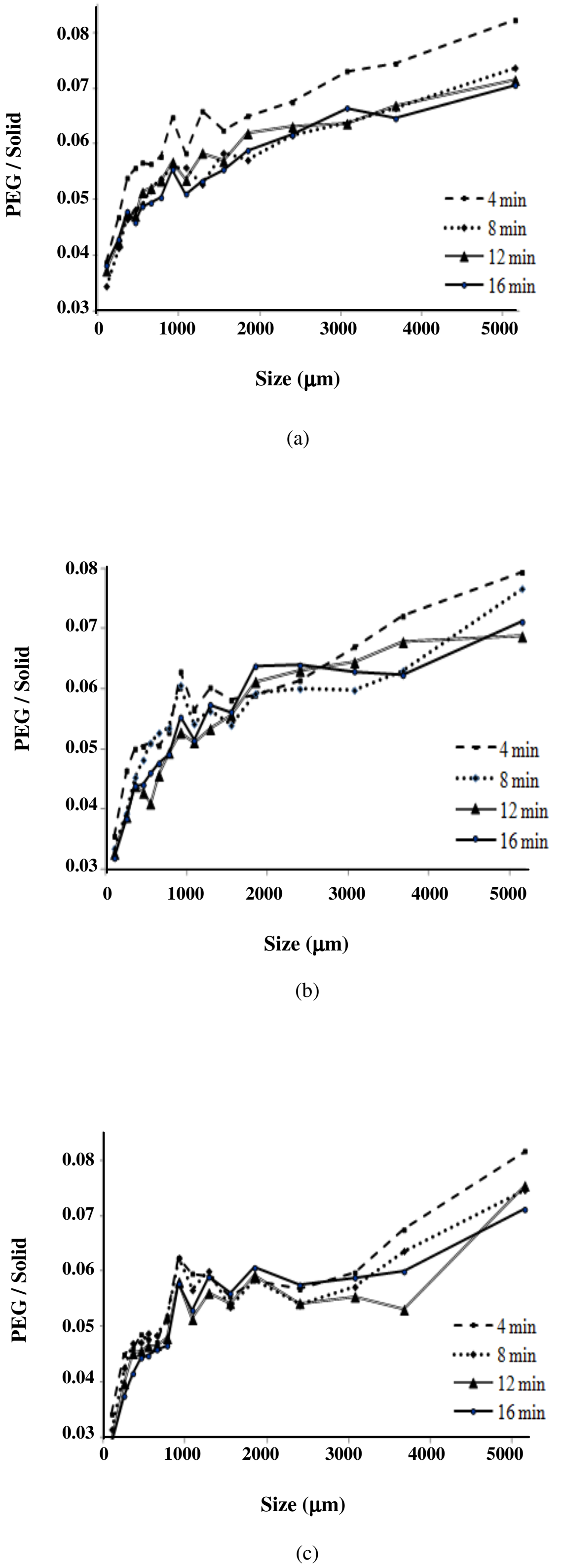


Fig. 5: Granule binder distribution at 200 rpm for (a) 36 g, (b) 33 g and (c) 30 g of binder.

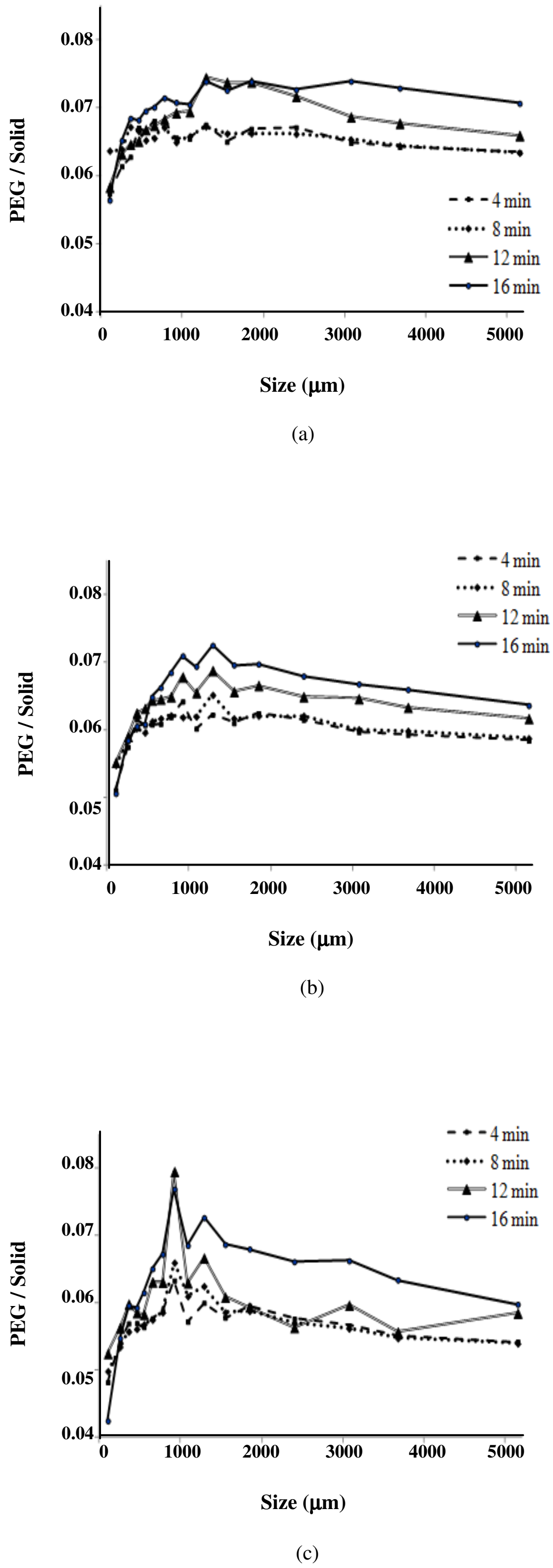


Fig. 6: Granule binder distribution at 300 rpm for (a) 36 g, (b) 33 g and (c) 30 g of binder.

At 400 rpm (Fig. 7), the variation of PEG-solid ratio across size is similar to that of 300 rpm, i.e. the binder content increases initially with size and then decreases or remains constant. However, the PEG-solid curves show different trends with time at different quantities of added binder. These discrepancies may arise from high intensity of agitation in the granulator and temperature increase during the process. For some of the experiments at 400 rpm and long granulation durations, the impeller was inevitably stopped to prevent further temperature increase and then the run was continued. This may have unintended and unknown influences on granulation behavior. So, the observed trends cannot be merely explained by dominant rate processes.

The effect of impeller speed on binder content distribution is presented in Fig. 8. In this paper, only the results of 36 g are shown for brevity, other binder values show similar trends. It is observed that for all granulation times, increasing impeller speed from 200 rpm to 300 rpm narrows the range of PEG-solid variation by promoting mixing quality and by homogenizing binder distribution. This effect is less pronounced for higher impeller velocities, due to intrinsic improved mixing quality at high impeller speeds.

CONCLUSIONS

By experimental work conducted in a conical shape, lab scale high shear granulator at relatively wide range of operating parameters, we conclude that granulation response to change in any process variable depends on all other operating variables. The results of present work are good evidence for the complexity of high shear granulation and highlight that the net effect of competing rate processes in a high shear granulator varies considerably from one operating condition to another.

Focusing on parameters interaction and its influence on granulation mechanisms, the present work supports and complements other related studies. Granules size and binder content have been examined over a suitable range of three operating variables (i.e. granulation time between 4 to 16 min, impeller speed between 200 to 500 rpm and binder mass between 30 to 36 g) and as an innovation, dominant rate process has been determined for each process condition from granule size distribution characteristics.

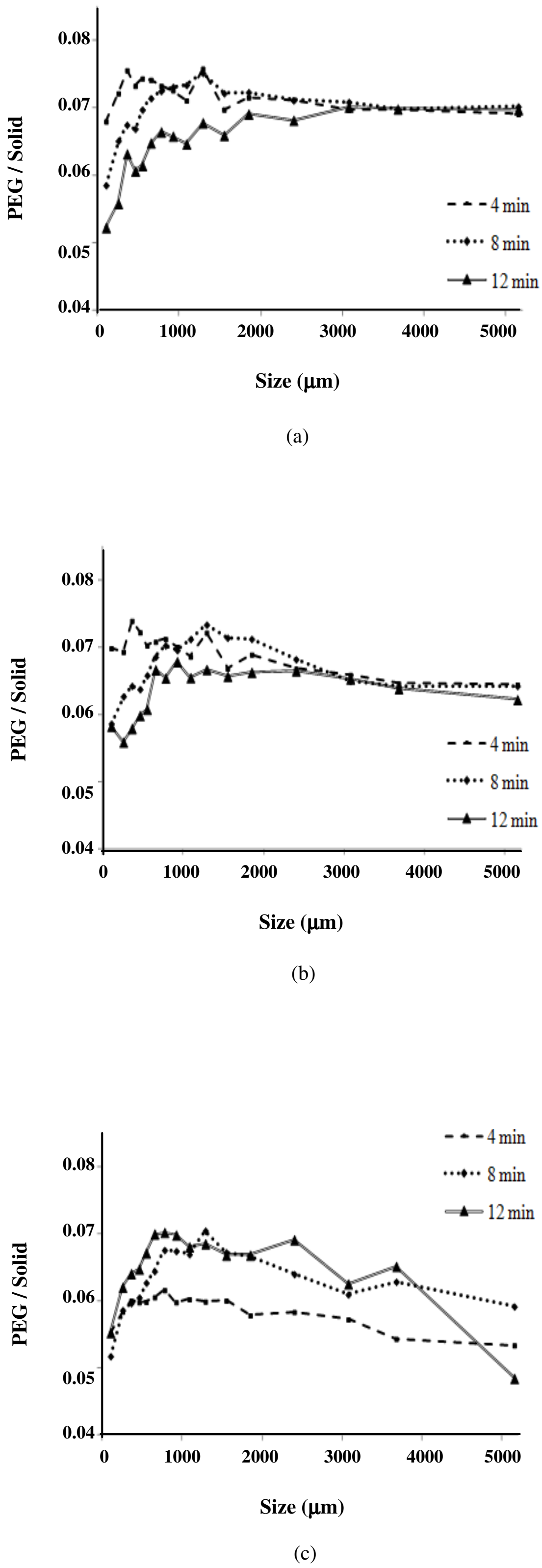


Fig. 7: Granule binder distribution at 400 rpm for (a) 36 g, (b) 33 g and (c) 30 g of binder.

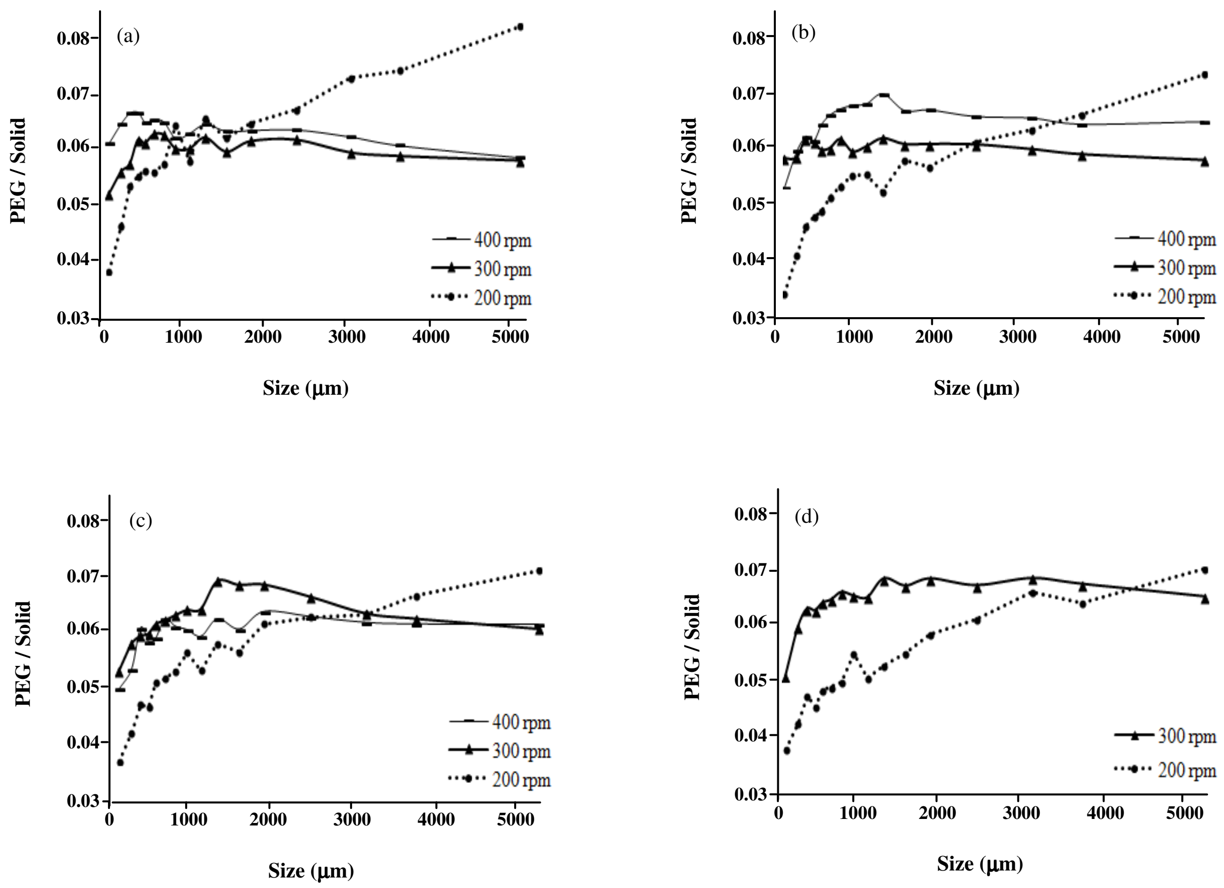


Fig. 8: Effect of impeller speed on granule binder distribution for 36 g of binder after (a) 4 min, (b) 8 min, (c) 12 min and (d) 16 min.

Nomenclature

d	Average size of collecting sieve and the one above or granule size, m
d_m	Granule mass mean size, m
d_{10}	The size which 10 % of granules are smaller than, m
d_{50}	The size which 50% of granules are smaller than, m
d_{90}	The size which 90% of granules are smaller than, m
i	Sieve number, -
m	Mass of granules retained on each sieve, kg
m_b	Mass of binder, kg
m_s	Mass of solid, kg
n	Total number of sieves, -

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