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Granule Breakage in Twin Screw Granulation: Effect of Material Properties and Screw Element Geometry

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ABSTRACT: This study is the first to explicitly measure the influence of material dynamic yield strength (DYS) and screw element geometry on the breakage process in twin screw granulation (TSG). Granule breakage is the key mechanism for controlling granule size within the TSG. Novel experiments which isolated breakage from other granulation rate processes were performed using conveying and distributive mixing element configurations and 2 and 3 mm cylindrical pellets of model materials (DYS from 0.5 to 137 kPa). Daughter size distributions and survivor pellet shape visualization was used to infer that the breakage mechanism in conveying elements (CE) is primarily edge chipping whereas in distributive mixing elements (DME), breakage is a combination of chipping and crushing. The maximum size of granule that could remain unbroken (3mm for CE and 2mm for DME) was determined by the largest available gap size in the element as measured by an analysis of the screw elements' open volume geometry. Below the maximum size, breakage probability varied inversely with granule strength up to 9kPa. For granules stronger than 9kPa DYS, breakage characteristics are independent of formulation properties and depend only on screw element geometry. This helps explain why twin screw granulation is more robust with respect to formulation changes compared to high shear wet granulation. Implications for using the results for both optimizing screw element design and calculating kinetic parameters for population balance modeling are discussed.

Keywords:

Twin screw granulation, granule breakage, conveying elements, distributive mixing elements, dynamic yield strength, granulation rate processes

1. Introduction

Granulation of powders is a widely used industrial unit operation designed to agglomerate small particles into larger granules in order to improve flow properties, minimize dust hazards, and reduce segregation risks. The process is complex and, thus, has been the topic of many research investigations [1–4]. Of particular interest in this work is continuous wet granulation. Continuous processing of material has been demonstrated to decrease product variability, offer improved process control, and improve usage efficiency. Continuous wet granulation has been commonly performed using high shear granulation and the area has been studied in detail by several researchers [2,5–9].

The present work focuses on the operation of a twin-screw granulator (TSG), a novel method for continuous granulation that has several advantages over other continuous granulators. TSGs are suitable for large material throughput, can be easily customized, have short material residence times, and reduced footprint and capital costs when compared to other continuous granulators. Hence, there has been considerable interest in TSG operation in recent years. Little is known about the physics of wet granulation in a TSG and, thus, there is potential for optimizing the twin screw granulation process.

2. Background and Objective

Twin screw granulation has been shown to provide better control over granule size and shape as compared to other granulation methods [10–12]. The differences between twin screw granulation and high shear wet granulation (HSWG), for example, are primarily due to (1) nucleation being separated, by design, from the other rate processes and (2) the short time scale of granulation in the TSG resulting in a different rate controlling mechanism in the granulator [13–16]. In order to understand what factors affect the attributes of granules produced using TSG, experiments designed to isolate the effects of particular rate processes are needed.

Several prior studies have focused on understanding the effect of screw element geometry on the performance of a TSG. It has been shown that a TSG operates in the mechanical dispersion regime with the breakage of wet mass being an important rate process for effective liquid distribution [17]. Breakage and liquid distribution in TSG is a strong function of the screw element geometry. For example, conveying elements (CEs), which are low shear transport elements, result in a bimodal granule size distribution due to poor liquid distribution [16,18,19]. Furthermore, the granule size distribution is observed to be more bimodal when the pitch of the screw elements is decreased [15]. In contrast, kneading elements exert significant shear on the wet mass and result in dense, elongated granules [13,20,21]. The offset angle between the kneading element discs and the number of kneading elements control the degree of shear within the kneading elements, which in turn affects granule size and porosity [22]. Distributive mixing elements (DMEs), also known as comb mixer elements, produce monomodal granule size distributions [14–16]. Breakage and layering have been shown to be the most important rate processes in this type of screw element. Since the screw element design is known to produce different granule properties, it is worthwhile to examine how these elements affect fundamental rate processes such as nucleation and wetting, coalescence and consolidation, and breakage [23]. In this work, we focus specifically on breakage.

Granule deformation and breakage primarily depends upon the dynamic yield strength (DYS) of the wet material, which in turn is a strong function of formulation properties such as primary particle size, binder viscosity, and binder surface tension [3,4,24] . The effects of formulation properties on breakage have been studied to some extent in the twin screw granulation literature. These studies have shown that increasing the binder concentration in the liquid phase results in a smaller mass fraction of fines and more uniform binder distribution throughout the different granule sieve cuts, especially in kneading elements [17,25]. However in conveying elements, increasing binder viscosity results in a more bimodal granule size distribution with larger mass fractions of fines and large granules [16,18,19]. The effect of changing powder binder wetting thermodynamics has been explored by using formulations with varying ratios of hydrophilic and hydrophobic powder components [26,27]. However, changing the concentration of the two powders also significantly changes the overall primary particle size distribution of the blend. This results in a convolution of the effects of the primary powder particle size as well as blend hydrophilicity on the overall granule properties. Differences in the primary particle size distribution of excipients has been shown to result in subtle differences between granule properties [17,28].

The effect of formulation properties on breakage has not been elucidated in the literature due to the use of multicomponent formulations and the fact that other rate processes occur simultaneously in the granulator. The effects of formulation properties on breakage have been studied in detail in HSWG [6,9,29], but similar studies have not been performed for a TSG. In this work, experiments have been designed to isolate granule breakage in order to study this key rate process.

3. Materials and Methods

3.1. Maximum Size Analysis

The maximum size of granules in CEs and DMEs was determined using computer aided drafting (CAD) files of the screw elements (refer to the Supplementary information for the procedure used to obtain these CAD files). Using the CAD files, the largest diameter sphere that fits between the screw element and barrel was determined, as shown in Figures 1 and 2.

3.2. Materials

Glass ballotini (Potters Industries LLC, OH, USA) of five different size cuts (0-10 µm, 63-90 µm, 125-180 µm, 180-250 µm, and 355-500 µm) were used as model material due to their spherical shape and well controlled properties (skeletal density = 2.47 g/cc) [3,6]. The particle size distributions of the glass ballotini were measured in a Malvern Mastersizer 2000 using water as the dispersing medium (refer to Figure S1 and Table S1 in Supplementary information for details of the distributions). Silicone oil (Sigma-Aldrich Corp., MO, USA, viscosity = 64 Pa.s) and a glycerol solution (Sigma-Aldrich Corp., MO, USA, water-to-glycerol ratio = 0.01 by weight, viscosity 0.7 Pa.s) were used as model binders of different viscosities [3,6]. These model powders and liquids have been previously used in mechanistic studies of wet granulation [3,6]. The silicone oil binder was dyed yellow using oil soluble aniline dye (Woodworker's Supply Inc., NC, USA; dye powder-to-silicone oil ratio = 0.01 by weight) and the glycerol solution was dyed using Nigrosin dye (Sigma-Aldrich Corp., MO, USA). Play Doh modeling compound (Hasbro Inc., USA) was used to prepare spheres of different diameters to perform preliminary breakage specific experiments in the twin screw granulator.

3.3. Pellet Preparation and Dynamic Yield Strength Measurements

To measure the dynamic yield strength (DYS) of the materials, cylindrical pellets with a height and diameter of 25 mm were prepared using a hand punch and die set. The glass ballotini and liquid binder were mixed together such that the binder-to-powder ratio by weight was 0.15. The solid fraction for these pellets was maintained at 0.63. For the 0-10 µm mixture with glycerol, the binder-to-powder ratio was 0.3 and the pellet solid fraction was maintained at 0.6. The pellet solid fractions were chosen to resemble those of granules produced by wet granulation of real powder blends. Cylindrical pellets with the same height and diameter were also prepared using Play Doh.

The DYS of each of the materials was measured using an Instron ElectroPlus E1000 material testing system with a platen impact speed of 10 mm/s. The strength of wet granules is a function of strain rate [3,30,31]. The typical particle impact speed observed in a TSG has been studied to some extent in the literature [18]. However, the results are not directly comparable to this work due to significant differences in operating conditions. On the other hand, the typical particle impact speed in a high shear granulator is estimated to be 10-20% of the impeller tip speed and is expected to be larger than in a TSG. Hence, the platen speed used in this study is approximately of the order of magnitude of 10% of the tip speed of the screw. The platens were greased with Vaseline in order to reduce the friction of the pellet with the platen surface during pellet deformation. The detailed methodology for pellet preparation and DYS measurements can be found elsewhere [3,32]. Images of the pellets before and after DYS measurement are shown in Figure S2 (Supplementary Information). A model material library was constructed by characterizing the materials using the DYS.

Preliminary breakage experiments were performed for CE and DME elements using Play Doh spheres in order to understand the effect of granule size on the breakage probability. For the CEs, Play Doh spheres of diameter 1.0 ± 0.1 mm, 2.0 ± 0.1 mm, 3.0 ± 0.1 mm and 4.0 ± 0.1 mm were prepared. For the DMEs, spheres of diameter 1.0 ± 0.1 mm, 2.0 ± 0.1 mm and 3.0 ± 0.1 mm were prepared. Twenty spheres were prepared for each diameter and the diameter of each sphere was verified using caliper measurements. For breakage specific experiments, cylindrical pellets of the model material systems of height 2 ± 0.1 mm and 3 ± 0.1 mm (height/diameter = 1) were prepared using a Natoli Carver Press tooling standard punch and die set (Natoli Engineering Company Inc., MO, USA) in the Instron ElectroPuls E1000 materials testing system. Pellet-to-pellet variation was minimized by maintaining uniform conditions for all pellets such as pellet weight, porosity, compressive stress and speed of compression. Although a spherical pellet is more representative of a granule, it was not possible to prepare a spherical pellet while maintaining uniform pellet preparation conditions. Twenty pellets were prepared for each model material. Images of the pellets are shown in Figure S3 (Supplementary Information).

3.4. Breakage Specific Experiments

The twin screw breakage specific experiments were performed in a EuroLab 16 mm Twin Screw Granulator (TSG), 25:1 length-to-diameter (L:D) ratio (Thermo Fisher Scientific, Karlsruhe, Germany). The TSG consists of two intermeshed, co-rotating screws encased in a segmented barrel with powder and liquid inlet ports. The granulator can be customized with several different types of screw elements such as conveying elements (CE), kneading elements (KE), and distributive mixing elements (DME) [33]. This study is focused on understanding the breakage phenomena in CEs and DMEs. For all experiments, microcrystalline cellulose (MCC) was used as a free flowing powder medium in order to simulate real flow conditions in the TSG. The MCC was fed into the TSG using a gravimetric feeder (Brabender Technologie, ON, Canada). The experiments were performed at 400 RPM with a 4 kg/h flow rate of MCC. Figure 3 shows a schematic of the TSG, which is divided into six zones of length 60 mm each and a 20 mm section on the end. The powder is fed from zone 4 for all experiments as shown in the Figure 3. Fifteen pairs of 16 mm double-flighted conveying elements L:D = 1 were used for CE breakage specific experiments. The pellets were fed one by one from zone 4 along with the powder feed. For DME breakage specific experiments, three pairs of DMEs were used in the Adjacent Reverse configuration [14] as shown in Figure 3b. The DMEs were placed at the front end of the granulator with CEs placed upstream to aid the transport of MCC into the DME zone. In order to make sure that the pellets only encounter DMEs, the pellets were fed one by one from a small aperture in the center of the 20 mm section in front of zone 1 as shown in the figure.

The pellet remnants and MCC were collected at the outlet of the granulator. Additional images of the experimental set up are shown in Figure S4, S5, and S6 (Supplementary information). All pellets that retain 75% or larger of their initial volume are considered “survivors”. For 3 mm pellets, the equivalent cylinder size (height/diameter = 1) of survivors is 2.8 mm and for 2 mm pellets it is 1.8 mm. A 2.8 mm sieve was used to separate the survivors in the case of 3 mm feed pellets and a 1.7 mm sieve was used to separate the survivors in the case of 2 mm feed pellets. For Play Doh spheres, the breakage does not necessarily result in several broken pieces due to the sticky nature of the modeling compound. Hence, any sphere showing a crack length equal to or larger than the sphere radius was considered a broken pellet. The daughter size distribution of the broken pellet pieces was measured using a $\sqrt{2}$ series of sieves from 0.5 to 2.0 mm. It was not possible to separate broken material smaller than 0.5 mm from the MCC because of the presence of MCC in lower sieve cuts. Daughter size distributions were plotted as the normalized mass frequency of the logarithm of the particle size according to,

$$f_i(\ln x) = \frac{y_i}{\ln(x_i/x_{i-1})}, \qquad (1)$$

where y_i is the mass fraction in size interval i and x_i is the midpoint of the size interval i. Two replicate experiments were performed for the 3 mm pellets in the conveying element configuration and 3 mm and 2 mm pellets in the DME configuration. Both sets of data are shown in the graphs.

3.5. Three dimensional survivor shape characterization

The survivor pellets were imaged using a Nikon SMZ-1500 Stereoscopic Zoom Microscope. The pellet was placed next to a prism in order to record the side and top views simultaneously. A representative survivor for each model material and screw configuration was chosen to study the survivor shape characteristics.

4. Results and Discussion

4.1. Geometric Analysis of Screw Elements

Figure 1 shows a CAD drawing of the CEs used in this work enclosed in the barrel of the TSG. Each CE is double flighted and has a length of 16 mm, diameter of 15.60 mm and the perpendicular distance between the two flights is 7.60 mm. The space between the flights and the barrel is the maximum space available for material in the CEs. When the twin screw with CEs is enclosed in the barrel, a sphere of maximum diameter 3.49 mm can fit in the region between the two flights of the CE and the barrel and is considered to be the maximum diameter of CEs.

For a pellet in the CEs, there are two paths that are possible as shown in Figure 1:

1) The pellet follows a helical path along the flights
2) The pellet is conveyed axially by the axial component of the flight velocity

In both cases, the pellet is always enclosed in the region between the flights and the barrel. Thus, the pellet always experiences a region of constant maximum diameter of 3.49 mm. Any granular material is therefore expected to have a maximum size less than or equal to this maximum diameter in CEs.

Figure 2 shows a CAD drawing of the DMEs used in the current work enclosed in the barrel of the granulator. Each DME has six teeth and the radius to the tip of the teeth is 7.8 mm. The thickness of the blades of the DME is 1.69 mm. The region between two DME teeth and the barrel is the maximum space available for the material to pass in the DME configuration of the granulator. The biggest possible sphere that can just fit in this space has a diameter of 3.18 mm, which is then the maximum diameter in DMEs.

As mentioned in previously, a pellet in the conveying elements can follow two possible unobstructed paths that offer a region of constant maximum size of 3.49 mm. However, in the case of DME, the direction of flow of the material in the twin screw granulator is perpendicular to the direction of rotation of the DME. Hence, there are two possible breakage mechanisms in DMEs:

1) Breakage of the pellet between the DME blades and the barrel
2) Breakage of the pellet by getting caught in the intermeshing zone of two DMEs.

It is important to consider both breakage mechanisms in a DME, as will be shown later.

4.2. Model Material Library

The model material library was characterized by measuring the DYS of the model material mixtures described in Section 3.3. A typical stress-strain curve for the model materials is shown in Figure S7 (Supplementary information). Figure 4 shows that the DYS of the materials decreases as the particle size of glass ballotini increases and as the binder viscosity decreases.

These results are consistent with previous work [3,4,23]. The results in figure 4 cover a Capillary number range from 0.001 to 0.5. In this range, viscous dissipation, interparticle friction and capillary forces all contribute towards the plastic deformation of the pellet and DYS will increase with binder viscosity and be approximately inversely proportional to primary particle size [3,34,35]. Furthermore, the DYS of the model materials span a wide range with the 0-10 μm glass ballotini and glycerol mixture being more than 100 times stronger than the 355-500 μm glass ballotini and glycerol mixture. This large range allows for a more complete understanding of the breakage mechanism for a wide variety of formulations in a TSG. The DYS of Play Doh was also measured in order to compare to the breakage studies using the model materials. The DYS of the Play Doh was found to be 13.5 ± 0.8 kPa.

4.3. Breakage in TSG: Effect of Granule Size

Figure 5 shows the breakage probability of the Play Doh spheres as a function of the sphere diameter for the CEs and DMEs. A 5% breakage probability represents one broken sphere. The scatter bars represent plus and minus one standard deviation in the measurement from three replicate experiments.

It is interesting to observe that spheres of diameter 4 mm showed 100% breakage probability while spheres of diameter 3 mm and smaller showed a negligible breakage probability in CEs. It was concluded that the spheres of diameter 4 mm were larger than the maximum size in the CEs and hence showed 100% breakage. Unlike the CEs, which show a significant increase of breakage probability from 0% to 100% as the size of the spheres is increased from 3 mm to 4 mm, the DMEs show a more gradual increase in breakage probability as the size of the sphere increases from 2 mm to 3 mm. Furthermore, the DMEs show 100% breakage of the 3 mm spheres, which is slightly smaller than the maximum size in the DME. The reason for this last observation is explained in section 4.5. It is expected that granules larger than 3 mm will also show 100% breakage in DMEs.

4.4. Breakage in Conveying Elements

The breakage experiments for CEs were performed using cylindrical pellets of diameters (height = diameter) 2 mm and 3 mm as described in Section 3.4. The number of survivor pellets out of 20 feed pellets for each material was noted and the percentage of pellets broken, or breakage probability, was plotted as a function of the DYS as shown in Figure 6. A 5% breakage probability represents one broken pellet. The number of survivors for each material was reproducible within 15% and was considered acceptable due to the stochastic nature of the path undertaken by the pellets. The horizontal scatter bars for the DYS represent plus minus one standard deviation in the measurement from five replicate measurements. Some of the data do not show scatter bars due to the scatter being smaller than the size of the marker.

For the 3 mm pellets, the breakage probability decreases as the DYS of the material increases. Interestingly, for materials having a DYS of 9 kPa or larger, none of the pellets broke. This result indicates that formulations with a strength larger than this critical value can resist breakage independent of their DYS. It is important to note that the Play Doh pellets of 3 mm diameter also showed 0% breakage probability. The DYS of the Play Doh was found to be larger than 9 kPa and thus is consistent with the other formulation results. The 355-500 microns glass ballotini and glycerol pellets were the weakest materials and showed 50% breakage probability. Any material weaker than this would slump under its own weight and, hence, it was not possible to prepare a self-sustaining pellet of weaker materials. It is also important to note that real powder formulations also typically have DYS values greater than 9 kPa [37,38]. In contrast, the 2 mm pellets show negligible breakage for all of the materials considered. The reason for this behavior is explained in the following paragraphs.

Figure 7 shows sectional views of the 2 mm and 3 mm pellets. The diameter and height of the pellet are 3.00 mm each whereas the diagonal length of the pellet is 4.24 mm. Figure 8 a-i) shows images of the typical survivor pellets for each model material. The pellets having DYS values larger than the critical value had distorted shape, but retained most of their mass. The weaker pellets showed a larger loss of mass with chipping primarily across the diagonal, which is greater than the maximum opening size in CEs. The height and diameter of the 2 mm pellets as well as the length of the diagonal are smaller than the maximum size in CEs and show zero breakage probability independent of the DYS of the material. Based on these observations, we conclude that if a granule has a size less than the element maximum opening size, then the breakage probability is extremely low (or near zero) irrespective of the material's DYS. If a granule has a size greater than the maximum size, then the breakage probability will decrease with an increase in the DYS.

Figure 9 shows the daughter size distribution of the 3 mm broken pellets passing through CEs. The original size of the pellet is marked by the vertical dotted line. The distribution shows a sharp peak at 2.4 mm along with a tail of fines for all of the materials considered. The shape of the distribution is characteristic of an attrition process where the sides of the pellet are chipped, retaining most of the mass of the pellet. The mean daughter size distribution shows a shift towards smaller granule sizes as the DYS decreases. This result shows that in CEs, materials weaker than 9 kPa are expected to undergo increased attrition resulting in a larger generation of fines. The daughter size distribution shows a peak at a mean sieve size of 2.4 mm. Considering 2.4 mm to be the height of the broken pellet, the length of the diagonal is 3.4 mm, which is the maximum size of the CEs. This trend demonstrates that the pellets undergo chipping primarily along the diagonal and are size reduced to the maximum opening size of the CEs.

4.5. Breakage in Distributive Mixing Elements (DMEs)

The breakage experiments in DMEs were conducted as described in Section 3.4 by using cylindrical pellets with heights (height = diameter) of 2 mm and 3 mm. Figure 10 shows the breakage probability as a function of the DYS for 3 mm and 2 mm pellets in the DMEs. All of the 3 mm pellets demonstrated breakage and showed zero survivors independent of the DYS. The breakage probability for the 2 mm pellets follows an interesting trend in which materials having a DYS greater than 9 kPa show a constant breakage probability of 20%, independent of the strength. One should note that the breakage probability of the 2 mm Play-Doh spheres was also 20%, as shown in Figure 5. Section 4.1 showed that the maximum opening size in the DMEs is 3.18 mm. Even though the 2 mm pellets have dimensions smaller than the maximum opening size in DMEs, breakage is observed for all of the materials considered.

The daughter size distribution of the 3 mm pellets and the 2 mm pellets aids in the understanding of the breakage mechanisms in DME. The daughter size distribution for the 3 mm pellets is shown in Figure 11. The original size of the pellets is marked by the vertical dotted line in the figure. The distribution is broader than those observed with CEs and is indicative of a crushing mechanism. The daughter size distribution is similar for all materials of dynamic yield strength greater than 9 kPa and shows a subtle shift to the left for the weaker materials.

Figure 12 shows the daughter size distribution of the 2 mm pellets in the DMEs. The daughter size distribution does not show a significant difference for all the materials considered. The weaker materials showing slightly higher amount of fines as expected.

As noted in Section 4.1, there are two possible breakage paths in DMEs. Chipping of the pellets will occur if the pellet is pressed between the teeth of the DME and the barrel. The crushing type breakage, on the other hand, will occur if the pellet is nipped in the intermeshing zone of the DMEs and will result in complete breakage of material. The probability of crushing breakage appears to be 20% for the 2 mm pellets and is independent of the DYS of the material. This observation is unique to TSG, as breakage-specific studies performed in high shear granulation show a strong dependence of breakage probability on material DYS [6]. There are currently no references about the influence of DYS on the probability of crushing breakage in TSG. All of the 3 mm pellets, in contrast, show crushing, as observed in Figure 11. As crushing results in complete breakage, any pellet experiencing nipping is not expected to survive. Hence, all the 2 mm survivor pellets experienced only chipping and did not get caught in the zone between the two DMEs. The chipping breakage probability is a function of the DYS of the material and increases as the DYS of the materials decreases. Materials having a DYS less than 9 kPa undergo both crushing and chipping breakage. On the other hand, materials having dynamic yield strength greater than 9 kPa show breakage only due to crushing as they are strong enough to resist chipping. It is important to note that the mode of the daughter size distribution is positioned at a sieve size of 1.70 mm (fragments in the sieve fraction 1.4-2.0 mm) for both 3 mm and 2 mm pellets in the DME as shown in Figures 11 and 12, respectively. This size is equal to the thickness of the blade of the DME, which is 1.69 mm as described in Section 4.1. This result is because the pellets are sliced by the blades of the DMEs with fines generated in the process. The daughter size distribution for the 3 mm pellets also shows some daughter pieces having a mean size greater than 1.70 mm and is possibly due to parts of the pellet escaping the intermeshing zone while traveling axially.

The chipping mechanism in the DMEs is different than in CEs. As mentioned previously, the material flows in the axial direction while the DMEs rotate radially and perpendicular to the direction of the flow of the granular mass. Hence, the survivors experience attrition from all sides due to impact of the DME blade and do not have a preferential axis of breakage. Figure 13 shows the images of the 2 mm pellet survivors after passing through the DMEs. As mentioned previously, all the survivor pellets experienced only chipping. The materials stronger than 9 kPa show deformation of the pellet from all sides whereas the weaker materials demonstrate a larger mass loss.

4.6. Understanding the performance of real formulations in Conveying and Distributive Mixing Elements

In a twin screw wet granulation operation, the liquid is typically injected using a drip nozzle positioned above the CEs [14,17]. The material in the twin screw granulator is, thus, a mixture of wet powder mass enveloped in dry powder. At low liquid to solid ratio, the primary rate process for liquid distribution is breakage and the rate of coalescence is low [17]. Due to the low shearing characteristics of CEs, wet mass is sheared by chipping to granules no larger than 3.49 mm. The chipped pieces along with the un-wet powder are accounted for in the fines region of the granule size distribution. Therefore, the granule size distribution for CEs is observed to be bimodal in shape. It is expected that granule size distributions in real formulations are also bimodal in nature with granules no larger than the maximum size in CEs in the breakage controlled regimes. This trend has been observed in several published reports using a similar scale of TSG geometry as used in this work [10,13,14,36]. However, it should be noted that although the breakage probability in the CEs is a strong function of the DYS, materials stronger than 9 kPa are able to resist damage.

Granule breakage in DMEs is a combination of chipping and crushing, and is characterized by the mode of the distribution positioned at a size equal to the thickness of the DME blades. The breakage in DMEs results in a broad monomodal granule size distribution with no dependence on granule strength. Hence, the granule size distribution is expected to have a peak at a granule size of 1.70 mm, which is the thickness of the DME blades. This trend has been observed in the literature using the same scale of twin screw granulator geometry as used in this work [14].

These results help explain why TSG is relatively robust in the face of changing formulation properties when compared to high shear wet granulation. Breakage is a key rate process that determines granule size and liquid distribution in TSG. Most real formulations have DYS values greater than 9kPa [37,38]. Under these conditions, the breakage characteristics are a function of process conditions only, especially screw element geometry, and not a function of granule strength. This suggests that measurement of the DYS of a new formulation is a quick and easy screening test to determine whether or not results during design and scale up will be sensitive to formulation.

The data presented for the breakage probability and the daughter size distribution can be directly used in the population balance modeling (PBM) of twin screw granulation in the breakage rate expression:

$$R_{breakage} = \dot{b}_{break} - \dot{d}_{break}$$

$$\dot{d}_{break}(s,l,t) = k_{break}(s,l) \times n(s,l,t)$$

$$\dot{b}_{break}(s,l,t) = \int_{s}^{\infty}\int_{l}^{\infty} B_{break}(s,l,s',l') \times k_{break}(s',l') \times n(s',l',t)ds'dl'$$

Where, $\dot{b}_{break}(s,l,t)$ is the birth rate of particles, $\dot{d}_{break}(s,l,t)$ is the death rate of particles, $k_{break}(s,l)$ is the breakage probability , $B_{break}(s,l,s',l')$ is the daughter size distribution and $n(s,l,t)$ is the number of particles as a function of time, defined by the solid content, liquid content in the particles. $k_{break}(s,l)$ is directly related to the measured breakage probabilities, and $B_{break}(s,l,s',l')$ are directly measured in this paper as a function of material properties and screw geometry without conducting full granulation experiments and therefore provide a good methodology for developing predictive PBM rate expressions for breakage.

5. Conclusions

The effect of formulation properties and screw element geometry was studied in a twin screw granulator using breakage specific experiments. The maximum size in conveying elements is determined by the space available between the two flights of the conveying element and the barrel. For conveying elements, the breakage mechanism is primarily chipping, in which granules larger than the maximum size undergo deformation or breakage. Granules smaller than the maximum size do not experience breakage due to the low shearing characteristics of the conveying elements. Breakage in distributive mixing elements is a combination of chipping and crushing mechanisms. The maximum size in the distributive mixing elements depends on the space between the teeth of the elements and the barrel. For granules greater than the critical size, 100% breakage is observed independent of the DYS of the materials and results in a broad monomodal daughter size distribution with mode corresponding to the thickness of the DME blades. The breakage probability for granules smaller than the maximum size is dependent on the granule strength and shows a combination of attrition and crushing breakage. The granule size distribution also exhibits the mode at a size equal to the thickness of the distributive mixing elements.

The understanding of the geometric aspects of the screw elements and their effect on the maximum size of granules from a Twin Screw Granulator is a step towards tailored granule size distributions. By changing the geometry of the screw elements, it is possible to change the maximum granule size produced by the granulator. By choosing a certain type and design of the screw elements, the breakage mechanisms in the granulator can be modified resulting in broad monomodal or bimodal granule size distributions. This understanding holds promise for designing screw element geometries to achieve better control over granule properties.

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