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ASYMMETRIC DISTRIBUTION IN TWIN SCREW GRANULATION

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ABSTRACT

PEPT (Positron Emission Particle Tracking) was successfully employed to validate measured transverse asymmetry in material distribution in the conveying zones of a Twin Screw Granulator (TSG). Flow asymmetry was established to be a property of the granulator geometry and dependent on fill level. The liquid distribution of granules as a function of fill level was determined. High flow asymmetry at low fill level negatively affects granule nucleation leading to high variance in final uniformity. Wetting of material during nucleation was identified as a critical parameter in determining final granule uniformity and fill level is highlighted as a crucial control factor in achieving this. Flow asymmetry of dry material in conveying zones upstream of binder fluid injection leads to poor non-uniform wetting at nucleation and results in heterogeneous final product. The granule formation mechanism of 60°F kneading blocks is suggested to be primarily breakage of agglomerates formed during nucleation. Optimisation of screw configuration would be required to provide secondary growth. This work shows how fill dependent flow regimes affect granulation mechanisms.

KEYWORDS

Twin Screw Granulation, Continuous Processes, Pharmaceutical Technology, Nucleation, PEPT

1. INTRODUCTION

Granulation is a size enlargement process wherein particles are brought together to form permanent agglomerates to improve their handling and mechanical properties. Twin screw granulation (TSG) is a new method of continuous wet granulation developing considerable interest within the pharmaceutical industry. Granulation has traditionally been employed as a batch operation within pharmaceutical processes. Recently development has shifted in focus towards continuous production due to the perceived achievable benefits in control and efficiency[1]. A twin screw granulator consists of two co-rotating screws confined in a barrel. Screws are modular and consist of conveying and mixing elements. Powder is fed into the base of the barrel and transported by conveying elements. Liquid is added to bind particles together and mixing zones typically consisting of blocks of broad kneading elements provide the intensive mixing and densification required for granulation to take place.

In the past two decades the depth of research into TSG has increased considerably. Research has looked into the response to process parameters including: screw speed [2-4], material feed rate [2,5-7] and liquid to solid ratio [4,8-10]. Despite this the mechanisms of TSG are still not well understood, the modular nature of the screws and differences in granulator size and geometry makes drawing comparison between different granulators difficult. Visualisation of the flow inside the granulator is inherently difficult due to the requirement of running within an enclosed barrel. Granulation mechanisms must usually be inferred from final granule properties. Dhenge et al [11] examined the steps in granule growth by stopping an actively running process and extracting samples of granules from the different regions of the granulator. El Hagrasy & Litster [12] used three dimensional shape characterisation to develop concepts for the dominant granulation rate mechanisms in the mixing zones of a twin screw granulator. Kumar et al [13] employed near infra-red chemical imaging in order to determine residence time distribution (RTD) and infer the degree of axial mixing with changes in process parameters. Visualisation of the flow of material inside an actively running granulator was achieved by Lee et al [14] who employed PEPT (Positron Emission Particle Tracking) to determine RTD and fill level occupancy across individual screw elements.

There remains a need to understand processes occurring within the granulator. This work examines fundamentals of material flow within conveying zones of the granulator and their dependence on fill level. Liquid distribution is examined and the importance of nucleation in determining granule properties is highlighted. Finally the conveying zone mixing efficiency is examined and typical granulation formation mechanisms suggested.

2. EXPERIMENTAL METHOD

Preparation of powder formulation

For granulation experiments a formulation consisting of 75% alpha lactose monohydrate (316/FAST-FLO®, Foremost Farms, USA), 20% Microcrystalline Cellulose (Avicel PH101, FMC BioPolymer, Ireland) and 5% Hydroxypropyl Cellulose (Klucel EXF, Ashland Inc, USA) was used. The formulation was blended in a Pascal lab mixer for 25 minutes to ensure homogenous mixing. Median particle size was determined to be ~125 µm through image size analysis (Sympatec QICPIC).

Granulation process

Granulation was performed using a lab scale co-rotating Twin Screw Extruder (TSE) (Haake, Thermo Scientific, Germany) with screw diameter of 16 mm and length to diameter ratio of 25:1. Granulation was performed based around screw configurations consisting of a single 60° Forwarding kneading block, assigned the notation [18x 1D **C**//4x 0.25D **K** 60°F//6x 1D **C**] where D represents the screw diameter, **C** conveying elements and **K** kneading elements. Thus from the base the configuration consists of 18 conveying elements each 1 diameter in length, 4 kneading elements each 0.25 diameters in length offset at an angle of 60° in the forwarding direction and finally 6 conveying elements each 1 diameter in length. Powder was fed into the barrel at the base of the screws via a volumetric twin screw feeder (T20, K-Tron Soder). Granulation liquid (distilled water) was added through a single injection port positioned above the screws approximately 9 diameters length from the base and provided by an 8 roller peristaltic pump (REGLO Digital, Ismatec, Swtizerland).

Determination of Transverse Distribution in Conveying Zones as a Function of Fill

In order to determine the mass load conveyed by each screw the Haake TSE was fitted with screws consisting of only conveying elements (25x 1D **C**). The dry formulation was fed through to the granulator and the fill level varied through material feed rate (1-7 kg/h) at set screw speed (100 and 400 rpm). A stainless steel sheet was positioned at the outlet aligned with the centreline to physically separate the material as it was discharged from each screw into separate vessels. The mass of material collected in each container at steady state was measured. The distribution of material at the discharge is believed to be representative of the transverse distribution of material across the entire conveying section.

Size Selective Segregation

The extent of size segregation in conveying zones was examined by feeding a bimodal mixture of spherical MCC pellets through the long conveying section of the Haake extruder. The mixture consisted of a 50/50% (w/w) blend of 1000 μm and 100 μm pellets (Cellets 1000 & Cellets 100, Pharmatrans Sanaq AG, Switzerland). No granulation liquid was added to ensure the surface forces between particles and the screws was the same and segregation was as a result of differences in particle volume only. Material was separated at the discharge as described above and the size distribution determined by the mass fraction of material which could be passed through a 500 μm aperture sieve.

Positron Emission Particle Tracking (PEPT)

PEPT was employed in order to determine the transverse distribution of material in the conveying zones of an actively running twin screw granulator. The experimental setup for PEPT was that of Lee et al [14] and the data was reprocessed in order to determine the transverse distribution of material in conveying zones. PEPT is a technique which allows for the tracking of a radioactive tracer particle in three dimensions within an actively running process. The tracers used in the experiments were 100 μm ion exchange resin particles labelled with Fluorine-18 through an ion exchange technique. ^{18}F was selected as the tracer radionuclide as it undergoes beta decay, has a short half-life and degrades to water. As the tracer undergoes beta decay it releases a positron which very quickly collides with a local electron and is annihilated to release two "back to back" 511 keV gamma rays. PEPT cameras are used to detect the γ-rays and the position of the tracer is determined from the intersection of multiple pairs. PEPT visualisation was performed on a specially modified twin screw granulator provided by GEA Niro, UK. The granulator barrel was machined down to a low thickness to minimise the attenuation of gamma rays and maximise tracer detection. The screws of the granulator were 19 mm in diameter with a length to diameter ratio of 10:1.

The tracer was fed through the granulator a minimum of 100 times. For each pass through the granulator the time spent by the tracer on either screw in conveying zones was determined, which is representative of the distribution of material. Due to the short residence time of material in conveying zones, the final transverse distribution was determined from the summation of the time spent by the tracer in all passes.

Liquid Distribution

The effect of fill level on nucleation and final liquid distribution was investigated in this study. Granules were produced using the Haake TSE, local fill level was controlled by including elements with longer residence times immediately upstream of the point of liquid injection. Long residence time elements have correspondingly high local material fill levels. Overall fill level was controlled through material feed rate at set screw speed. The screw configurations used to achieve desired fill level distributions are shown in Table 1. Fill levels were not measured directly and ‘high’ and ‘low’ fill levels are qualitative relative descriptions. Granulation was performed at 400 rpm screw speed and 2 kg/h mass feed rate. Liquid distribution was first determined gravimetrically through loss on drying using the following method. Liquid was fed into the granulator at a liquid to solid ratio (L/S) of 0.15, after production of a minimum of 100 g of granules they were immediately transferred to a series of sieves (British Standard 5600 - 63 μm). Granules were sieved using a vibratory sieve shaker (Fritsch, Germany) at an amplitude of 2 mm for 10 minutes, samples of a minimum of 1 g were taken from each sieve and weighed. Granules were then oven dried at 100°C for 1 hour in order to measure the loss on drying.

Liquid distribution was secondly determined through tracer analysis following a procedure similar to el Hagrasy et al [12]. A 0.1% (w/v) Nigrosin dye solution was used as the granulation liquid. Granules were produced at an L/S ratio of 0.18 and oven dried at 100°C for 2 hours. Dried granules were sieved through the same series of 14 sieves. Samples representing less than 1% of the original sample mass were discarded. 2 g samples were taken from each sieve and dissolved in 10 ml of distilled water. Samples were sonicated for 1 hour to ensure complete dissolution of granules before being centrifuged at 4000 rpm for 10 minutes in order to separate insoluble excipients. Approximately 1.5 ml of the supernatant was drawn off each sample and the dye concentration determined from absorbance of light at 565 nm using a UV/Vis spectrophotometer (Aquarius 7500, Cecil, UK).

Table 1. Screw configurations explored in liquid distribution study.

3. RESULTS

Asymmetric Material Distribution in Conveying Zones

The distribution of material in conveying zones is highly asymmetric, with one screw carrying the bulk of material. Figure 1 shows the distribution of material collected at the discharge of the granulator outfitted with conveying screws only. In order to differentiate between the two screws the well filled right screw has been termed the 'driving' screw as it carries the bulk of material forward while the poorly filled left screw has been termed the 'loading' screw as the main function is to load material onto the driving screw. The granulator operates in a starved fed fashion where material flow rate is determined by an upstream feeder. Increasing feed rate will lead to increasing fill level up to the point of complete fill at places in the barrel.

Figure 1. Transverse material distribution of dry ungranulated formulation conveyed at a range of feed rates a) 100 rpm screw speed, b) 400 rpm screw speed.

Figure 1b Shows that at low barrel fill level at low feed rates the transverse discharge of material is totally asymmetric with 100% of material discharged from the driving screw. The transverse distribution only approaches homogeneity at the maximum conveying capacity of the granulator at approximately 4.5 kg/h feed rate and a reduced 100 rpm screw speed in Figure 1a. The distribution of material at discharge is representative of the transverse distribution of material throughout the entire length of the conveying zone assuming uniform fill level.

Figure 2 – Example image of the HAAKE TSE showing the distribution of material during granulation. Looking down the barrel rotation is counter-clockwise.

Figure 2 illustrates how conveying asymmetry can appear during granulation, note the well filled driving screw and poorly filled loading screw. Here the granulator had been brought from running to an emergency stop, so the loading of material is representative of the actual case during granulation. In this example the kneading block is operating highly filled and the fill level in the conveying section is comparably low. As the residence time of the kneading block is much longer than conveying sections it defines the feed rate the granulator is capable of operating at. Feed rates above this will lead to accumulation downstream and eventually blockages. As such, conveying sections in granulation systems are likely to typically operate partially filled.

Figure 3. a) Overhead view of the GEA Niro TSG showing each detection of the PEPT tracer in 100 passes through the granulator at 10 g/min and 150 rpm b) End on view showing the position of the tracer in the conveying sections of the TSG, rotation is counter clockwise and flow in direction of the reader c) Single pass of a tracer through the TSG with screw configuration [4x 0.25D **K//** 4x 1.5D **C**// 6x 0.25D **K**// 1x1.5D **C**] to illustrate conveying load asymmetry, axial flow is away from the reader and rotation is clockwise.

PEPT visualisation of the conveying zones of a twin screw granulator was undertaken in order to confirm the measured asymmetric transverse distribution. Figure 3a illustrates an overhead view of the GEA TSG with screw configuration [2x1.5D **C**//7 x 0.25D **K** 30°F//1x 1.5D **C**//6x 0.25D **K** 30°F//1 x 1.5D **C**] where each data point represents a detection of the tracer in approximately 100 passes through the granulator. The detection density is much higher in the mixing zones of the TSG indicative of the much longer residence time compared to conveying zones. Figure 3b shows the vertical and transverse position of tracer detections within the conveying sections of the same geometry. The higher detection density to the bottom right of the image indicates the higher material loading of the driving screw. Figure 3c illustrates an example flow path of a tracer through the TSG with a single mixing zone showing the asymmetrical conveying flow path. The results of the PEPT study are shown in Table 2. The fraction of time spent by the tracer on the driving screw is greater than 50% for all the conditions investigated. Implying evidence toward the predicted flow asymmetry in conveying zones. The original PEPT experimental setup had a broader investigative focus. Data was reprocessed to look exclusively at conveying zones in an effort to capture novel information. Due to this the useful measurement time is relatively small. In addition the flow of wet granular compared to dry powder material is likely to be different. As such the values generated through processing the PEPT data are not necessarily representative of all systems. In spite of this, as a tool for visualising the flow and distribution of material PEPT adds valuable evidence toward the measured and expected flow asymmetry.

While the time spent in conveying is so low, asymmetry here upstream of fluid addition could be very important. The difference in distribution of material is not as extreme as may be predicted from the outlet distribution of the Haake extruder in Figure 1 due to the higher overall fill level established by the mixing sections. Additionally the wet granular material is 'stickier' than the dry powder formulation fed through the Haake extruder, as such material is more likely to adhere to the screw surface under these conditions promoting the transfer from the driving to the loading screw. By adding liquid the ‘rheology’ of the material changes, wet granular material will have a higher wall friction angle aiding the transfer of momentum from the screw. This makes it more inclined to follow the rotation of the screw and be transferred to the loading screw. It is then interesting to note that despite this, differences in screw material loading can be observed through PEPT. When considering the transfer of material between screws with granulators from different manufacturers the differences in screw clearances may have an important impact. Despite the differences in screw diameter the granulators in this study had similar screw-screw and screw-barrel clearances (~0.2 mm), thus any effect of clearances is expected to be similar. In systems where clearances are larger or smaller, particularly with respect to the particle size flow patterns may be different. Larger clearances may result in new flow paths, such as between the screws or extruded against the barrel wall which may affect the transfer of material between screws.

Table 2. Sum of time spent by the PEPT tracer in each zone of the granulator.

Figure 4. Front on schematic representing the transverse fill profile which establishes across the entire length of conveying zones and the transfer of material between screws. Axial flow is toward the reader.

The asymmetric distribution of material in conveying zones is a result of the screw geometry and the effect of gravity. Figure 4 illustrates the transverse distribution of material in conveying zones. In the partially filled environment of conveying zones the bulk material will tend to remain below the screws under gravity. Under rotation, the flight of both screws will push material axially toward the barrel discharge and sideways in the direction of rotation. The screws are co-rotating meaning that material below the screws is conveyed in the same direction. This results in material being conveyed from the loading screw onto the driving screw and from the driving screw against the side wall of the granulator. As a result the rotation of the driving screw acts mainly to drive material axially forwards. For material to follow the rotation of the driving screw there has to be sufficient adhesion between the material in the limited volume of the flight channel and surface of the screw. Carryover of material to the top of the driving screw is thus dependent on the fill level in conveying zones, the force between material and screw surface, the stickiness and wall friction angle of material. Carryover of material will increase with fill level: under well filled conditions the radial flow pattern above the screws will be reverse of that below the screws, with the driving screw loading material onto the loading screw resulting in a "Figure of eight" flow. This is likely to only occur when the volume of material exceeds the free volume of the driving screw. Under low fill conditions material will remain isolated in the flight chambers of the driving screw undergoing plug like flow with minimal transverse mixing. In general the fill level in conveying zones will be low, to achieve high fill levels in conveying zones a high overall fill level is required which may result in blockages forming in mixing zones [15].

Size Selective Segregation in Conveying Zones

The crucial factor in flow asymmetry is the resistance to transfer of material from below the driving screw. The force acting on the material comes from the friction or transfer of momentum from the screw and resistance to flow from gravity acting on the material. Thus the higher the frictional force between a particle and the screw surface the greater the probability that it will be carried over. Carry over is selective to large particles due to their size relative to the screw clearance and the greater momentum imparted to them. Their greater volume results in compression against the barrel wall and increase in friction against the screw surface. Table 3 shows the distribution of a bimodal size mixture of MCC pellets fed through the conveying section of the Haake extruder. Screw speed was constant at 200 rpm and mass feed rate increased to an arbitrary fill level where material discharge from the loading screw could be observed. The bulk of material is again conveyed by the driving screw. It can be seen that the material conveyed by the loading screw consists mainly of large particles (72% of material collected), illustrating the segregation effect and dependence on particle volume.

Table 3. Distribution of a 50/50 wt% blend of 1000 and 100 μm MCC pellets discharged from either screw in a long conveying section at 200 rpm.

Size segregation in conveying zones is a result of screw geometry. It occurs due to the higher probability of large volume particles to be carried over from the base to top of the driving screw. Material that is carried over following the rotation of the driving screw may additionally be segregated in transfer to the loading screws. Depending on the clearance between the screws fine particles may follow the rotation of the driving screw and fall through the gap between screws. Above the screws particles larger than the screw-screw clearance may become trapped between the flights of both screws and be conveyed forward with a rolling motion. The screw-screw clearance of the Haake extruder is 0.2 mm and thus larger than the size of the fine particles. This is likely to contribute to the segregation effect, forming a flow path which the coarse particles cannot take. However not all fine particles fall through this clearance, as implied by the fine material collected from the loading screw. The geometry of different extruders and their clearances varies between manufacturers. The clearance and relative particle sizes in any given system are likely to contribute to the degree of segregation.

Fill Level and Liquid Distribution

Figure 5 shows the size and normalised liquid distribution of granules produced by a single 60°F kneading block at low overall fill level. Liquid is non-uniformly distributed and skewed toward the top end of the granule size range. The bimodal size distribution is characteristic of twin screw granulation and becomes sharply monomodal at high L/S ratio [2,10,16]. Despite being consistently monomodal in size distribution the granules produced at high L/S ratio are typically too large to be used directly for tabletting. In addition liquid remains non-uniformly distributed at increasing L/S ratios. Granules produced at lower liquid to solid ratios are within a more useful size range, however the inherent heterogeneity indicated by the bimodal size distribution causes difficulties in downstream processing. The short 60°F mixing section in the screw configuration explored here is not adequate to ensure homogeneous liquid distribution. Table 4 shows the mass weighted coefficient of variance (CoV) in liquid distribution at various fill levels. At higher local fill levels at the site of liquid injection the variance in liquid distribution is slightly reduced. Changes in size distribution are minimal and remain bimodal however the distribution of liquid becomes more uniform at high fill level as reflected by the values of CoV in Table 4.

Figure 5. Size distribution and normalised liquid content of granules produced at 2 kg/h – 0.15 L/S – 400 rpm.

Table 4. Coefficients of Variance in liquid distribution of granules produced at increasing local fill level at the site of liquid injection.

The distribution of liquid at low and high overall fill level is shown in Figure 6. Liquid distribution is skewed at low fill level, however at high fill level is considerably more uniform across the entire range. Despite the improvement in liquid distribution at high fill the size distribution remains bimodal, with a greater proportion of fines. The low liquid variance would imply that despite the differences in size these granules are more similar in structure and properties. Thus high fill levels promote more homogeneous liquid distribution.

Figure 6. Size and liquid distributions of granules formed with a 1D 60°F kneading block at low and high overall fill level a) 1 kg/h – 0.18 L/S – 200 rpm, b) 4 kg/h – 0.18 L/S – 200 rpm.

Granules formed at low and high fill level are shown in Figure 7, black surfaces contain the nigrosin dye and are indicative of wetted areas. Granules produced at high fill are more rounded and have smoother surfaces, implying higher compaction of material. Granules produced at low fill contain elongated rectangular agglomerates and smaller granules can be seen to be fragments of these. It is believed these agglomerates are formed from the breakage of initial agglomerates formed during nucleation wetting. As granules are primarily fragments it can be inferred that the main role of the 60°F kneading block is breakage of these agglomerates. The rough surfaces of granules formed at low fill imply that the compaction in the kneading block is insufficient to cause significant densification and incorporation of fines. The low amount of mixing is not sufficient to ensure re-distribution of liquid. The surfaces of granules formed at high fill are lighter in colour showing the consolidation and incorporation of fines into the granule structure. It is believed that the higher fill level in the conveying zone promotes mixing of material and improves the distribution of liquid following nucleation.

Figure 7. Optical micrographs of granules formed at low (top) and high (bottom) overall fill a) & c) 710 – 1000 µm, b) & d) 1000 – 1400 µm.

4. DISCUSSION

Asymmetric Transverse Distribution

Asymmetric transverse distribution in conveying zones is inherent to the granulator geometry. Distribution of material between the screws is dependent on the fill level in the granulator. Raising the fill level in conveying zones improves distribution of material and induces mixing. Operating at higher fill levels will mitigate the inherent asymmetric distribution, however the higher mass loads in the granulator will affect granulation mechanisms and increases the risk of blockages occurring. This is implied by the average torque at steady state of 5.23 Nm at low fill level and 36.27 Nm at high fill level.

As some asymmetric transverse distribution is inherent to the process it can not necessarily be avoided, thus it is important to consider how it affects granulation mechanisms. Transverse loading will be uneven in that the driving screw carries more material than the loading screw. This means the driving screw does more work in conveying material, however the magnitude of this may be irrelevant when compared to the work carried out in mixing zones. A non-uniform transverse fill profile may establish in conveying zones leading into mixing elements. As the fill level rises in the approach to mixing zones, transverse distribution becomes more uniform, however for a non-cohesive material Figure 1 would imply uniform distribution is only established when the region is fully filled. The results of Lee et al [14] demonstrate that maximum fill is reached at the second kneading disc of the kneading block, but 100% fill level is only reached in this region with kneading blocks with 90° offset angle. Forwarding flow in kneading blocks is determined by a combination of kinetic and dispersive mechanisms, kinetic from the conveying action of the screw and dispersive following the axial fill profile of the block. If the transverse fill profile is non-uniform the kinetic force of the loading screw is lower and dispersive driven flow from the intermeshing region will have a greater tendency toward back flow. Given the differences in fill above and below the screws this may establish as "recirculation" of material between the upper and lower intermeshing regions of kneading discs. This may be why lower axial mixing is observed by Kumar et al [13] at higher fill levels, kinetic force is high and the degree of backflow is reduced. Visualisation of this flow pattern could be achieved through modelling or an in depth PEPT study.

Size segregation in conveying zones

The size selective segregation of material in conveying zones results from the asymmetric transverse distribution and again can be mitigated by operating at higher fill level. Wetting of material in conventional liquid injection leads to non-uniform liquid distribution where larger granules contain more liquid as in Figure 5. Given the characteristic bimodal size distributions of granules produced by TSG this means granules are heterogeneous in both size and properties. If the action of conveying elements segregates these different particles it is important to consider how this affects the granulation process. Segregation of particles may reduce the degree of mixing and distribution of liquid. Primary agglomerates are formed at the site of liquid injection under immersion type wetting [17], variance in initial wetting results in a mixture of large wetted agglomerates and dry fines. Segregation of these two types of particles may result in separate flow streams and reduce their interaction, meaning wet particles mix primarily with other wetted particles and dry fines with other fines. Redistribution of liquid is restricted as wetted material is isolated. The differences in particles may be alleviated in kneading blocks due to their superior mixing action, however conceptually segregation of particles may establish in flow of "wet" and "dry" streams fed into the top and base of the kneading block respectively. Thus good radial mixing would be required in order to ensure liquid distribution.

Segregation establishes in long conveying sections operating at a low fill level, thus will only be apparent at the discharge depending on the positioning of mixing zones and final fill profile. This has implications for the use of in-line particle sizing: if segregation is established at the discharge optical sizing techniques may result in skewed size data dependent on the position of the detector. To avoid this the separated streams of particles should be collated or the entire discharge flow sampled.

Size selective segregation is a fill level dependent effect of conveying zone asymmetry. The free volume under low fill conditions allows for segregation to take place, therefore segregation can be controlled through fill level. Segregation of particles may not necessarily have a strong effect on mixing mechanisms but is a property of twin screw granulation that should be taken into consideration when establishing screw configuration.

Fill Level and liquid Distribution

The distribution of liquid is poor at low fill levels. The asymmetric, plug flow which establishes in conveying zones results in poor mixing of wetted agglomerates and dry fines and increases the mixing load of the kneading block. During nucleation material undergoes immersion wetting. Increasing the local fill level at the site of liquid injection improves the liquid distribution during nucleation. When the fill level at the site of nucleation is higher the liquid is injected into a greater instantaneous volume of powder material. Liquid is better distributed throughout the greater volume of powder, where under low fill intensely wetted agglomerates would form. Similarly the transfer of material between conveying screws in this region mixes the newly wetted powder and aids distribution of liquid to un-wetted material. The higher the local fill level the better the distribution and the more uniform final liquid distribution. Additionally the longer residence time of material at higher local fill level may aid in minimising any effect of liquid feed pulsation. In this study liquid was fed via an 8 roller peristaltic pump, due to the nature of peristaltic pumps some pulsation in liquid is always present. Longer residence times dissipate the pulse of liquid allowing it to become more uniformly distributed throughout the powder. This would otherwise result in wet and dry plugs of material, demanding good axial mixing from the downstream kneading block to ensure distribution. The quality of primary nucleation wetting affects the downstream mixing demand.

Feeding liquid directly onto the kneading block led to periodic surging of material discharged from the granulator. Fill levels are highest across kneading blocks, given the improvement in liquid distribution found at higher fill level (Table 4) and the superior mixing action of kneading elements it would be expected this would result in superior dispersive mixing of liquid at nucleation. However the periodic surging of material arises due to the long residence times and poor conveying capacity of kneading blocks. Material in the block becomes intensely wetted changing the rheology and forming a viscous paste like substance resistant to forward flow. The pasted material forms a pseudo-blockage where dispersive forward flow is low. Sufficient kinetic force must be developed in the conveying zone upstream of the kneading block to clear the pseudo-blockage. Kinetic force increases as material accumulates in the conveying zone until it is sufficient to clear the pasted material, travelling as a plug of over-wetted lumps followed by a tail of fines. Once cleared, the pseudo-blockage will re-establish as the conveying fill level drops. Thus with this configuration the granulator never reaches true steady state. The material discharged does not contain true granules but instead a mixture of over-wetted lumps and ungranulated fines, with high variance.

Liquid distribution is most improved at high overall fill level as demonstrated by the differences in distribution in Figure 6. Despite the size distribution of granules remaining bimodal at high overall fill level the similar liquid distribution implies that granules across the size range are more uniform in structure and properties than those produced at low fill. There are three factors which lead to the improvement in liquid distribution, firstly the greater volume of powder gives more uniform immersion wetting during nucleation. Secondly maintaining the increased fill level throughout the conveying zone results in fully developed flow and induces mixing between the screws. Thirdly the higher overall mass load in the granulator increases compaction and consolidation of material. In addition to the mixing achieved in the conveying zone, primary agglomerates are restricted in size in the reduced free volume and are abraded through shear and collisions with other particles. Breakage is higher at high fill level indicated by the greater proportion of fines, however the surfaces of particles in Figure 7 indicate greater compaction and the lighter surface colouration shows consolidation and incorporation of fines into the granule structure. Granules can be seen to be fragments of large elongated granules implying the 60°F kneading block mainly causes breakage of primary agglomerates formed by nucleation wetting. The mixing ability of the kneading block is not sufficient to reincorporate material and promote growth through coalescence of granules. High fill levels increase consolidation and compaction of these granular fragments but optimisation of screw configuration is required to promote secondary growth of granules and overcome increased breakage rates. This is why proportionally more fines are present at high fill level despite the composition of granules being more uniform.

5. CONCLUSIONS

PEPT was successfully employed to validate measured transverse material asymmetry in the conveying zones of a twin screw granulator. Flow asymmetry is inherent to the granulator geometry but can be mitigated through control of fill level. Flow establishes as poorly mixed plugs of material travelling axially with minimal transverse mixing at low fill. Size selective segregation of particles occurs within conveying zones and further reduces the degree of mixing. The plug flow in conveying zones negatively affects liquid distribution at low fill level. Primary agglomerates formed by immersion wetting at nucleation travel as plugs and the mixing capabilities of 1D 60°F kneading blocks are insufficient to ensure redistribution of liquid. Raising the fill level at the site of nucleation reduces the variance in primary wetting. Maintaining high fill level throughout conveying zones induces mixing and improves liquid distribution. This work demonstrates that granule uniformity is dependent on wetting during nucleation. With 60°F kneading blocks the mechanism of formation is suggested to be primarily breakage in the mixing zone of large primary agglomerates formed during nucleation.

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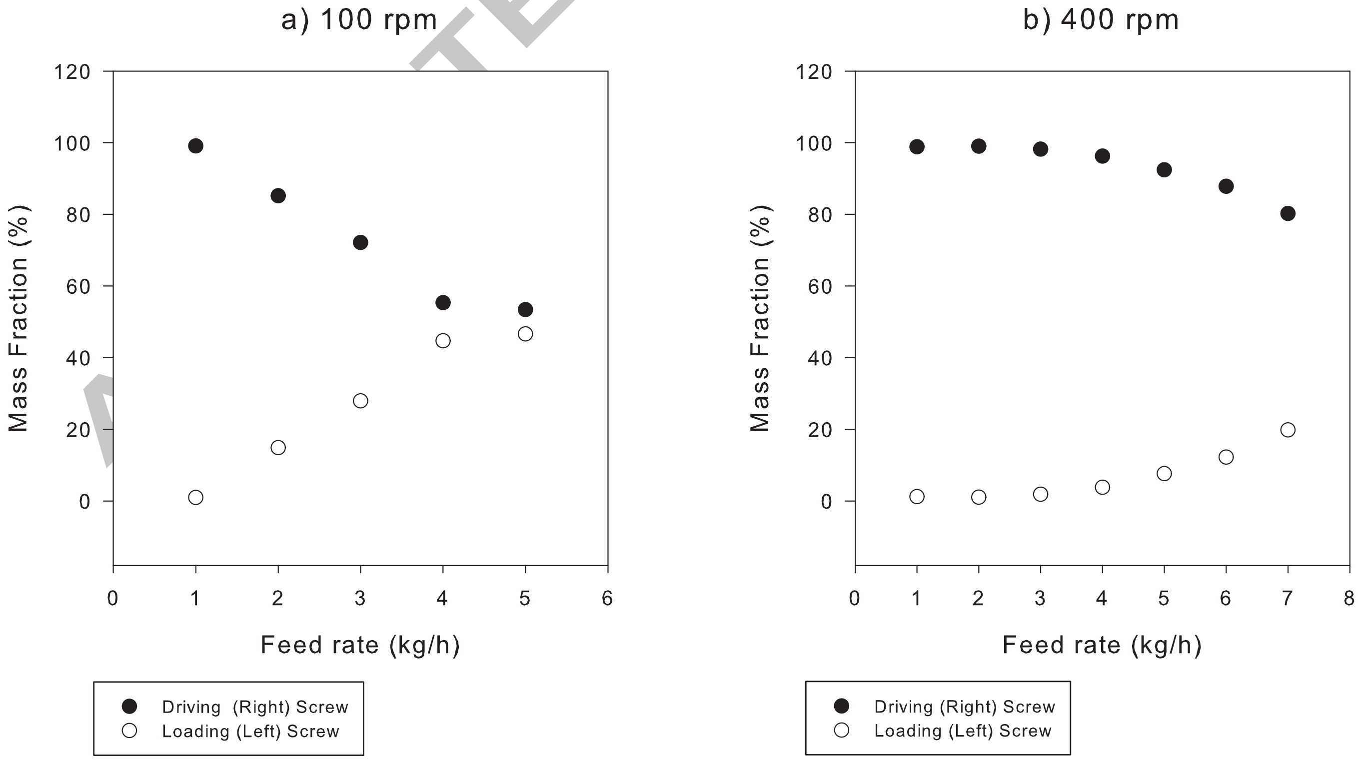
Figure1


a) 100 rpm
b) 400 rpm
Mass Fraction (%)
Feed rate (kg/h)
120
100
80
60
40
20
0
0
1
2
3
4
5
6
7
8
Driving (Right) Screw
Loading (Left) Screw

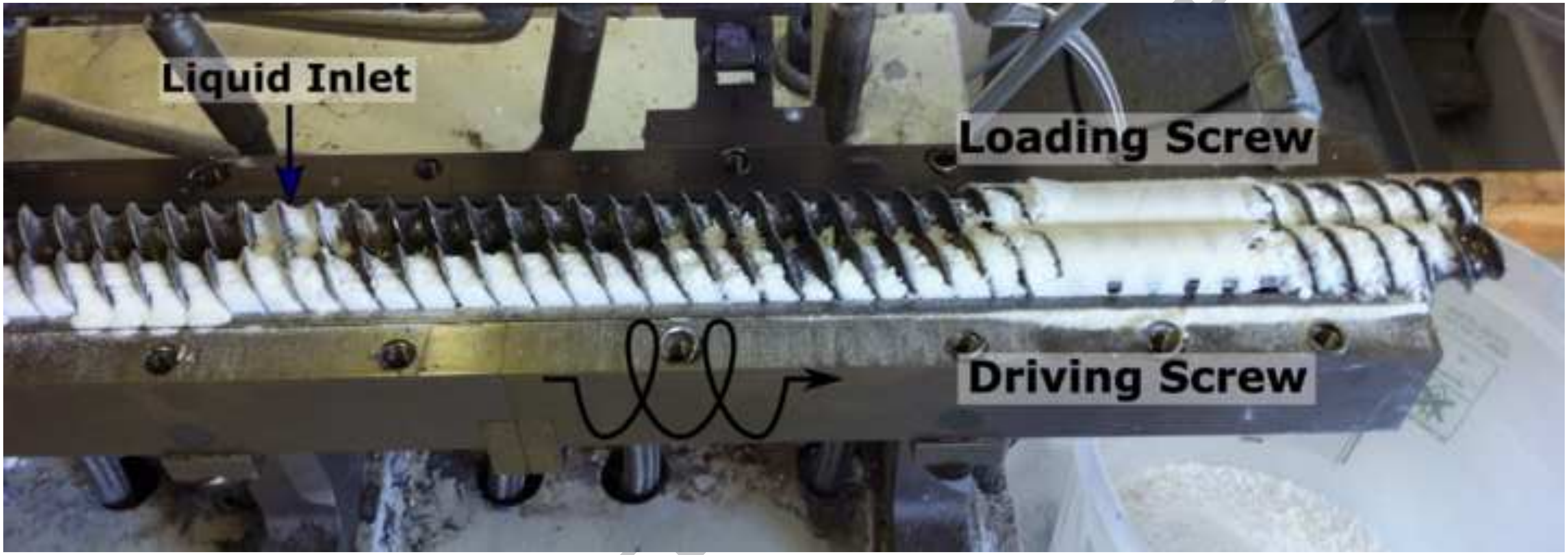

Liquid Inlet
Loading Screw
Driving Screw

Figure3a

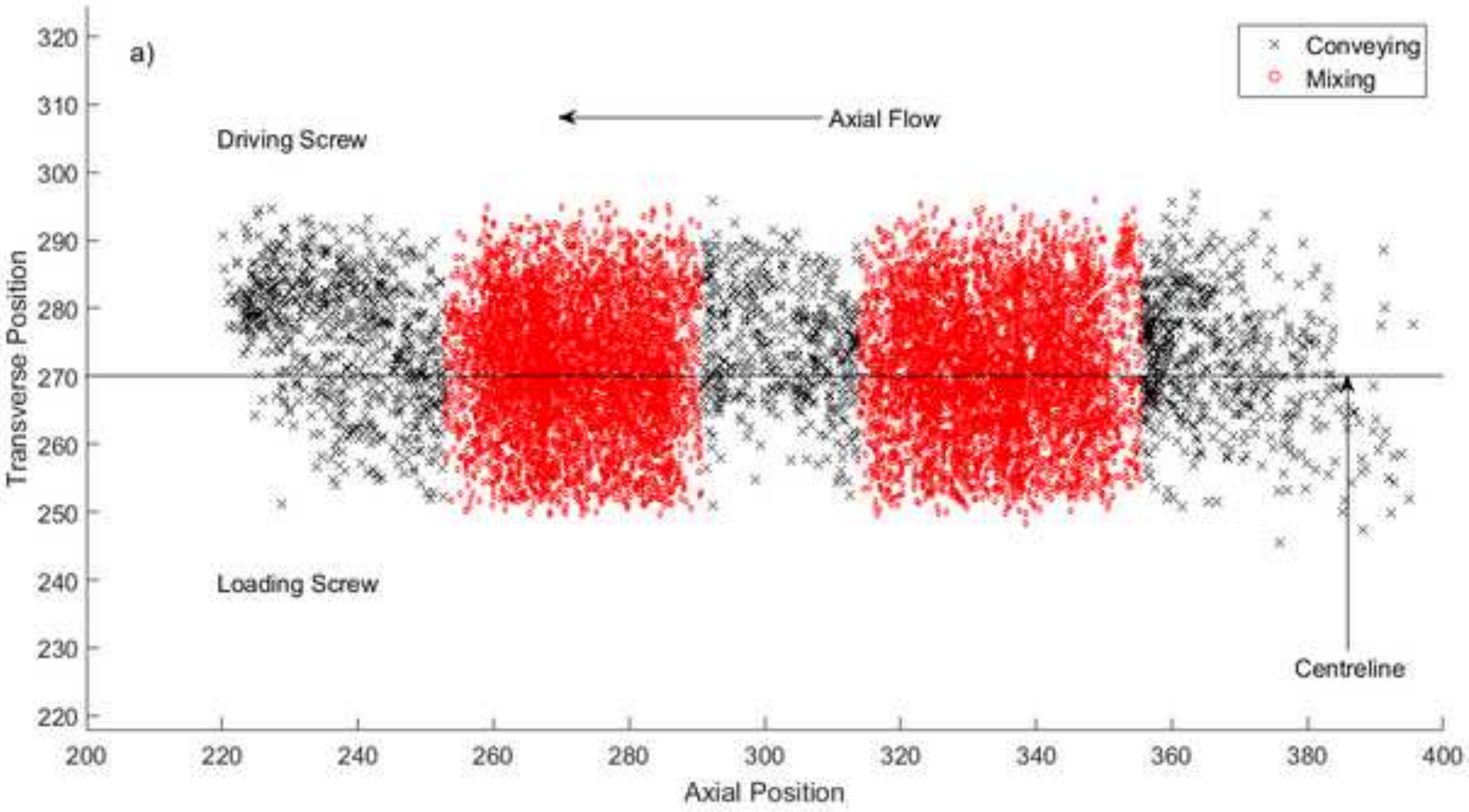

a)
× Conveying
○ Mixing
Axial Flow
Driving Screw
Loading Screw
Centreline
Transverse Position
Axial Position
320
310
300
290
280
270
260
250
240
230
220
200
220
240
260
280
300
320
340
360
380
400

Figure3b

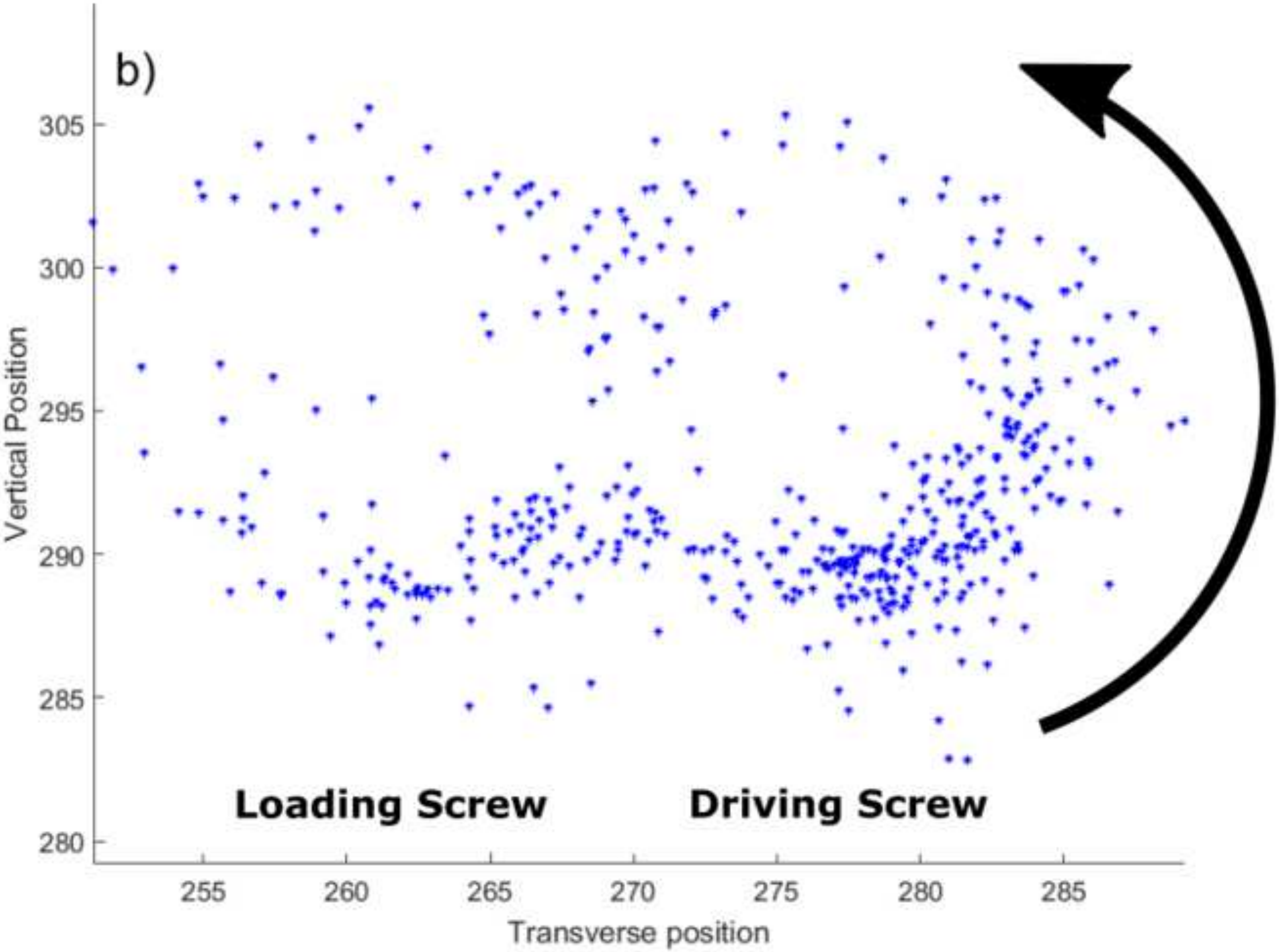

b)
305
300
295
290
285
280
Vertical Position
255
260
265
270
275
280
285
Transverse position
Loading Screw
Driving Screw

Figure3c

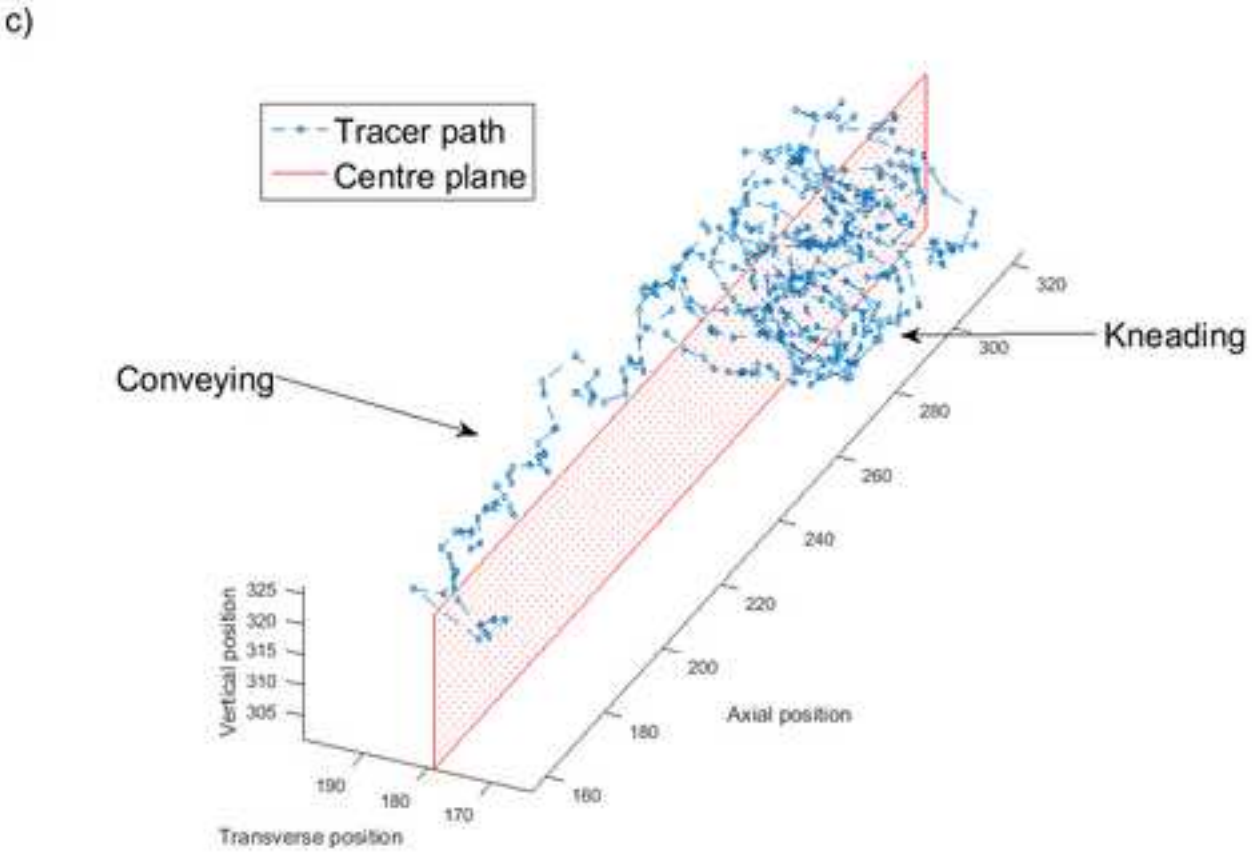
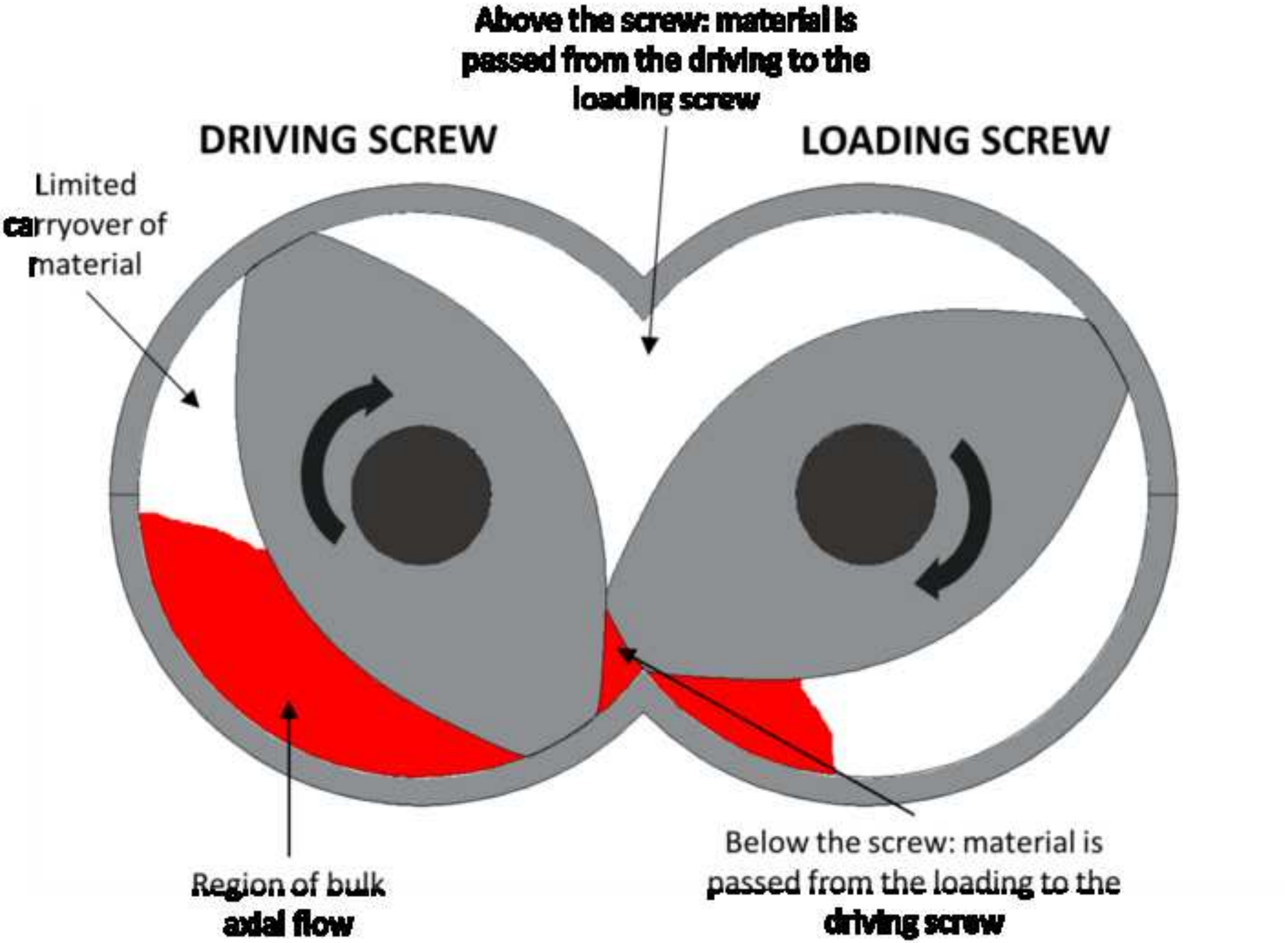
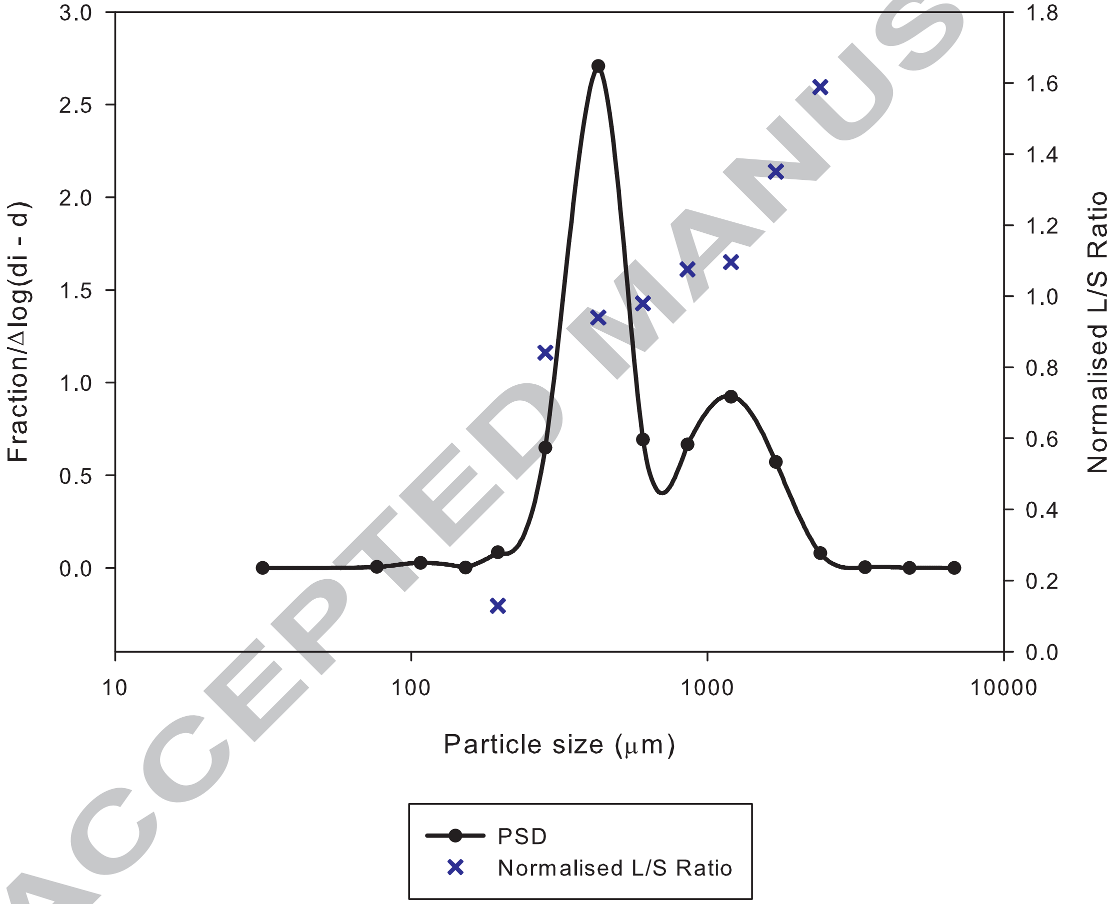
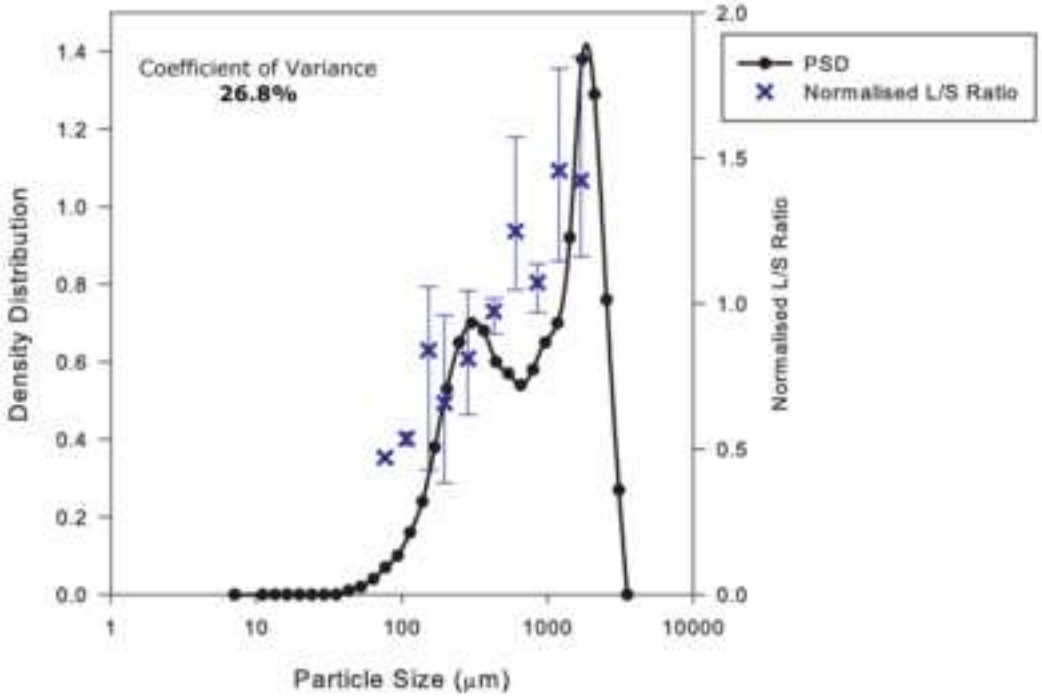

c)
Tracer path
Centre plane
Conveying
Kneading
Vertical position
325
320
315
310
305
Transverse position
190
180
170
Axial position
160
180
200
220
240
260
280
300
320

Figure4

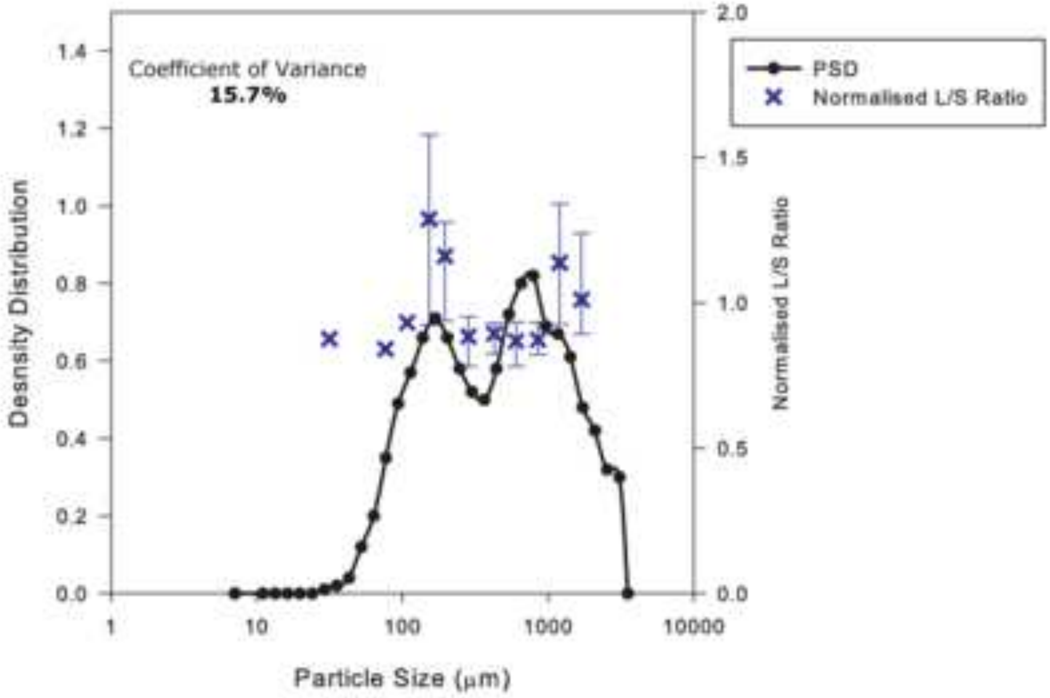

Above the screw: material is passed from the driving to the loading screw
DRIVING SCREW
LOADING SCREW
Limited carryover of material
Region of bulk axial flow
Below the screw: material is passed from the loading to the driving screw


3.0
2.5
2.0
1.5
1.0
0.5
0.0
Fraction/Δlog(di - d)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Normalised L/S Ratio
10
100
1000
10000
Particle size (μm)
PSD
Normalised L/S Ratio

a) Low Fill



b) High Fill



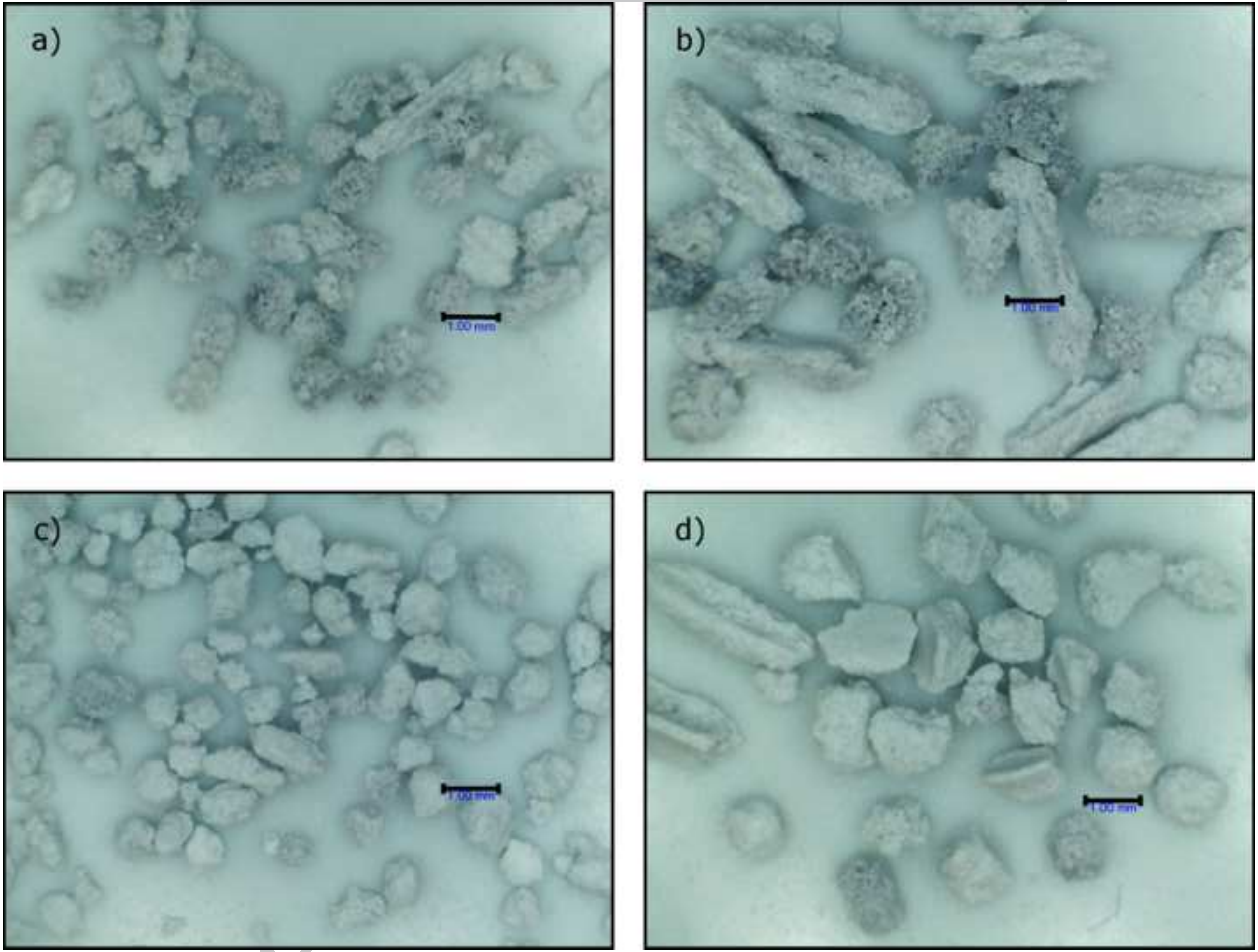

a)
1.00 mm
b)
1.00 mm
c)
1.00 mm
d)
1.00 mm

Table 1. Screw configurations explored in liquid distribution study.

Local Fill Level	**Material Feed Rate (kg/h)**	**Screw Speed (rpm)**	**Screw Configuration**
Unmodified	2	400	18x 1D **C**//4x 0.25D **K** 60°F//6x 1D **C**
Low	2	400	7x 1D **C**//3x 1D **C** (0.25D Pitch)//8x 1D **C**//4x 0.25D **K** 60°F//6x 1D **C**
Medium	2	400	7x1D **C**//4x 0.25D **K** 60°F//10x1D **C**//4x 0.25D **K** 60°F//6x 1D **C**
High	2	400	7x1D **C**//4x 0.25D **K** 90°//10x1D **C**//4x 0.25D **K** 60°F//6x 1D **C**
Overall Fill Level			
Low	1	200	18x 1D **C**//4x 0.25D **K** 60°F//6x 1D **C**
High	4	200	18x 1D **C**//4x 0.25D **K** 60°F//6x 1D **C**

Table 2. Sum of time spent by the PEPT tracer in each zone of the granulator.

	Total Time (min)		Time in conveying zones (s)		
Configuration	Mixing	Conveying	Loading Screw	Driving Screw	Percentage of Time - Driving Screw (%)
10g/min - 150rpm					
30°F KB	21:45	4:18	93.6	164.3	64
60°F KB	28:24	4:13	92.7	159.9	63
90° KB	25:40	3:24	47.4	157.1	77
10g/min - 300rpm					
30°F KB	12:42	2:18	59.6	78.5	57
60°F KB	17:27	1:42	34.2	67.8	66
90° KB	17:09	1:27	39.6	47.0	54
20g/min - 300rpm					
30°F KB	9:27	1:36	38.3	58.0	60
60°F KB	14:19	2:43	53.4	109.2	67
90° KB	20:22	1:48	51.5	56.4	52

Table 3. Distribution of a 50/50 wt% blend of 1000 and 100 µm MCC pellets discharged from either screw in a long conveying section at 200 rpm.

	Mass load Conveyed % (gram)	
	Driving Screw	Loading Screw
Total Material Conveyed (100g)	78	22
Coarse Particles (1000 µm)	45 (35.1g)	72 (15.8g)
Fine Particles (100 µm)	55 (42.9g)	28 (6.2g)

Table 4. Coefficients of Variance in liquid distribution of granules produced at increasing local fill level at the site of liquid injection.

Local Fill Level	Liquid Distribution Coefficient of Variance (%)
Unmodified – No element	32.2
Low – 0.25D pitch conveying element	33.4
Medium – 1D 60°F Kneading block	30.5
High – 1D 90° Kneading block	26.2

Graphical abstract