An Experimental Study on Free-surface Rolling Segregation and Correlations with Angle of Repose and Particle Sphericity

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Bulk solids commonly segregate when particles move on an inclined surface, often causing significant variations in content uniformity of particle properties such as size and species. Often called free surface rolling segregation, this is one of the most common segregation phenomena for powders and granules, where different sized and shaped particles become separated due to differential movement on the surface. Intensity of rolling segregation is widely understood to depend upon various factors including differences in particle size and shape as well as flow properties and densities. These influences have been studied and reported previously, but correlations between levels of rolling segregation and particle shape assessed by particle sphericity or sphericity ratio with consideration of angle of repose has not been investigated in detail.

This is an experimental study of rolling segregation of one particulate material (calcium carbonate) with different particle size ranges; using a single material eliminates variations of influences not being studied, especially solid density. The influence of bulk and particle properties has been studied systematically, especially particle size distributions, angle of repose and particle sphericity. The segregation results show an increased trend with higher particle sphericity and especially sphericity ratio. Also, the results indicate higher levels of segregation with a low angle of repose, but little segregation when angle of repose is higher than 32 degrees. However, the authors caution that this angle of repose of 32 degrees should not be taken as a general indicator of a limitation of the susceptibility of blends to free-surface rolling segregation, for materials other than the one tested.

Key words: Rolling Segregation; Particle Shape; Sphericity; Sphericity Ratio; Angle of Repose

1 Introduction

Segregation of solid particulates in terms of size, shape or density is a phenomenon that leads to significant variations of particle size, shape and density in different parts of a batch being processed, compromising the original homogeneity [1]. This segregation can cause many problems in industrial processes. For example, in pharmaceutical and food industries, significant variation of particle size distributions (PSDs) and heterogeneity of blend components in material handling processes leads to compromised quality of final product eliciting failures during quality control [2]. In order to control segregation of powders by either changing formulations of materials or process methods, extensive work has been carried out both theoretically and experimentally [3, 4-5].

Free-surface rolling segregation is one of the most common segregation phenomena, happening when solid particles drop on an inclined surface and roll down an angle of repose.

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Different sized and shaped particles segregate as a result of different trajectories on the surface, caused by different friction opposing their momentum [6]. It is frequently observed in handling of particulate solids especially during filling and discharging of material into and out of a vessel or process [7]. This rolling segregation has attracted considerable attention in the past due to its significant influences on process performance and final product quality. For instance, significant change of particle sizes leads to variation of a critical component (typically an active pharmaceutical ingredient (API) or flavouring), and failure in blending processes, with a focus on free-flowing materials [2]. Considerable research has been done to better understand the mechanisms and the factors affecting rolling segregation using numerical simulations [8, 9], which mainly focus on particle size and density without considering particle shape or bulk flow properties, except for a few studies [1].

However, it is known from industrial practice that in influencing rolling segregation, particle shape and bulk flow properties can be just as important and certainly in cases can overcome the influences of particle size and density. Anecdotally, many formulators express the strategy of reducing segregation susceptibility of their blends by introducing components that make them cohesive (*i.e.* prevent them from being free flowing) [10]. Experimental studies of the current authors for industrial clients (unpublished due to confidentiality) have shown that with highly engineered particles such as pet-food kibbles, careful selection of particle shapes in certain size ranges can be used to oppose the natural segregation caused by the different sizes. However, in spite of these known industry practices, the influences of particle shape and flow properties have never been objectively quantified in research. Therefore, it was felt necessary to investigate rigorously the influence of particle shape and particle flow properties on rolling segregation, in relation to the known effects of particle size, to aid formulators to use these techniques more effectively to design blends that are more resistant to free-surface rolling segregation.

In this study, an experimental investigation of the effects of the particle shape and angle of repose on free surface rolling segregation has been conducted for calcium carbonate powders with different particle size and size distributions, and the effect of particle sphericity and angle of repose on segregation are discussed.

2 Surface rolling segregation

Free-surface rolling segregation was reported during heap formation many years ago [11-13]. It typically happens when particulates are loaded into a bin or stockpile and then discharged, as shown in Fig. 1(a). A short overview of rolling segregation and the quantification of its severity as a "segregation index" are given below.

2.1 State of arts

In surface rolling segregation, typically different sized particles come to rest preferentially in different locations of the surcharge or heap, due to different movement of the larger and smaller particles. Most commonly, the top of the heap comprises more fine particles whereas coarser particles tend to move further down towards the bottom. The size variation in the heap can result in significant change of particle size distributions ("PSDs") in sub-lots when the material is divided into small batches (in packs, tablets etc) in a following process [12].

Sometimes, the changes of PSDs can be crucial, *e.g.*, in pharmaceutical industries where blends are generally composed of coarse particles as excipients and fines as active pharmaceutical ingredients (APIs). Variation of fines causes variation of API ("content uniformity") compromising the quality of final products [2].

According to Brown [14], rolling segregation was described as particles moving with different velocities due to different levels of friction opposing their momentum, where the form of the heap surface was approximately constant. However, the study by Fan, et al. [8] suggested that particle movement during heap formation can be affected by not just particle dynamics (such as velocities), but also the nature of the surface of the heap and the properties of the solids.

Tang and Puri [13] classified all segregation mechanisms into four categories of trajectory, sieving (or sifting), fluidisation and agglomeration segregation in general. For rolling segregation, trajectory and sifting segregation are commonly observed as the major mechanisms rather than fluidisation and agglomeration segregation. Specifically, in the moving layer descending the surface of a heap, sifting allows the fine particles to fall through the coarse ones to stratify the sizes in the moving layer, then the finer particles experience more friction against the underlying stationary surface, holding back their movement compared to the coarser ones. As pointed out, the influence factors on levels of rolling segregation can be particle dynamics (initial kinetic energy), particle properties (size, shape, friction), and the formed surface properties (see Fig. 1(b)) [8, 9].
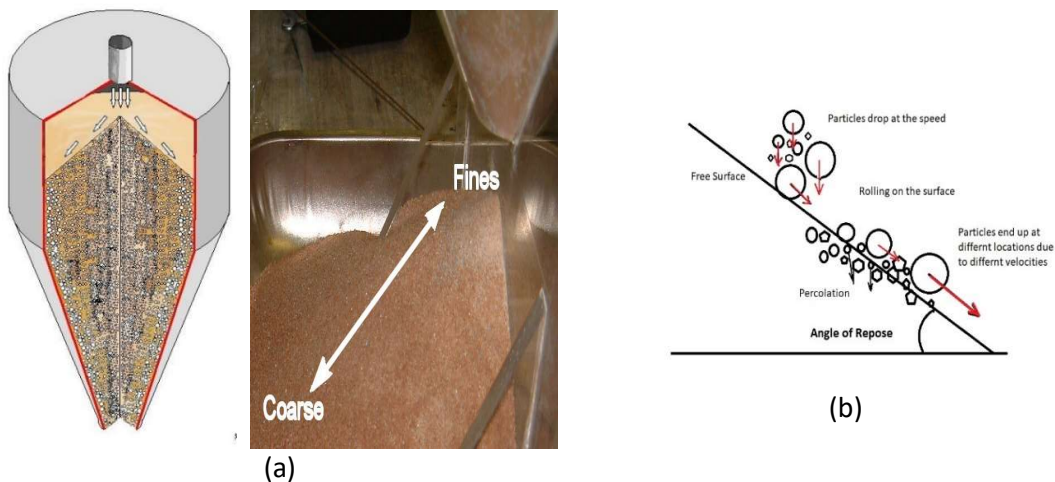

Fig. 1: (a) Particles segregate in a bin during filling, (B) Particles rolling on an inclined surface with segregation mechanisms

In the past, significant research focused on the dynamic influences, which were promoted by mathematical modelling of segregation [8, 9]. Fan et al. [8] summarised the kinematic influences and applications in continuum models of heap segregation and pointed out that further work would need to consider particle properties such as particle shape and cohesion. On the other hand, Liss et al. [6] asserted that the particle movement would be controlled by ratios of friction between particles to particle momentum gained from the initial velocities.

With his idea, surface segregation directly relates to material physical properties (size, shape, friction) and heap surface formation. From these studies, it is evident that rolling segregation can be subject to both initial particle movement and the physical properties of the particles.

In mechanical principles, large particles can have high initial kinetic energy and momentum, but low friction even on a rough surface due to rolling effects (it is a matter of common experience that a football will roll much more readily on a stony beach then will a tennis ball). Hence large particles have high possibility to move further down compared to small particles. To quantify the particle movement on a heap, three things need to be considered: initial kinetic energy, momentum of particles, and friction [31]. The initial kinetic energy has been used for many studies in relation to mass conservation and velocity change in flowing layers [4, 8, 15, 16]. As reviewed [1, 8, 17], it is difficult to characterize the kinematics of particles in rolling segregation if the material physical properties are unknown. A general conclusion from these works is that the physical properties of the material are key to any quantifying study of surface rolling segregation based on mechanisms, dynamics and kinematics.

Influences of material properties on surface rolling segregation of granular materials have been studied, and particle size and density have been especially investigated [17–19]. Effects of shape and/or surface roughness of particles have also been explored by other researchers [20]. The studies concluded that particle size, shape and density had influences on segregation levels of powders. The difference in density, and sometimes particle shape had a greater effect on particle segregation than the difference in particle size. Remy et al. [21] explored the effect of particle roughness of cohesionless glass beads and particles of various roughness with different friction coefficients. They found that the amplitude of the velocity fluctuation of components increased as particle surface roughness was increased. Another complexity for rolling segregation is surface formation, which also depends on material properties. In bounded heap flow, the percolation and angle of repose effects are the two major factors that determine the final particle segregation configurations [8, 16]. Due to the percolation mechanism in the moving layer, smaller particles have more contact with the heap surface tending to retard them, but also have higher possibility of getting caught in a texture feature on the surface compared with bigger particles. So, surface formation is important in assessment and prediction of surface rolling segregation, where particle shape and cohesiveness need to be considered even if percolation is not involved [20].

Consequently, the current study focuses on rolling segregation influenced by particle shape and cohesion, while the kinematic influences are kept constant at a low level by fixing a falling height that is small compared to the length of the sliding surface.

2.2   Segregation index and links to particle properties

For rolling segregation, many numerical simulation methods have been developed including continuum approaches [8, 22]. A continuum approach has some advantages in its applicability and accuracy in predicting the final particle configuration. However, complexity of running simulation models limits predicting segregation in practical industrial situations. Instead, segregation indices may provide a simple indicator for levels of segregation.

A segregation index is a dimensionless number, which incorporates the concentrations of the species of interest in both homogeneously blended and segregated powder, to quantify the change. Many segregation indices introduced previously [23-25] are based on a comparison between a statistical quantifier such as variance for the concentration of a reference component and the variance for the measured sample, such as Lacey index [26]. Zigan et al., [23] introduced a segregation index with consideration of mass ratios when a scaling-up problem of segregation in different dimension silos was discussed. In case of a single material, Zigan's segregation index can be as a ratio of concentrations of fines before and after segregation at the size i.

$$I_{s(i)} = \left[\frac{C_i - C_{o(i)}}{C_{o(i)}}\right]^2 \qquad (1)$$

where C_i is the (mass or volumetric) concentration of fines after segregation at the size i and $C_{o(i)}$ the (mass or volumetric) concentration of fines in original homogeneous blend at the size i. The purpose of the square here appears to be to ensure that the result always comes out as a positive number, however this has the unfortunate effect of giving disproportionately larger numbers to larger deviations and vice versa. In industrial practice, most quality control (QC) operations are concerned with the proportionate variation from the intended content of a component (API tolerance is often ±10% of the nominal value), and also recognise that it is useful to understand whether a variation is in the upwards or downwards direction in any give sub-lot, so this is better informed by calculating the change of the concentration without the square power, as follows:-

$$P_{s(i)} = \left(\frac{C_i - C_{o(i)}}{C_{o(i)}}\right) \qquad (2)$$

This will give a positive or negative number according to whether a blend is "over-strength" or "under-strength". These segregation indices are simply a statistical representation of concentration of a component in a process, without inherent links to any physical influences of particle or process properties. Zigan *et al.* [23] attempted to establish a group of dimensionless numbers for individual physical factors to correlate its contributions in the segregation, which is shown in Eq. 3.

$$I_s = f(\rho_a, E, d, S \dots) = f(f_{air}, f_s, f_{ES} \dots) \qquad (3)$$

where ρ_a is air density, E is air extraction rate, d is particle diameter and S is solid feeding rate, and the corresponding physical factors: f_{air} is the air flow factor, f_s is the factor of particle size and silo dimension and f_{ES} is the factor of feeding rate and air flowrate. Note that Zigan was studying air-induced segregation, which is excluded from this study, hence the importance of air properties to him; nevertheless, this is presented here to illustrate a general approach to solving the problem of predicting segregation, which correlates the segregation index with the influence from individual physical parameters so the levels of segregation under different conditions could be better evaluated. Therefore, for rolling segregation, Eq. 3 may become Eq. 4, which the factors can be in different forms: dynamic factor, f_v (moving velocity difference), frictional factor, f_{rr} (cohesiveness as indicated by angle of repose), particle size

factor, f_s (size and size difference), particle shape factor, f_c (shape factor and difference in this between components) and f_{cc} (flow functions).

$$I_s = f(f_v, f_{rr}, f_s, f_c, f_{cc} \dots)$$ (4)

In Eq. 4, particle shape can be considered as an independent factor, which has been used in many numerical simulations [20, 27-28] but not been applied in segregation index. As shown in a recent work, the shape factor was considered by using definition of parameter aspect ratio (AR) [29]. Nevertheless, the shape factor used in this paper applies the same definition for solution of f_c, which is described by the term of particle sphericity, S_c. However, in this paper the suitability of S_c in assessing rolling segregation with segregation index is also studied.

In 2D dimensions, sphericity of a particle is also known as circularity, which is the degree to which the particle is similar to a circle, taking into consideration of the smoothness of the perimeter. It accounts for both the particle overall form and roughness. Thus, the further away from a perfectly round, smooth circle a particle becomes, the lower the sphericity value. With definition in ISO 9276-6 [29], the sphericity can be defined as:

$$S_c = \sqrt{\frac{4\pi A}{P^2}}$$ (5)

where P is the perimeter of a particle, and A is the projected two-dimensional area. In this study, the authors were particularly interested in how differences in particle shape drive segregation or difference , so using particle sphericity from Eq. 5, the shape difference factor, f_c could also be a ratio of the sphericity of a segregated component to the average particle sphericity of the homogeneous blend:

$$f_c = \frac{S_{c(i)}}{S_{cv(i)}}$$ (6)

where $S_{c(i)}$ is the particle sphericity of segregated particles at size i, and $S_{cv(i)}$ is the particle sphericity of the virgin materials at the same size i.

3 Experimental procedure

To quantify influences of particle shape and cohesiveness on surface rolling segregation, this experimental study has included segregation tests on a system of different bulk solids of the same chemistry and production route from a common feedstock, ground to different degrees to yield different degrees of cohesiveness (indicated by angle of repose) and particle properties including PSDs and sphericity.

3.1 Segregation tester

It must be recognised that particulate segregation, and therefore the segregation properties of any bulk solid, do not exist in isolation from the processes that cause segregation, but are the result of interaction of the material properties and the process. The same bulk solid may segregate more in one process than in another. Therefore, when quantifying the segregation tendency of a bulk solid it is important to define clearly the process to which this quantified tendency relates, so that it may be used as a reference case against which to judge other cases. A method has been chosen that has been shown clearly to represent (in a scalable

manner) a scenario that is especially common in industrial processing, that of material rolling down an angle of repose where the initial kinetic energy of the particles hitting the surface is not sufficient to be influential (specifically, not sufficient to cause the particles to exhibit large bounces on the heap surface that cause them to lose contact with substantial parts of the length of the surface).

To this end, surface rolling segregation tests were carried out on a segregation tester (QPM) developed at the Wolfson Centre, University of Greenwich [30] as shown in Fig. 2 (a). The tester is used to quantify the intensity and distribution of segregation in a heap formation, where particle segregation is noticed due to rolling (including percolation) mechanisms. The advantage of this tester is that the results are accurately scalable to different heap lengths [30], making it easy and convenient to use the results to predict segregation in industrial processes using this tester, providing the same mechanisms dominate.

The QPM segregation tester consists of a cubic mixer and an adjustable inclined trough (Fig. 2 (a)). The mixer can be run at a controlled speed for a defined time period, for example, 25 rpm and 30 minutes for the current tests to ensure the powder is uniformly blended. The blend is discharged into the trough which is set to the angle of repose to make a smooth and continuous heap of powder, which has a segregation pattern and is ready for sampling. Six equally sized compartments can be formed by sliding gates as shown in Fig. 2 (b). Each section can be discharged individually and the sample material in the compartment can be collected for further analysis. The section numbers of 1-6 is set for the samples from top to bottom of the trough. The trough is approximately 380 mm long and cross-section is about 55 mm wide by 55 mm high. A sample of approximately 0.5 litre bulk material is used for a test.
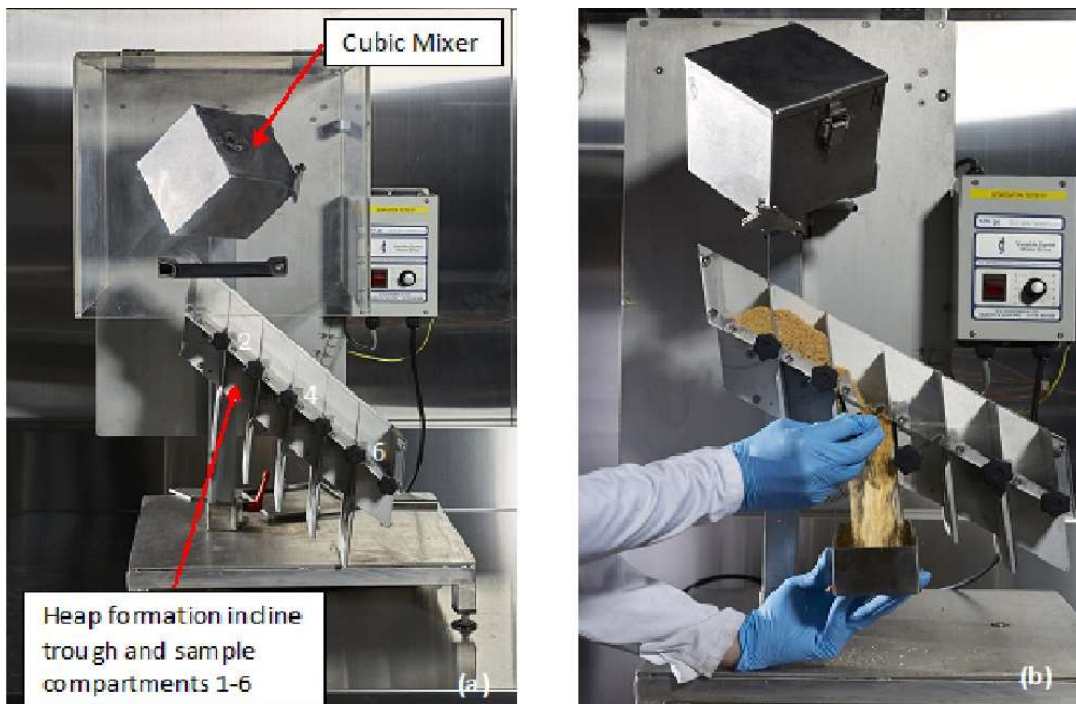

Fig. 2: Surface segregation tester (QPM) for heap formation: (a) Tester; (b) Sampling

3.2   Test method

The segregation investigation was carried out by using the QPM segregation tester shown above. The test materials (about 0.5 litre) were blended in the cubic mixer initially to homogenous distribution of particle sizes. The blended materials were sampled first for virgin material property determination including PSDs before discharging into the test trough. Then the rest of the material in the mixer was discharged to form a heap in the trough. The heap material was sectioned by sliding gates shown in Fig. 2. The sectioned samples were collected in sampler for further particle size analysis. Note that once filled, section 1 is to the left hand side of the apex of the fill so represents a complete heap length of small size whereas sections 2, 3, 4, 5 and 6 are to the right hand side of the heap apex therefore dividing a complete heap length into 5 parts. The samples in top (section 2), middle (section 4) and bottom (section 6) compartments were sub-divided into small proportions of samples and 3 of the small samples were used for the particle size measurement by using Malvern Mastersizer2000. Average and standard deviation of the measurements were used for results analysis of the segregation tests.

3.3   Test materials and material properties

In this study, Calcium Carbonate with several size ranges was used for the study of particle shape influence, with consideration of particle surface roughness or friction that is assessed by the angle of repose. A list of the test materials (named Eskal series manufactured by KSL Staubtechnik GMBH, Germany) and materials properties is given in Table 1, which includes median sizes (D50) and size span, solid density (measured using Pycnometer) and bulk density, angle of repose (measured by a 20 mm diameter funnel raised slowly to form a standard cone) for the original samples and particle sphericity measured by using Malvern G3.

Table 1: Test materials and material physical properties

Name of materials	Median Size D_{50} (µm)	Size Span $(D_{90}-D_{10})/D_{50}$	Solid Density (kg/m^3)	Bulk Density (kg/m^3)	Angle of Repose (°)	Sphericity (-)
Eskal 10	10.3	1.19	2800	1254	41.5° ±0.3	0.882
Eskal 15	18.2	0.93	2800	1262	34.6° ±0.4	0.875
Eskal 30	27.0	1.29	2800	1354	32.5° ±0.5	0.883
Eskal 80	80.6	0.91	2800	1483	30.8° ±0.3	0.885
Eskal 150	128.7	0.65	2800	1489	27.6° ±0.3	0.885
Eskal 0.1-0.5	346.4	2.27	2800	1496	29.1° ±0.6	0.881
Eskal 0.5-0.8	594.9	0.42	2800	1502	28.6° ±0.5	0.881

Particle size distributions (PSDs) of the blended virgin powders were measured by Malvern Mastersizer2000 and the averaged PSDs are shown in Fig. 3. The Eskal materials are manufactured in the same process from a single feedstock which has high chemical uniformity and classified for different size categories based on different applications. In this study, all powders are named with their original commercial names (e.g. Eskal 10), which are named according to their size categories. Six of the materials tested (Eskal 10 - Eskal 150 and Eskal 0.5-0.8) show a relatively narrow size range with different median sizes. The Eskal 0.1-0.5 has a relatively wide size range.
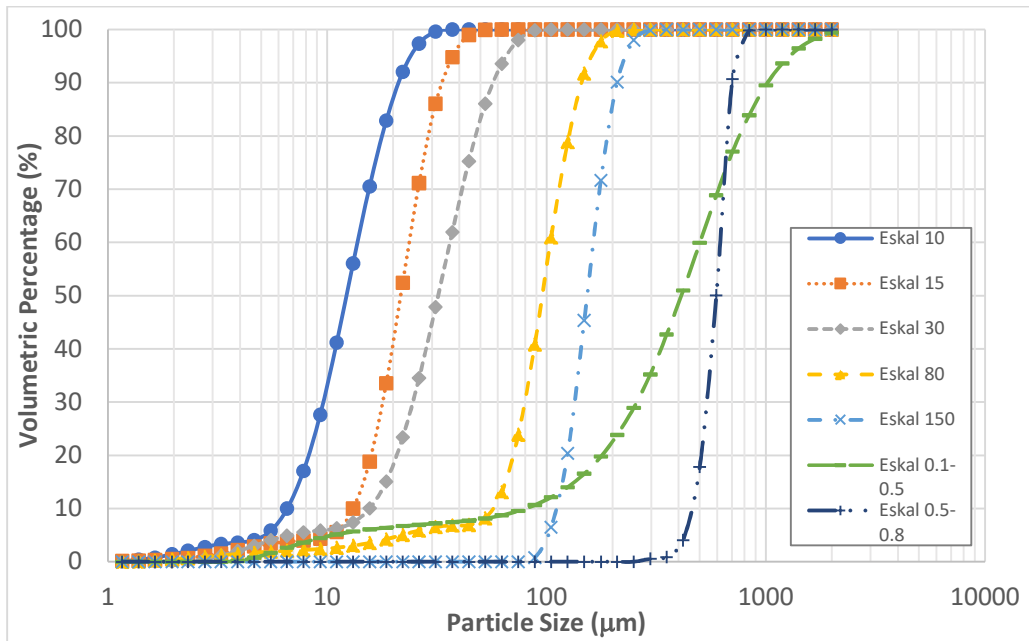
Fig. 3: Particle size distributions (PSDs) for the 7 Calcium Carbonate test materials

SEM images of the particles for 4 materials (Eskal10, 80, 150 and 0.5-0.8) are given in Fig. 4 (a-d), which show that particle shapes for different sizes of particles are quite similar.
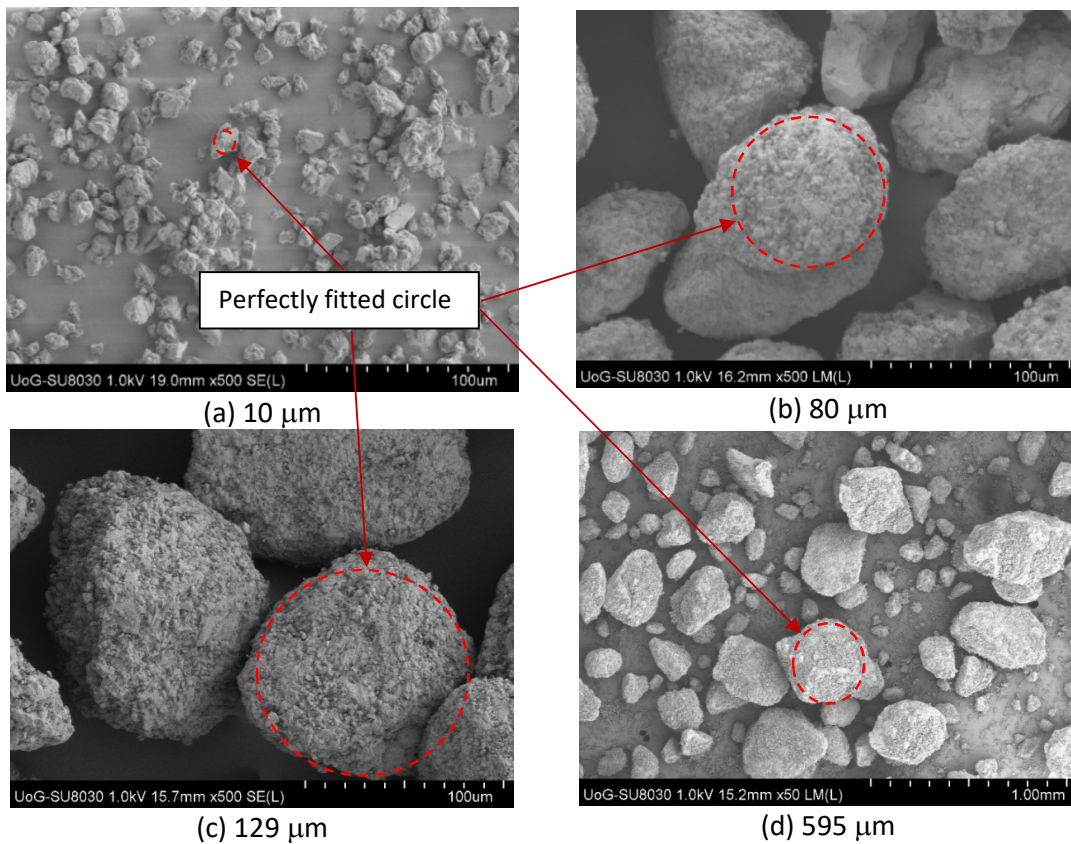

Fig. 4: SEM images of particles for the 4 different sizes of Calcium Carbonate with best fitted circles: (a) Eskal 10; (b) Eskal 80; (c) Eskal 150 and (d) Eskal 0.5-0.8 (in different scale).

Also, it demonstrates that the shape factor being measured is the ratio of the projected two-dimensional area to the perimeter of the particle shown in the images. It can be found that the particles are likely to have high roundness for all the size with rough surfaces, and large particles tend to be coated with fines if there are fines present.

3.4   Variations in cohesiveness (flowability) and use of angle of repose as an indicator

The different grades of calcium carbonate covered a range of cohesiveness, above 80 microns the materials were visibly observable as being free flowing whereas smaller sizes were increasingly cohesive. It is common for most dry bulk materials to be free flowing above a certain critical particle size then increasingly cohesive below this. This trend shows up in the angle of repose for Eskal, being broadly similar from 80 microns upwards and increasing below this. Nevertheless the authors caution the reader against using angle of repose generally as an indicator of the cohesiveness of a bulk solid; there are many other factors apart from cohesiveness that influence angle of repose, and in spite of it being quite commonly cited as an indicator of flowability, experience shows that it can be extremely misleading to link the two except within certain close limits. One such set of limits, applicable in this case, is that all the bulk solids are of the same material made in the same way, so within the limits of this study the angle of repose did give a useful indicator of flowability.

4   Results and discussion

4.1   Angle of repose and particle size

Segregation index, I_s, calculated from Eq. 1 and volumetric concentration change of fines, P_s, calculated from Eq. 2 at the top section of the segregation tester as a function of angle of repose are depicted in Fig. 5. The I_s and P_s are calculated by the concentrations at the median particle size (D_{50}) for all tests. The top section is selected for the figure just because the P_s are positive, which means percentage of fine contents is increased.
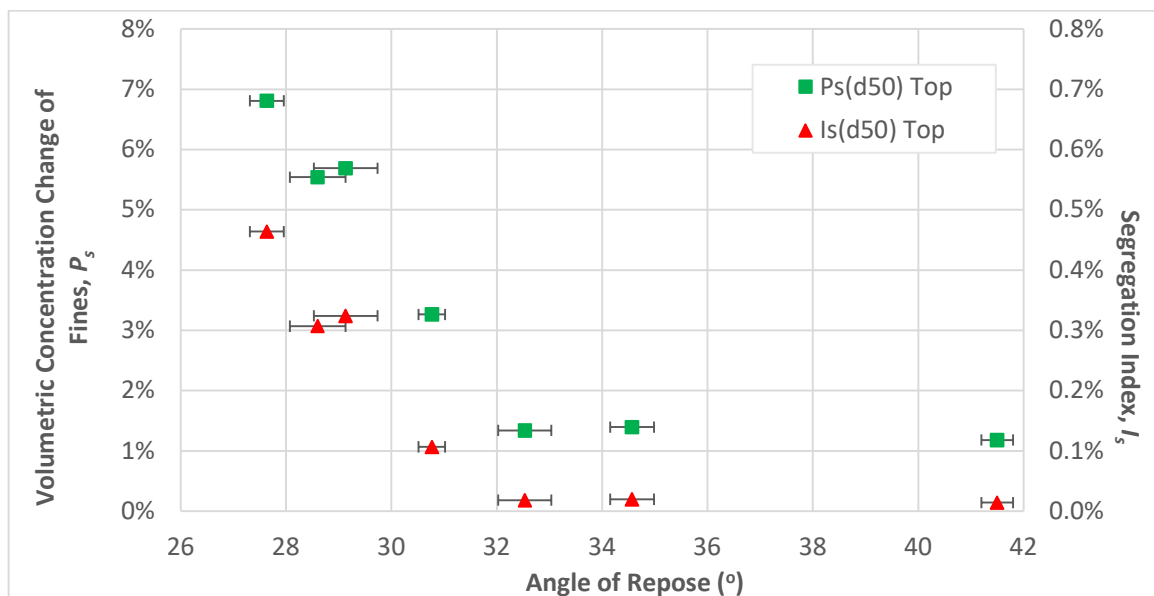

Fig. 5: Segregation index I_s and volumetric concentration change P_s at D_{50} for the samples in the top section versus angle of repose

The results of segregation index (I_s) and the change of volumetric concentration change of fines (P_s) at D50 clearly show a sharp increase, when the angle of repose is smaller than 32° for the materials tested. For the test materials, the angle of repose is larger than 32°, it means the powder is cohesive where segregation is consistently low as shown, but it shows more or less a linear increase below 32°. This angle of repose, 32° is thought that is strongly related to particle contact forces such as cohesiveness and contact areas, and particle shape. With different materials, the material may have different cohesion (contact force) and variation of particle shape with the same size distribution, which must result in a different specific angle of repose for segregation. So 32° is thought only for the material tested in this study.

Comparison of segregation indices in term of median particle sizes is shown in Fig. 6. Segregation (P_s) was low with median particle size of 40 μm and below, but increased from there to about 150 μm, and above which it remained similar.
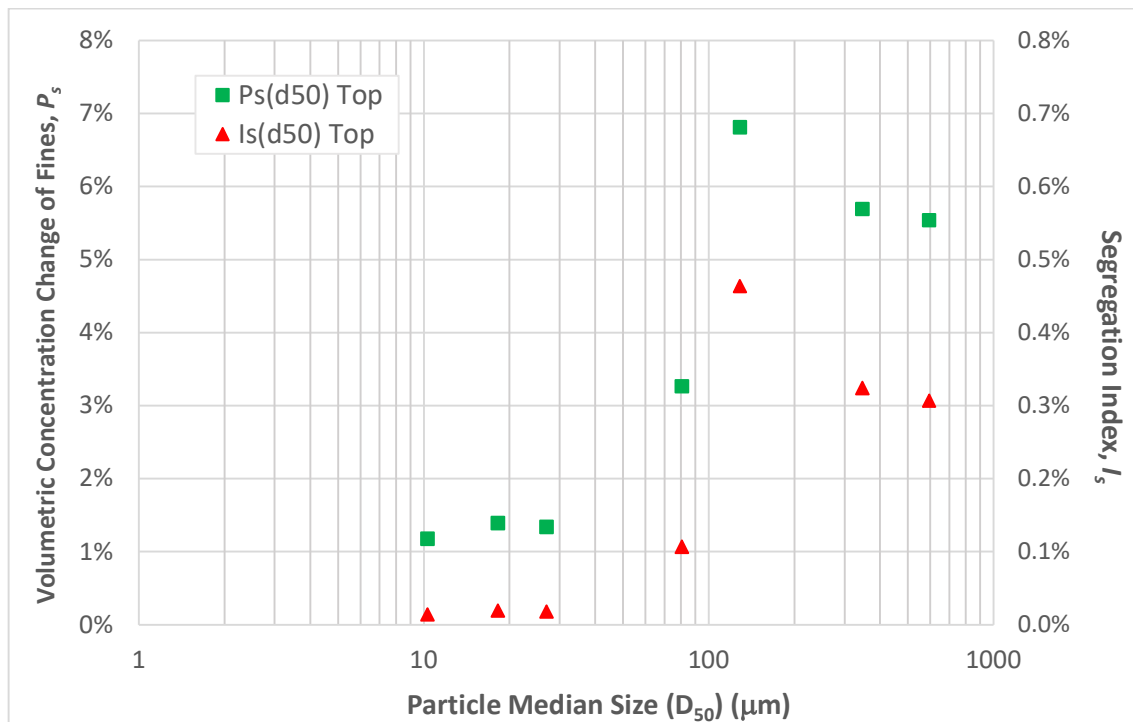

Fig. 6: Segregation index, I_s and volumetric concentration change, P_s at D$_{50}$ for the samples in the top section versus median particle size, D$_{50}$

The results in Fig. 5 and 6 shows that samples with bigger particle size and less cohesion will have more tendency to segregate. Inspection of these two figures also showed a boundary beyond which the samples undergo little segregation. Full segregation results of concentration change of fines (P_s) at D$_{50}$ for the top, middle and bottom section versus angle of repose are given in Fig. 7, and versus particle median size in Fig. 8, where the P_s was selected is because the concentration change can give out direction of the component change as the fines are increased as a positive value or decreased as a negative value.

The data shows a slight difference in the trend between segregation observed at the top and bottom of the heap. First, the material at the top showed increasing segregation when the

AOR fell below 32.5 degrees whereas material at the bottom showed increasing segregation when the AOR fell below 34.5 degrees. Second, the segregation at the heap bottom was generally higher than at the top, right across the range. This seems not to be simply experimental variation, as the results are aggregated across several tests which were highly repeatable, so these seem to be real trends. It could be that segregation at the top of the heap is slightly attenuated compared to further down, because the material near the top has had less opportunity for the fines to percolate (sift) downwards though the coarse particles.
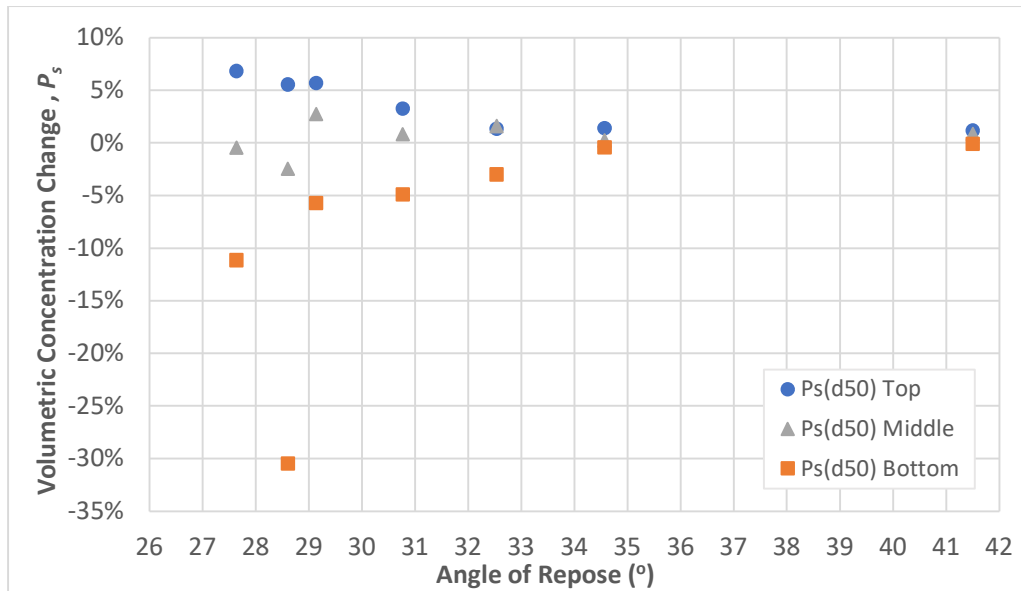

Fig. 7: Volumetric concentration change of fines for particle size of D_{50} in different test sections of an inclined slope (top, middle and bottom section) versus angle of repose
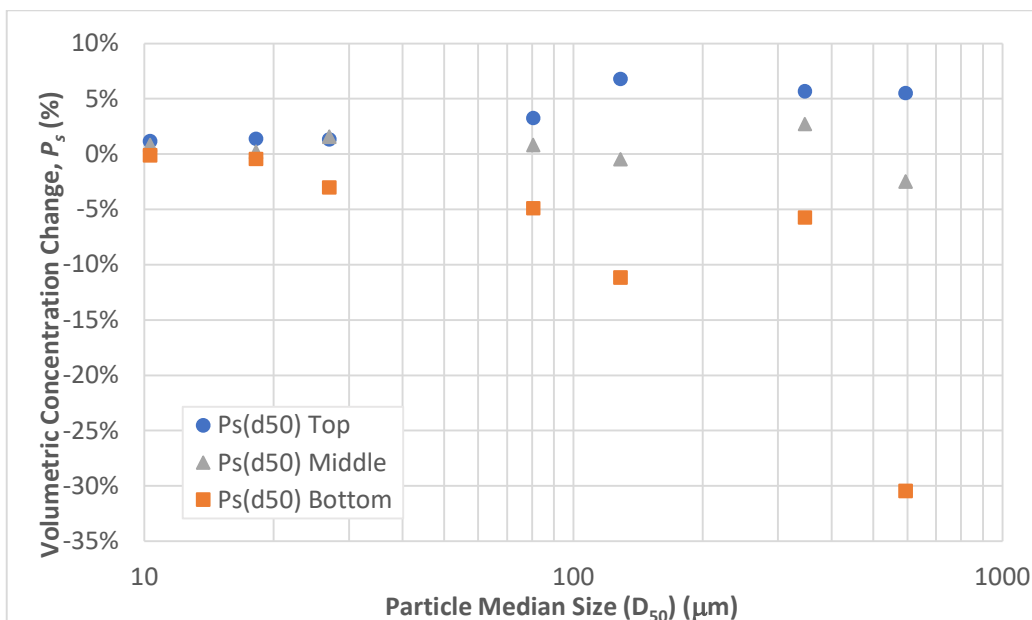

Fig. 8: Volumetric concentration change of fines for particle size of D_{50} in different test sections of an inclined slope (top, middle and bottom section) versus particle median size

The results in Fig. 7 clearly show a segregation trend with angle of repose that the material with low angle of repose is expected to have large segregation on an inclined surface. Volumetric concentration of fines (P_s) at D_{50} in top sections changes positively, which means the percentage of fines for the same particle size is increased compared to that in the original material. With increased angle of repose, the change of fines in top sections is decreasing. For bottom sections, the results in Fig. 7 show opposite changes, which negative changes of volumetric concentration of fines means coarse contents increased. In middle sections, the changes are minor. There is a similar cut-off for angle of repose at 32-34°. In Fig. 8, the same data have been plotted against particle median size, showing increased segregation with particle size. It is noticed that the segregation is very small for particles smaller than 30 ⬚m. It was considered that particle size range (as well as median sizes) may have an influence on particle segregation, so the segregation data against the size spans (Table1) have been presented in Fig. 9. The results here show some interesting points: 1) it confirms a previous study [7] that a material with a large size span (a wide range or a small D_{50}) is expected to have large segregation, 2) however, it shows that a material with a small size span (a narrow range or a large D_{50}) is also expected to have strong segregation, 3) only a material with the size span around 1 to 1.2 is expected to be less segregated. It reveals that not only a wide size range, but also a very narrow size range will result in large segregation due to containing some extra-large particles (causes a large D_{50} or a wide size range).
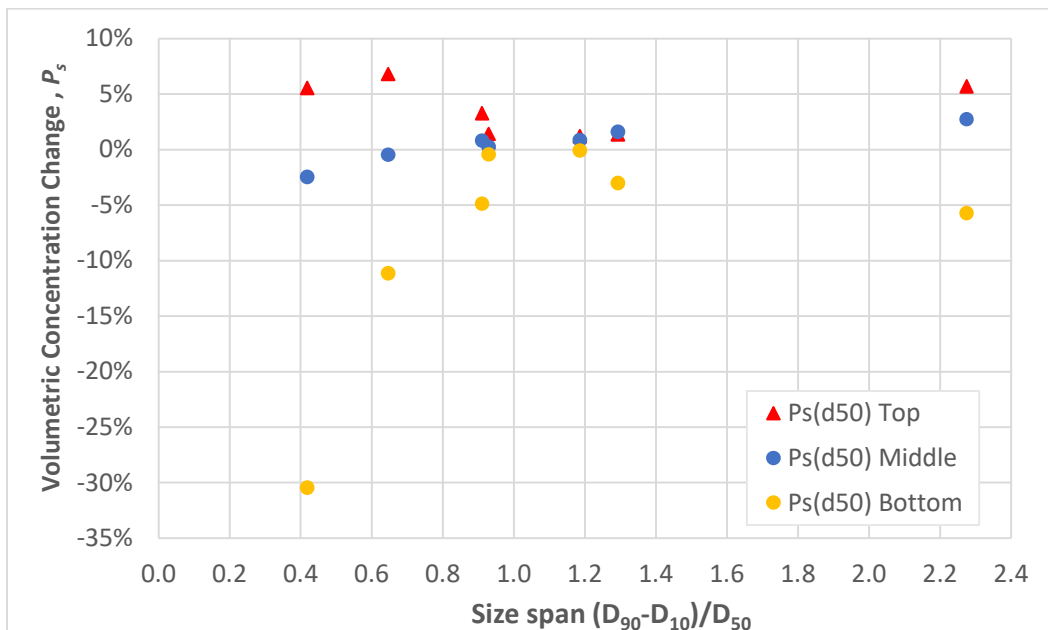

Fig. 9: Volumetric concentration change of fines for particle size at D_{50} in different test sections (top, middle and bottom section) of an inclined slope versus particle size span

4.2   Particle shape and sphericity

In rolling segregation, particle shape can make significant contributions if the particles are rounded. However, it is hard to correlate the segregation with particle shape quantitively. In this study, particle sphericity calculated by Eq. 5 for the materials in different segregation test

sections has been determined and the results of particle sphericity for the materials in different segregation test sections are given in Fig. 10.
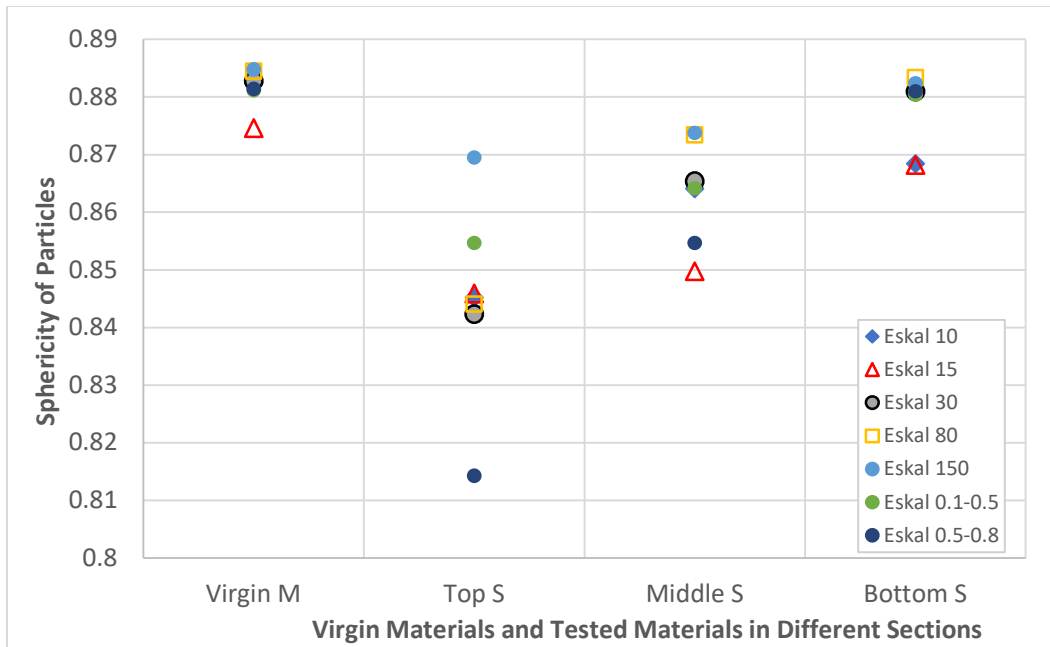

Fig. 10: Sphericity of segregated particles in different test sections (top, middle and bottom section) and particle sphericity of the virgin materials

The results in Fig. 10 show that sphericities of the particles in sections are clearly different. In bottom sections, the particles generally have a high sphericity (0.87-0.88), which is close to the values for virgin materials. In top section where contains more fine particles, the particles seem to have a relatively low sphericity (0.81-0.87). The sphericity for the particles in middle section lies in between (0.85-0.875). The sphericity results seem to show that larger particles tend to be more spherical and small particles are more angular. However, the sphericity for the virgin materials shows a fair similarity for all different particle sizes. Therefore, the results can reveal that the particles with different shapes are segregated due to rolling effects.

By physics, it is known that spheres must suffer from a low friction if the spheres are rolling. In terms of volumetric concentration changes shown in Fig. 11, the level of segregation can be heavily driven by particle shape when the particle sphericity is close to 1, which is perfectly spherical. Also, when the particles become more angular, it shows the level of segregation has not changed significantly. For the top section where P_s increased, a low sphericity means particles are more angular. For the bottom section where P_s decreased, the particles tend to have a high sphericity. Generally, it reveals in rolling segregation that coarse particles can be driven by both the size and the shape compared to fine particles. The results in the figure show that level of segregation at the bottom (5%-10%) is generally higher than that in top sections. Therefore, for rolling segregation particle shape has a strong influence when the particles have a high sphericity. The results confirm that particles with high sphericity tend to move to the bottom and angular particles have a high chance to remain at the top.
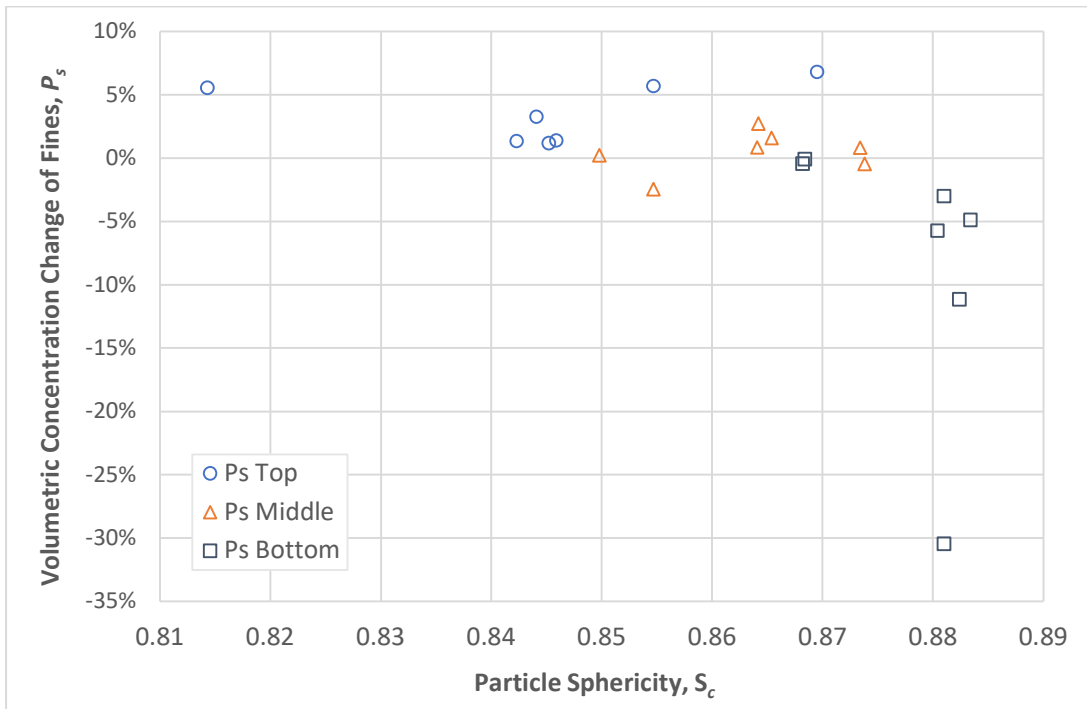
Fig. 11: Volumetric concentration changes of fines at the median particle size (D_{50}) for the materials at different test sections versus particle sphericity measured of the samples

4.3   Shape factor and sphericity ratio

For surface rolling segregation, the experimental results obtained have shown that particle shape can be significant to drive segregation when the particles are highly close to spherical. Especially for coarse spherical particles, levels of segregation can be enlarged by the spherical particle shape compared to any angular particles. However, although the influence of particle shape on rolling segregation is clear, it is hard to establish a proper correlation between segregation tendency of a powder and a particle shape factor which would represent the particle shape influence. Generally, spherical particles will have high segregation tendency due to a small rolling friction whatever the particle size is. when the particles become angular, segregation of particles will purely depend on friction between the particles when rolling effect is negligible. By the results in Fig. 11, particle sphericity may not reflect variation of particle shape and its influence in segregation. Therefore, particle sphericity ratio in Eq. 6, is used for assessing the variation. The results of particle sphericity ratio, fc, are shown in Tables 2 and the corresponding particle sphericity measured are also given.

The sphericity ratios in the table clearly show the difference between the top and bottom sections, where the particles in bottom section have the same sphericity to the virgin materials (f_c=1.00) and that the particles in the top section are quite different (f_c=0.96 in average). A low sphericity ratio means that the particles are more angular (less spherical) than the particles in virgin materials. So, in the top section more angular particles are presented and in bottom section the particle shape remains as no change.

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Table 2: The sphericity and the sphericity ratios for the materials at different test compartments (Top, Middle and Bottom) compared to the sphericity of virgin materials

Test Materials	D_{50} (µm)	AoR	S_c Virgin	S_c Top	S_c Mid.	S_c Bot.	f_c Top	f_c Mid.	f_c Bot.
Eskal 10	10.3	41.5	0.8823	0.8452	0.8641	0.8684	0.958	0.979	0.984
Eskal 15	18.2	34.6	0.8746	0.8459	0.8498	0.8682	0.967	0.972	0.993
Eskal 30	27.0	32.5	0.8828	0.8423	0.8654	0.8810	0.954	0.980	0.998
Eskal 80	80.6	30.8	0.8845	0.8441	0.8734	0.8834	0.954	0.987	0.999
Eskal 150	128.7	27.6	0.8848	0.8695	0.8738	0.8824	0.983	0.988	0.997
Eskal 0.1-0.5	346.4	29.1	0.8812	0.8547	0.8642	0.8804	0.970	0.981	0.999
Eskal 0.5-0.8	594.9	28.6	0.8814	0.8143	0.8547	0.8810	0.924	0.970	1.000

With the data in Table 2, segregation index I_s at particle size of D_{50} for different test sections (Top, Middle and Bottom) versus particle sphericity ratio, f_c is shown in Fig. 12. The results show that highly segregated particles have the same particle shape as the particle sphericity ratio is close to 1. Clearly the particles in the bottom section show an exponential relationship between the sphericity ratio the segregation index, but for other two sections the relationships are not very clear. Therefore, although the results show that the particles with different shapes behave differently in rolling segregation, the sphericity ratios only indicate the shape differences in the segregation. To have a clear picture of particle segregation versus the sphericity ratio, the results of the particle sphericity ratios for particles in different test sections against the volumetric concentration change of fines, P_s, are shown in Fig. 13.
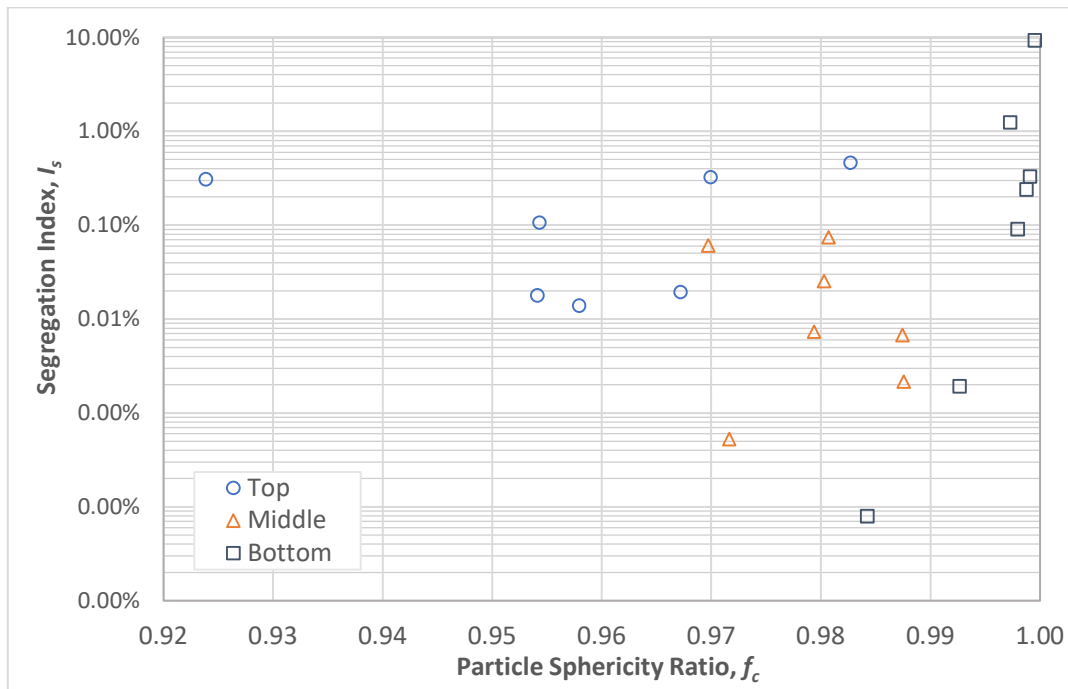

Fig. 12: Segregation index I_s at particle size of D_{50} versus particle sphericity ratios for particles in different test sections (Top, Middle and Bottom) of the test materials
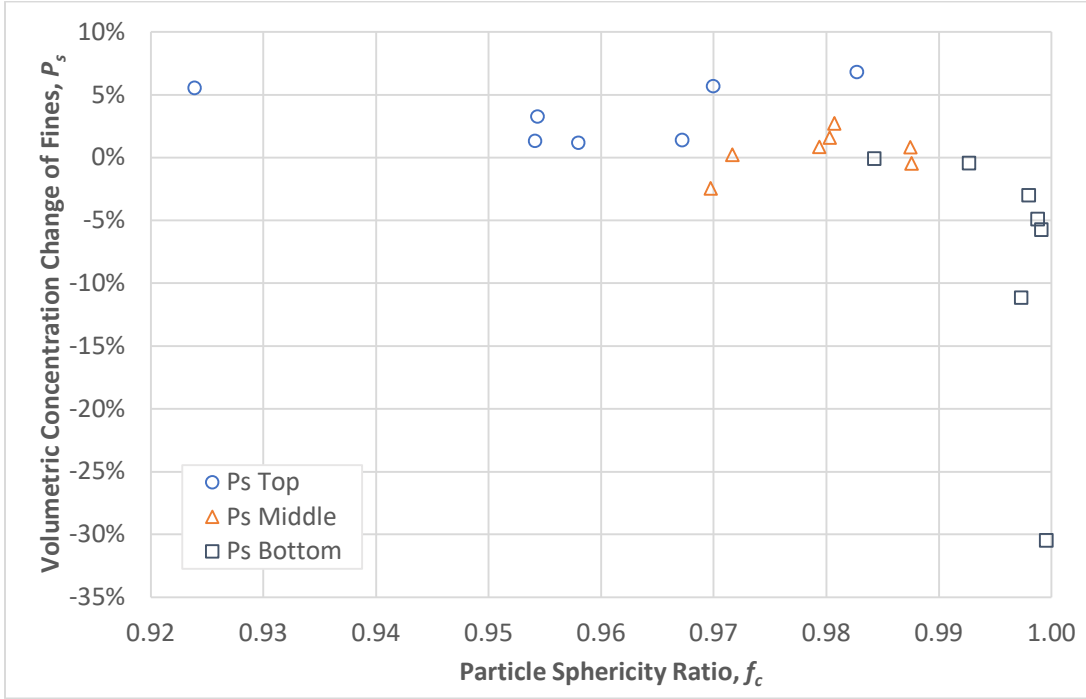
Fig. 13: Volumetric concentration change of fines, P_s at particle size of D_{50} versus particle sphericity in different test sections (Top, Middle and Bottom) for the test materials

In Fig. 13, it shows a relatively high change of particle shape for the particles in top section, where the fines content has increased (as the sphericity ratio, f_c is getting small). For the particles in bottom section, the particles seem to have much similar particle shape whereas the sphericity ratio is close to 1. Interestingly, the particles with higher similarity of particle shape (the f_c is close to 1) have high segregation where the particle size tends to be bigger. Inversely, the particles with high fines contents tend to be more angular (the sphericity is getting small) and the shape is more different to that in virgin materials. In terms of particle sphericity change magnitude in rolling segregation, the variation of particle sphericity, R_{sc} can be defined in Eq. 7, which could be an indicator for the particle shape influence.

$$R_{sc} = \left| \frac{S_{cv(i)} - S_{c(i)}}{S_{cv(i)}} \times 100\% \right| \qquad (7)$$

where $S_{c(i)}$ is the particle sphericity of segregated particles at size i, and $S_{cv(i)}$ is the particle sphericity of the virgin materials at the same size i.

In Fig. 14, the variations of particle sphericity for the segregated particles in all test sections versus the volumetric concentration change of fines are given. It shows an increased trend when the variation increases. A best-fit trendline is given. Although the trend seems weak, it clearly shows an increased fines content when the variation of particle sphericity is increased. It can conclude that spherical particles (shape variation is close to zero) tend to segregate much more than any angular particles. When the particles become more angular, the influence of particle shape in rolling segregation tend to disappearing.
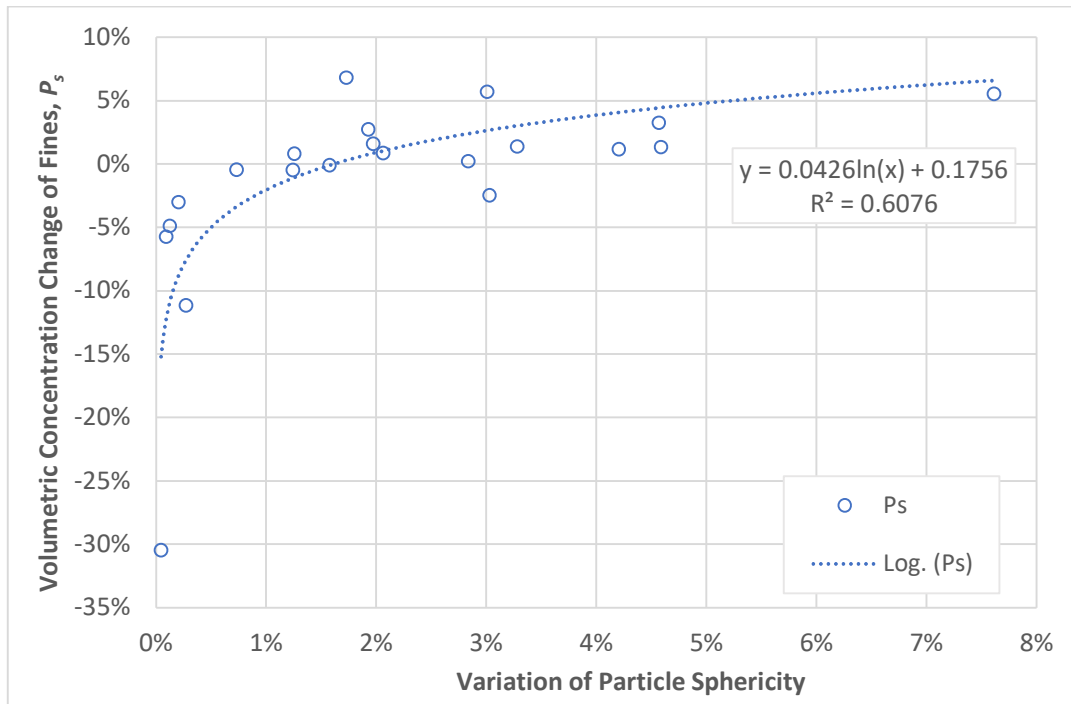
The chart shows the equation:
$$y = 0.0426\ln(x) + 0.1756$$
$$R^2 = 0.6076$$

Fig. 14: Volumetric concentration change of fines, P_s at particle size of D_{50} versus variation of particle sphericity for the test materials in all segregation test sections

5   Conclusions

In this study, influences of particle shape and angle of repose on surface rolling segregation have been studied by incorporating segregation index and volumetric concentration change of fines for one powder with different size and size ranges. It is concluded that:

1) For surface rolling segregation, particle shape and flow property (represented by angle of repose) have significant influences in case of low AORs and more spherical shape.

2) It shows that there is a critical value of AORs for segregations of a material (e.g., 32o for the material tested), above it the material is less susceptible to segregate, but lower than that the level of segregations can be linearly proportioned to its angle of repose.

3) Particle size can strongly influence segregation levels in rolling effects. The materials with large particle size tend to segregate more. However, there is a low limit of particle size, beyond that the material undergoes little segregation (e.g. 30 ?m for the material tested). A large size span will give high segregation, but a material with a small size pan can be more sensitive to the segregation. A size span of (1-1.2) gives less segregation.

4) For rolling segregation, influence of particle shape only becomes strong when particle sphericity is high. The results confirm that particles with different shapes are segregated due to the rolling effects. The particles with high sphericity are likely to move towards the bottom of a heap and angular particles have a high chance to remain near the top.

5) Particle sphericity ratio and variation of particle sphericity were studied as indictors for the particle shape factor. The results show that It is hard to establish a correlation between the segregation index, but they may work for the volumetric concentration change of fines.

6) The results show a general decrease of fines contents in rolling segregation when the sphericity ratio increases. The particles with higher similarity of particle shape (the fc is close to 1) have high segregation where the particle size tends to be bigger. The variation of particle sphericity also shows the same trend to indicate particle shape influence in rolling segregation.

7) It is concluded that spherical particles (shape variation is close to zero) tend to segregate much more than any angular particles. When the particles become more angular, the influence of particle shape in rolling segregation tend to disappearing

8) Therefore, variation of particle shapes in a material can be a driven factor as the same as particle shapes in sphericity.

In a summary, particle flow and particle shape can have strong influences on rolling segregation of a powder and need to be considered as important factors. Angle of repose can indicate the influence of powder flow. For shape influences, sphericities and variation of the sphericities of a powder need to be studied.

Acknowledgement

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Table A1: Segregation index and concentration change for the different components (top, middle and bottom) and the different size category of (D10, D50 and D90)

Test Materials		Particle size (μm)				Seg. Index (I_s)			Conc. Change (P_s)		
		Top	Middle	Bottom	Virgin	T - V	M - V	B - V	T - V	M - V	B - V
Eskal 0.5-0.8	D10	500.7	501.8	515.4	512.6	1.65%	1.32%	0.08%	12.84%	11.48%	-2.84%
	D50	800.1	824.5	855.2	851.4	2.09%	0.63%	0.02%	14.45%	7.95%	-1.32%
	D90	1307.0	1373.3	1420.6	1416.9	0.49%	0.09%	0.00%	6.98%	3.04%	-0.30%
Eskal 0.1-0.5	D10	48.9	51.3	51.2	50.2	0.01%	0.00%	0.01%	1.18%	-0.64%	-0.91%
	D50	269.4	283.3	283.3	277.9	0.06%	0.02%	0.02%	2.47%	-1.30%	-1.46%
	D90	622.0	637.1	646.5	632.0	0.00%	0.00%	0.01%	0.37%	-0.28%	-0.83%
Eskal 150	D10	94.4	95.9	96.2	95.5	1.03%	0.09%	0.22%	10.13%	-3.07%	-4.70%
	D50	129.8	131.5	131.9	131.1	0.12%	0.01%	0.05%	3.50%	-1.20%	-2.19%
	D90	177.8	180.2	181.0	179.6	0.01%	0.00%	0.00%	0.74%	-0.26%	-0.58%
Eskal 80	D10	48.6	50.4	51.5	50.8	0.89%	0.03%	0.04%	9.42%	1.80%	-1.94%
	D50	78.9	81.2	83.2	82.2	0.78%	0.08%	0.07%	8.81%	2.85%	-2.59%
	D90	118.6	118.0	121.7	120.3	0.07%	0.01%	0.00%	2.57%	1.18%	-0.68%
Eskal 30	D10	12.8	12.8	13.7	13.1	0.27%	0.23%	1.00%	5.19%	4.80%	-9.99%
	D50	26.7	26.7	27.4	27.0	0.01%	0.01%	0.05%	0.95%	1.15%	-2.16%
	D90	47.6	47.7	48.7	48.0	0.00%	0.00%	0.00%	0.29%	0.21%	-0.58%
Eskal 15	D10	11.0	11.1	11.1	11.1	0.10%	0.00%	0.01%	3.21%	-0.10%	-0.90%
	D50	18.1	18.2	18.2	18.2	0.02%	0.00%	0.00%	1.39%	0.23%	-0.44%
	D90	27.9	27.9	28.0	28.0	0.00%	0.00%	0.00%	0.27%	0.09%	-0.19%
Eskal 10	D10	5.5	5.5	5.6	5.5	0.04%	0.01%	0.02%	2.11%	1.00%	-1.41%
	D50	10.2	10.2	10.3	10.3	0.01%	0.01%	0.00%	1.18%	0.86%	-0.09%
	D90	17.7	17.7	17.7	17.8	0.00%	0.00%	0.00%	0.04%	0.11%	0.05%