

Correlations Between Segregation Intensity and Material Properties such as Particle Sizes and Adhesions and Novel Methods for Assessment

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Assessment of powder segregation is essential in most powder-based industries when homogeneity of the powder is a concern. This study focuses on exploring correlations between segregation possibilities of the powders with a wide size range and some common material properties such as particle size, size ratio and adhesion, so that a simple method for assessing segregation intensity of a powder just based on the material properties can be developed, although the current study is designed for surface rolling segregation only.

In this study, eight blends were made from different grades of calcium carbonate, which were selected based on different particle size ranges and particle adhesions. Segregation of a sample was determined by measuring segregation index, which was defined as a ratio of fines concentration change between segregated samples at the top and bottom sections of a surface segregation tester and the averaged concentration. Particle size distributions of the blended samples were measured before segregation tests and then the segregation tests were completed for the same blends. Powder adhesion of the virgin blends was also examined and represented by measuring the “bond number” using a mechanical surface energy tester developed at the Wolfson Centre.

The experimental results showed correlations between the segregation index and the material properties such as particle size, size ratio and bond numbers, and was fitted to multiple empirical functions. With the correlations determined, novel methods for assessing surface segregation have been developed; one method considers just the particle sizes and the other one considers both particle sizes and particle adhesion identified by the bond number.

Keywords: Surface segregation, Segregation index, Particle size, Particle adhesion,

1 Introduction

Segregation of solid particles is a well-known phenomenon in many industrial processes such as pharmaceuticals because particles in terms of different size, shape or density are separated in movement of particles, e.g. loading and off-loading process [1, 2]. The segregation in powders can result in uneven mixing and bias to a single component. For example, in pharmaceutical industries, significant variation of particle size distributions (PSDs) and heterogeneity of blend components in material handling processes leads to compromised quality in the final product eliciting failures during quality control [3]. Prescott *et al.*, [3] showed a significant change of particle sizes led to variation of a critical component (typically an active pharmaceutical ingredient (API)), and failure in blending processes, with a focus on free-flowing materials. It brings challenges to process engineers either the materials do not mix very well if they are too cohesive or the materials are segregated even after mixing if they are too free flowing. Finding an optimised window for selecting material properties is not easy to identify, the material with this window would be suitable for blending but have little segregation. To make this decision clearly, it would normally require many experimental tests or use powerful simulations [5, 6]. For practicality, it would have benefits if segregation of a powder can be identified by its physical properties such as particle size and adhesion.

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For surface segregation, Brown [4] described it as particles separating by moving with different velocities due to different levels of friction opposing their momentum, where the form of the heap surface was approximately constant. To evaluate segregation due to rolling mechanism, investigations were made experimentally and numerically. Fan *et al.* [6] suggested that particle movement during heap formation could be affected by not just particle dynamics (such as velocities), but also by the nature of the surface of the heap and the properties of the solids. Obviously, mechanisms for surface rolling segregation have two aspects: particle dynamics and nature of solid properties.

Influences of particle dynamics on surface segregation have been studied and shown that the particles' initial kinetic energy is a major factor on levels of segregation generated [7]. With identical initial velocity, different sized particles have different momentum. However, particle movements on the slope surface are subject to the friction forces between the particles and the surface formed by the stationary heap of particles. Therefore, the particle movement is influenced by particle rolling tendency and movement of the surface. Both movements are subject to the nature of these particle properties, such as difference in particle sizes, particle shape, density, adhesion, *etc.* An extensive study on the influences of material properties on surface rolling segregation has been carried out recently [8-10]. It is concluded that the significant influential factors are size, size difference and adhesion [11-12], but sometimes differences in density and particle shape can have a more significant influence than the particle size [13]. If it is presumed that the dynamic effects would be constant under certain conditions, levels of segregation that a powder could suffer can be identified by its material properties.

In the pharmaceutical process, it has more concerns on the segregation of blended materials in buffer hopper as consequent influences occur in tablet compression process. Lakio *et al.*, [14] suggested that smaller particle size gave better tablet-ability, but the effects of particle size and particle size distribution of granules alone could not explain how the granules behaved when the particles were compressed. Other factors influencing tableting included the flowability and surface texture of the granules, while porosity, density and particle shape of the granules could also have an effect. For assessing material segregations, because of the complexity of the nature of material properties, enormous efforts are required in terms of particle size, size difference, particle densities and particle shapes, and the other influential factors such as friction and adhesions. Especially for pharmaceuticals, where APIs often need to mix with different excipients, assessment of segregation can be a big challenge at an early stage, as there is not enough quantity of the materials available [15-16]. In practice, 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) [17]. However, by taking the level of cohesiveness of the components, it is hard to decide cohesiveness of the materials that do not segregate but are still able to be mixed. To have an optimised decision, assessment of material segregations must have a large quantity of materials available for the experiments.

To solve this problem by using a small quantity of the materials, a hypothesis was developed for a method that can indicate material segregation intensity by its material properties such as particle sizes and adhesions, that way the amount of materials is sufficient for material characterisation rather than needing to perform too many segregation tests. The method also has great advantages by limiting the number of segregation experiments while validating numerical simulations. This study aims to explore possible correlations between segregation intensity of a powder defined by a segregation index and the nature of material properties including particle size, size range and particle adhesion, thereby establishing empirical models between the segregation index and the material properties.

With the models, intensities of surface segregation of a powder can be evaluated by its size distribution and adhesion if influences of particle density and shape are not considered.

2 Materials and methods

2.1 Sample materials and preparation

For this experiment, a total of eight blends were made from different grades of calcium carbonate, giving a wide range of material properties including PSDs, flow functions, and particle adhesions as varied levels of fines contents. The calcium carbonate was named with Eskal series grades and manufactured by KSL Staubtechnik GMBH, Germany. Because the Eskal materials used in this study are manufactured in the same process from a single feedstock, thereby showing high chemical uniformity and classified for different size categories based on different applications, particle density and shapes are likely the same.

Virgin materials used for the sample preparation are listed in Table 1. Summarized particle size percentile values (volume % measured by the Malvern laser diffraction method) for the materials are also shown in the table with other physical properties including solid particle density; ρ_s measured by a nitrogen pycnometer, bulk density, ρ_b at relatively low consolidation stress (about <1 kPa), particle flow functions ($1/ff_c$) measured by a particle flow tester (PFT, Brookfield) [18], and bond number (B_o) by a mechanical surface energy tester (details see section 2.2).

Table 1: Virgin materials used for making blends and material physical properties

Name of materials	Particle Size (μm)			Size Span ($D_{90}-D_{10}$)/ D_{50}	Solid Density (kg/m^3)	Bulk Density (kg/m^3)	Flow Function ($1/ff_c$)	Bond Number (B_o)
	D_{10}	D_{50}	D_{90}					
Eskal 2	1.1	3.3	8.6	2.26	2800	711	1.57	11.40
Eskal 4	1.6	4.0	8.7	1.77	2800	1055	0.93	8.64
Eskal 10	4.6	10.0	18.4	1.37	2800	1254	0.56	6.57
Eskal 30	12.5	26.7	46.1	1.26	2800	1354	0.14	5.69
Eskal 80	46.1	82.3	129.5	1.01	2800	1483	0.14	4.75
Eskal 150	93.7	131.1	183.1	0.68	2800	1489	0.13	4.13
Eskal 0.1-0.5	62.6	367.9	1125.9	2.89	2800	1496	0.33	3.80
Eskal 0.5-0.8	507.4	898.1	1498.9	1.10	2800	1502	0.09	3.64

Eight designed blends were made from the eight grades of calcium carbonate shown in Table 1 at different ratios in weight, which were selected in terms of particle size ranges of the blends and flow functions and cohesiveness which the blends produced. For each designed sample, two virgin materials were selected proportionally in a designed ratio of weights as shown in the blends' names of (see Table 2) and prepared by a Turbula T2F powder mixer.

The blender for the sample blending process was run at 40 rpm for 1.5 hours to achieve a homogenous mixing as suggested in the literature [19]. The blended samples were characterised and shown material properties in Table 2 including particle sizes, size ratios (D_{90}/D_{10}), bulk density, flow functions and the bond number (B_o). The prepared samples were collected into a single container and subdivided into a few sub-samples by a mechanical riffler to ensure using identical samples to measure the material properties and for the segregation tests.

Table 2: Blended sample materials for segregation tests and physical properties

Designed Blends (ratio of weight)	Particle Size (μm)			Size Ratio D_{90}/D_{10}	Bulk Density (kg/m^3)	Flow Function ($1/ff_c$)	Bond Number (B_o)
	D_{10}	D_{50}	D_{90}				
E2-E80 (20%-80%)	3.2	76.4	120.3	37.59	1384	0.80	6.64
E2-E0.1 (20%-80%)	3.5	292.6	706.1	201.7	1164	1.00	11.12
E4-E30 (50%-50%)	2.1	7.1	40.1	19.09	1173	1.00	10.53
E4-E0.5 (20%-80%)	434.2	859.3	1413	3.25	1382	0.91	10.98
E10-E80 (80%-20%)	5.7	11.9	90.9	15.95	1499	0.55	6.58
E10-E150(20%-80%)	10.6	119.0	171.3	16.16	1741	0.26	6.50
E10-E0.1(50%-50%)	6.0	132.9	637.9	106.3	1673	0.85	8.71
E30-E0.5(50%-50%)	22.6	727.6	1344	59.46	1431	0.34	6.47

2.2 Particle adhesion and the 'bond number'

Regarding the influences of material properties on levels of segregation that may occur, adhesion between particles is identified to be important. Strong particle adhesion produces high friction, which can be determined by the 'bond number' (B_o) in terms of particle median size such as D_{50} . The 'bond number' (B_o) is defined as a ratio of adhesion force between a particle and an attached solid surface (or adhesion force for dissimilar particles) to the particle weight due to gravity as shown in Figure 1(a) [20, 21]. To measure the 'bond number' (B_o) of a powder, a mechanical surface energy tester has been developed at the Wolfson Centre and shown in Figure 1(b) [22]. The 'bond number' (B_o) method is also used for study of interparticle forces from powder flow properties recently [23].

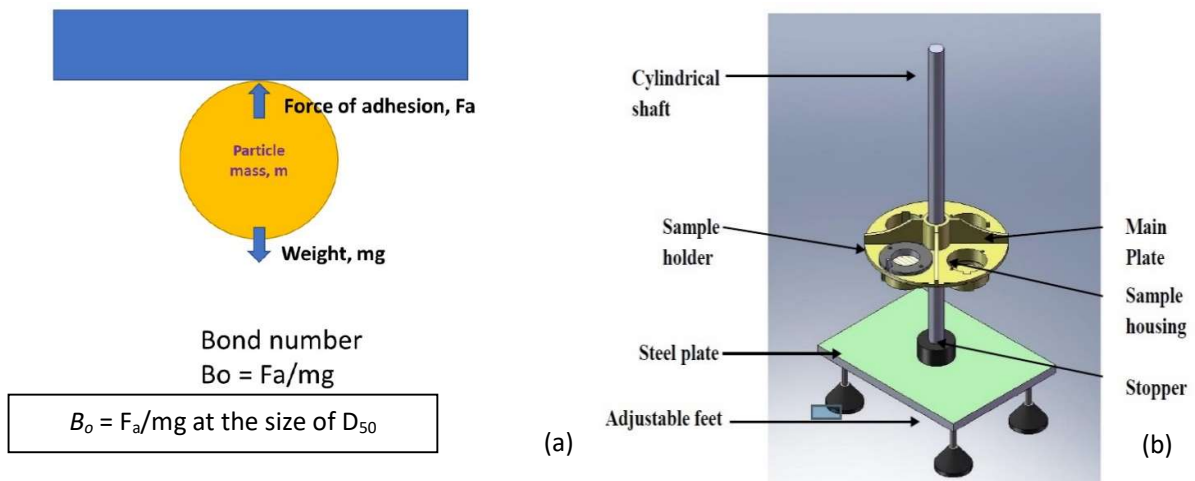


Figure 1: (a) Principle of the 'bond number' (B_o) defined, (b) Sketch of a mechanical surface energy tester [22]

For measuring the 'bond number', about 50 milligrams of the sample particles of a powder are deposited onto a prepared tablet surface that is either compacted from a few grams of commonly available powder or made of the same sample powder. The tablet represents the solid surface and the particles are used for the 'bond number' measurement. The tablet is then turned upside down and fitted into the sample holder (Figure 1 (b)). The sample holders and the housing are lifted to a certain height, and then released to drop. The tablet is halted against a buffer at the stopper, allowing a proportion of the attached powder to be detached from the solid surface. The deceleration value of the tablet is recorded at the same time. The detached particles are collected and measured for total mass. The particles are also examined under a Malvern G3 microscope to ascertain the nature and

the number of the detached particles for measuring particle median size. Finally, by using the deceleration value, the particle 'bond number' (B_b) at the median size is calculated (see Figure 1(a)).

2.3 Rolling segregation tester and test procedure

For surface rolling segregation tests, a segregation tester (QPM) developed at the Wolfson Centre, University of Greenwich [24] has been used (see Figure 2), which can quantify segregation intensity in a heap formation, where particle segregation is observed due to rolling (including percolation) mechanism. The tester has advantages and is convenient for segregation prediction in industrial processes such as material loading and off-loading from a hopper without compromising the prediction due to equipment scaling up [1]. The tests can also be repeated to give a statistic result, but typically requires a certain amount of test materials (about 0.5 litres) for a single test. If several parameters need to be investigated, significant tests and sample materials are required.

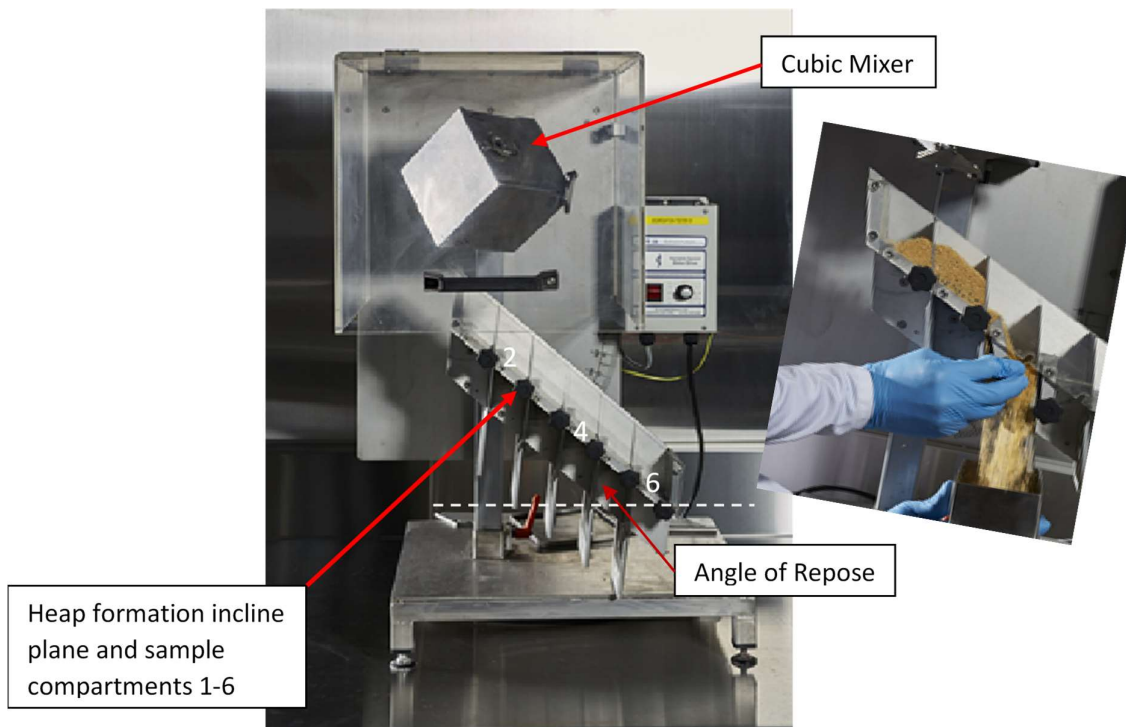


Figure 2: Surface segregation tester (QPM) for heap formation and surface segregation analysis

The QPM segregation tester consists of a cubic mixer and an adjustable inclined trough. In this study, the cubic mixer has not been used because some of the blends are very cohesive and not suitable for a cubic mixer. In the experiments, the blended sample is discharged by a screw feeder at about 25 g/s feed rate with a drop height at about 5 cm above the first compartment (the same height as the cubic mixer outlet). The trough is placed at an angle equivalent to the angle of repose for the substance to create a smooth and consistent heap of powder. The sample forms a slope of a heap with segregated patterns and is ready for sampling. There are six equally sized compartments which can be formed by sliding gates. Each section can be discharged individually and the sample material in the compartment can be collected for further analysis. The section numbers 1-6 are set from top to bottom of the trough. Only the sample from compartment 2 is used as the 'Top' section sample and the sample from compartment 6 as the 'Bottom' section sample. The trough is approximately 380 mm

long and cross-section is about 55 mm wide by 55 mm high. A sample of approximately 0.5l bulk material is used for a single segregation test.

The segregated samples collected from the segregation tester are usually too big in quantity for particle size analysis. Therefore, all segregated samples are subdivided by using a mechanical riffler splitter, so appropriate samples (about 40–50 grams) can be obtained for size analysis without biasing the results. To minimise random errors, segregation tests have been repeated if the results are not showing consistency.

2.4 Particle size analysis

Particle size distributions (PSDs) are determined by the laser diffraction method using a Malvern Mastersizer 2000 with a dry dispersion unit. In the measurement, a sample of approximately 20g is introduced into a dry dispersion unit and, in total repeated ten times. For the measurement settings, the air dispersion pressure is set at 1.5 bar with a vibration feed rate of 70% for all the tests. The particles' volume distribution is calculated using the 'general-purpose model' in the Mastersizer software. PSD of each sample is calculated with all the repeats, and the average with standard deviation is reported and used for data analysis. With this method, volumetric concentration of the PSDs is given and particle sizes at D10, D50 and D90 are also found. PSDs of the blended materials in Table 2 are shown in Figure 3.

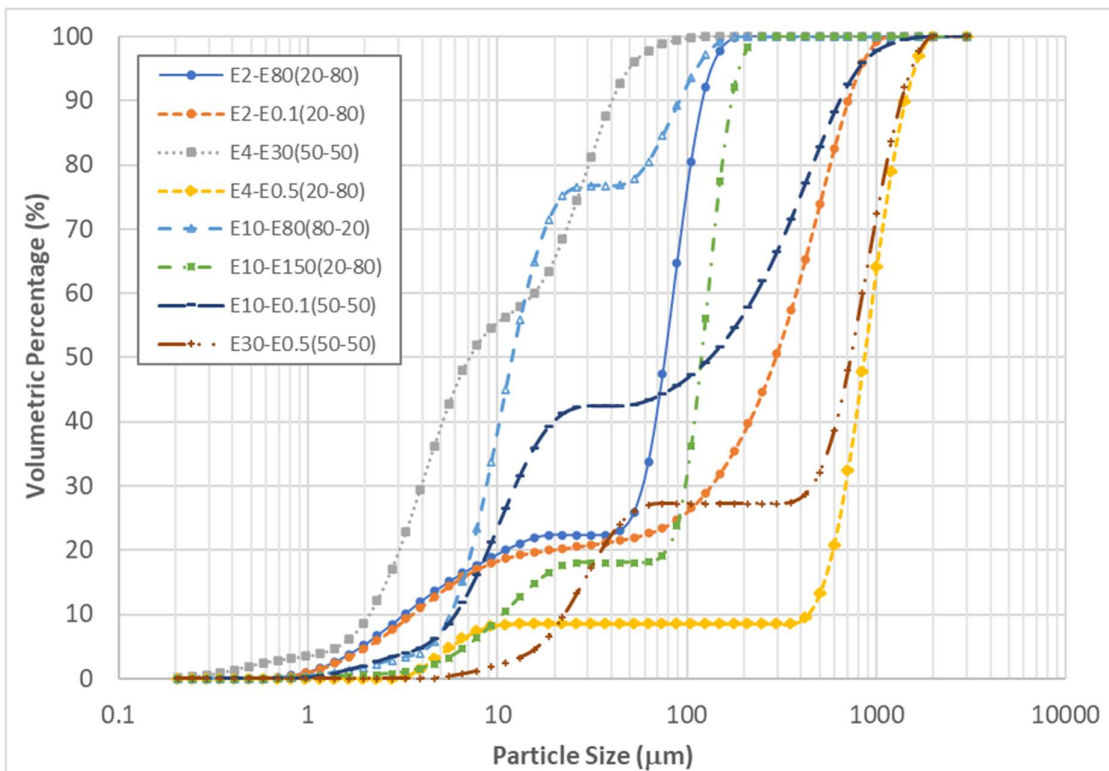


Figure 3: Particle size distributions for the eight blended powders of calcium carbonate

In this study, the blends are named with their original commercial names in short (e.g. E10 for Eskal 10) according to their size categories and the blending ratio in weight percentage. Eight blended materials tested (see Figure 3) show a wide size range with different median sizes. It is noticeable that some of the blends, *i.e.* E4-E0.5 and E30-E0.5, do not match the designed ratios exactly, because

of a possibility of losing fines in blending or in the sampling process. However, it does not influence the segregation study here and consequent conclusions.

2.5 Segregation index

A segregation index is defined as a dimensionless number, which is used to identify the change of the components of interest between homogeneously blended and segregated powder. To quantify segregation of blends, a number of indices have been introduced 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 [25-27]. Our previous study [13] shows that a segregation index based on the variance compromises resolution of segregation measurements, when segregation levels are small. Therefore, a segregation index based on the volumetric (or mass) concentration change of fines has been introduced as:

$$P_{s(i)} = \left(\frac{C_i - C_{o(i)}}{C_{o(i)}} \right) \quad (1)$$

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 the original homogeneous blend at the size i . However, $C_{o(i)}$ could have a problem if the sampling errors in the segregation tests are significant. In order to minimise the sampling errors before and during segregation tests, the segregation index used in this study is calculated based on the segregated samples. It is defined as a ratio of the concentration difference of the fines between segregated samples at different locations (bottom or top section) on a surface segregation tester and the average of the concentrations at the particle size, i , as:

$$P_{s(i)} = 2 \left(\frac{C_t - C_b}{C_t + C_b} \right)_i \quad (2)$$

where C_t (or C_b) is the concentration of fines after segregation at the top location (or bottom location) for the size i . The sizes used in this study are D_{10} , D_{50} , or D_{90} . Averaged absolute segregation index, \bar{P}_s or maximum absolute segregation index, $P_{s(max)}$ are:

$$\bar{P}_s = \frac{\sum_n |P_{s(i)}|}{n} \quad \text{or} \quad P_{s(max)} = \max_n |P_{s(i)}|_n \quad (3)$$

The averaged absolute \bar{P}_s or the maximum $P_{s(max)}$ represents the segregation possibility of a powder across the whole size range. In the results of processing, the averaged absolute \bar{P}_s or the maximum $P_{s(max)}$ at the particle sizes of D_{10} , D_{50} , and D_{90} are used for the assessments to avoid random sampling errors in PSDs analysis and systematic errors in the tests.

3 Results and discussion

3.1 Segregations of the blended samples

Segregation study of the blends has been carried out by the method described in section 2.3. A typical result of the E30-E0.5 (50-50) tests is given in Figure 4. The results show that the material has a size difference at different locations (top and bottom sections) after the material was deposited onto the inclined trough. Compared to the virgin blended material, the top section's material contains more fine particles of about 30% (<100 μ m), but the material in the bottom section only has about 26% fines at the same size. Although the blended sample was designed for a 50%-50% blending ratio in weight, the size measurement shows about 27%-73% in a volumetric ratio (virgin blended sample). It is believed that some fine particles must be lost in the blending and sampling process before carrying out the subsequent segregation tests. This systematic error generated leads to difficulties to apply

virgin sample analysis for the segregation assessment, which is commonly used by other segregation tests. Therefore, in this study the segregation of a sample is assessed by using the samples only after the segregation tests and calculated with the Eq. (2) to minimise the systematic errors generated in operation.

The segregation results of the blends are shown in Table 3, as averaged results of absolute segregation index ($abs \bar{P}_s$) and a maximum absolute segregation index ($abs P_{s(max)}$), which are used for interpretation of the segregation intensity of a sample powder across the size. In the table, the particle size ratio of D_{90}/D_{10} is given. Also, the bond numbers for the virgin blends are included as an indication. Detailed results calculated by Eq. (2) can be found in Table A1 in the appendix for all the samples and the selected repeated tests.

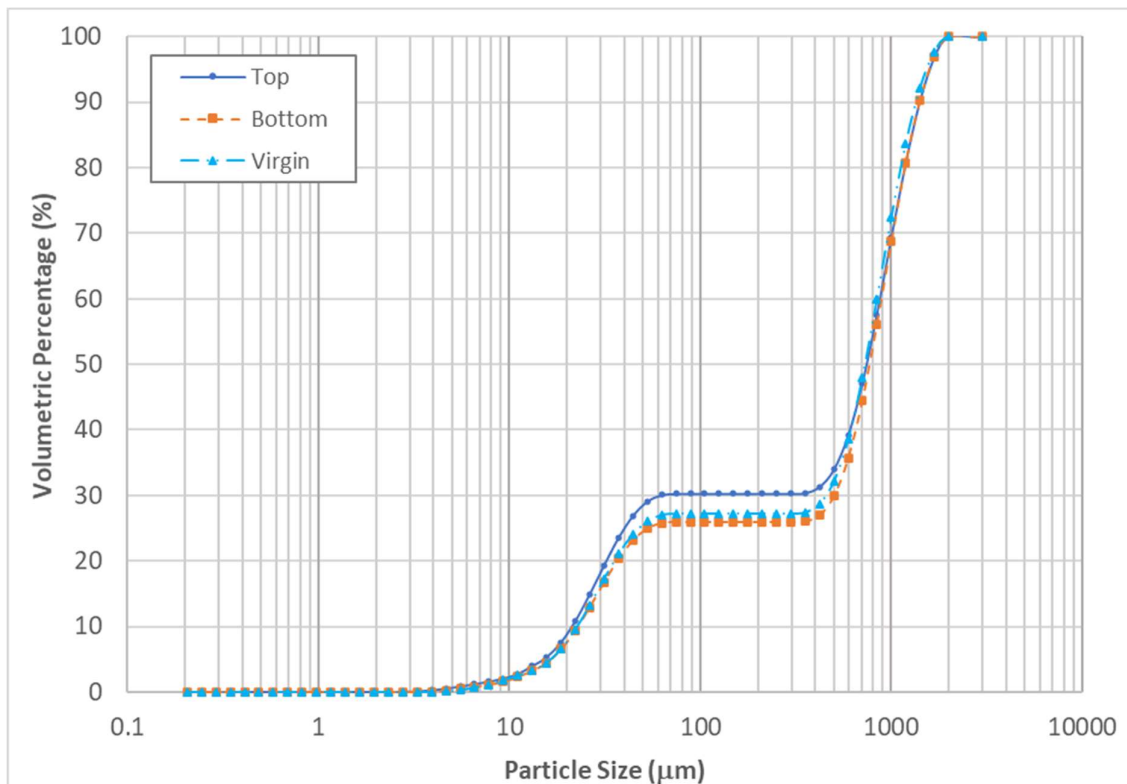


Figure 4: Particle size distributions (PSDs) for the test samples of E30-E0.5 (50-50) blends

Table 3: Segregation results of the blended sample materials

Binary-sized Blends (ratio of weight)	Particle Size (μm)			Size Ratio D_{90} / D_{10}	Bond Number	Average $abs \bar{P}_s$	Maximum $abs P_{s(max)}$
	D_{10}	D_{50}	D_{90}				
E2-E80 (20%-80%)	2.9	74.3	127.8	43.3	6.64	2.2%	5.0%
E2-E0.1 (20%80%)	3.5	273.9	834.6	239.7	11.12	9.2%	17.1%
E4-E30 (50%-50%)	2.1	7.5	38.9	18.4	10.53	0.4%	0.8%
E4-E0.5 (20%-80%)	4.4	746.4	1336.5	303.7	10.98	17.6%	43.3%
E10-E80 (80%-20%)	5.4	12.6	248.1	45.9	6.58	4.3%	8.4%
E10-E150(20%-80%)	12.2	118.9	170.1	14.0	6.5	2.1%	3.8%
E10-E0.1(50%-50%)	6.1	140.1	747.6	122.0	8.71	7.8%	15.7%
E30-E0.5(50%-50%)	22.2	775.5	1430.0	64.3	6.47	6.5%	13.9%

3.2 Influence of particle size on segregation

Commonly a powder with a wide size range tends to have a high segregation while the material moves on an inclined plane. Extra-large particles introduce extra kinetic energy to the slope surface than the fines, but the large particles' movement also depends upon the flowability of the powder and the surface created. If particles are presumed to start with an identical initial velocity, particles stop subject to frictional forces acted and initial kinetic energy carried out, where particle properties play key influences on the particle movement including particle size, cohesiveness, density, and particle shape. Apart from the dynamic effects of the particles such as initial particle velocities which are constant, levels of segregation that a powder could suffer can be identified by its material properties. A correlation between segregation intensity of a powder and particle size has not been clearly established before although a general trend has been shown in our previous study [13]. The trend in the previous study shows that a powder with increasing particle size and size spans has high segregation intensity, but there is not a clear correlation. The same work has been repeated and the results for the blends are plotted versus particle size and shown in Figure 5, as averaged absolute \bar{P}_s against particle sizes of D_{10} , D_{50} and D_{90} .

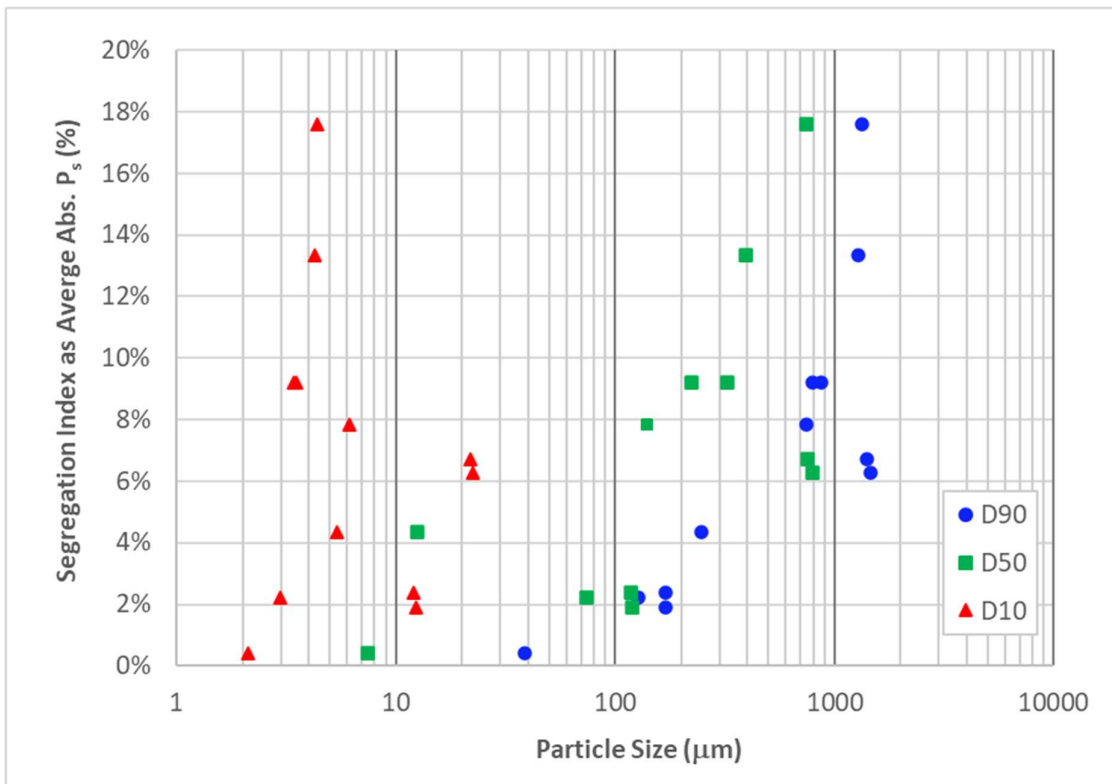


Figure 5: Segregation levels for the blends as averaged absolute \bar{P}_s versus particle size of D_{10} , D_{50} and D_{90}

To study influence of particle size difference, the segregation results against particle size ratio (D_{90}/D_{10}) are shown in Figure 6. For the averaged absolute \bar{P}_s , there is a better fitted linear relationship with a R^2 value of 0.8569. For the maximum absolute $P_{s(max)}$, the linear fitted correlation is poor as R^2 value is 0.6909. However, the results shown in Figure 6 indicate that particle size ratio has a more robust relationship compared to the particle size.

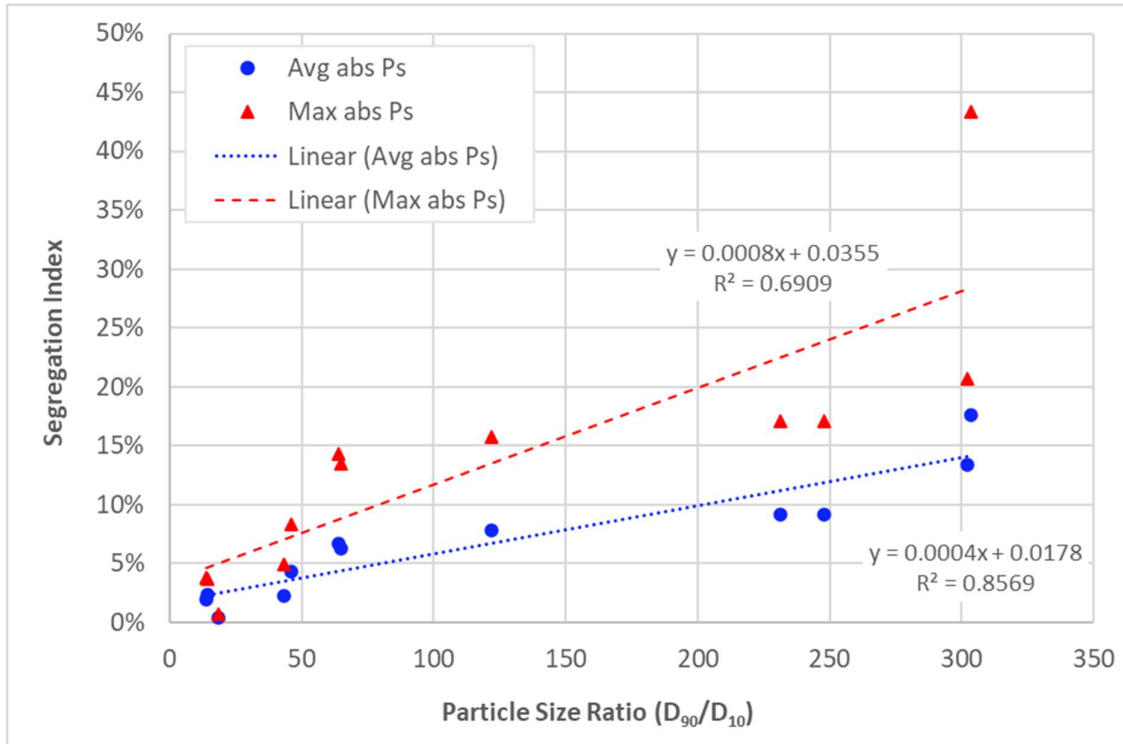


Figure 6: Segregation indices of the blends tested against particle size ratios

3.3 Influence of particle flow functions and adhesion on segregation

The segregation results are plotted against flow functions of the virgin blends as shown in Figure 7. A multiplicative inverse for flow function coefficient ($1/ff_c$) is used to interpret the results with linear trends fitted to the averaged absolute \bar{P}_s , and the maximum $P_{s(max)}$. This result does not show any correlations, although our previous study [13, 28] has indicated that flowability of a powder has a clear effect on its segregation intensity. A cohesive powder has little segregation, but a free-flowing powder tends to segregate more. In Figure 7, it shows that some cohesive powders do segregate less due to high cohesiveness, but not all the cohesive powders follow this hypothesis. It is understood that cohesiveness of a powder cannot rule out the control of particle size difference in surface segregation when the size difference is significant. For surface segregation, consolidation stress in the particle moving layer is negligible, but a traditional flow function measured of a powder normally means material flowability under some consolidation stresses [29], therefore direct correlating the flow functions to surface segregation levels does not work.

The results show that the particle size and the size ratio can influence segregation intensity of a powder strongly in surface segregation, as well as powder cohesiveness having a similar effect. Highly cohesive powders have little segregation compared to less cohesive powders. In the case of this study shown in Figure 7, it is impossible to establish a clear correlation of powder flow function to the segregation intensity. A better indicator to describe the influence of powder cohesiveness on surface segregation therefore is needed. Because the bond number describes the particle adhesion at zero consolidation stress for a given particle sizes, it is believed that the bond number is more suitable for this purpose. In section 2.2, the bond number has been explained and measured at the median particle size (D_{50}). The bond numbers must be different with different particle sizes; therefore, a bond number can describe particle adhesion at a specific particle size. A ratio of the particle size ratio to the bond number is taken in correlation to the segregation. The segregation results in Figure

8 show a better correlation of R^2 value of 0.9062 for the \bar{P}_s , and 0.7446 for the $P_{s(max)}$. Compared to the results in Figure 6, the correlations for a linear fit are improved.

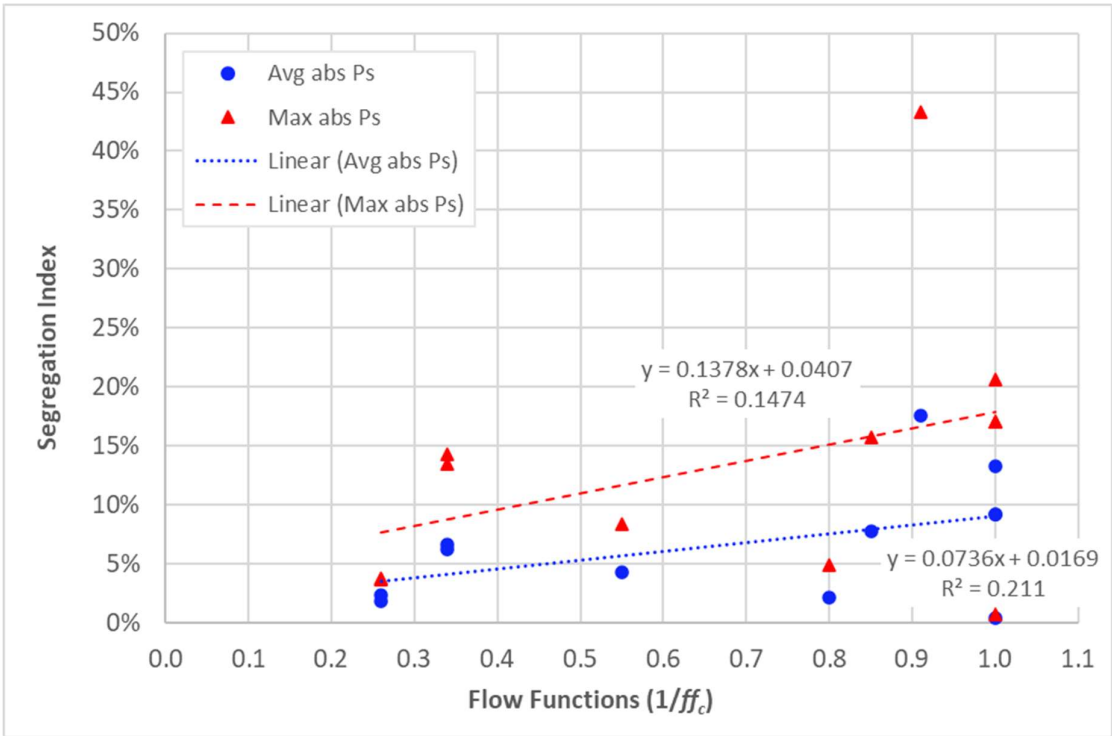


Figure 7: Segregation indices of the blends tested versus flow functions ($1/ff_c$)

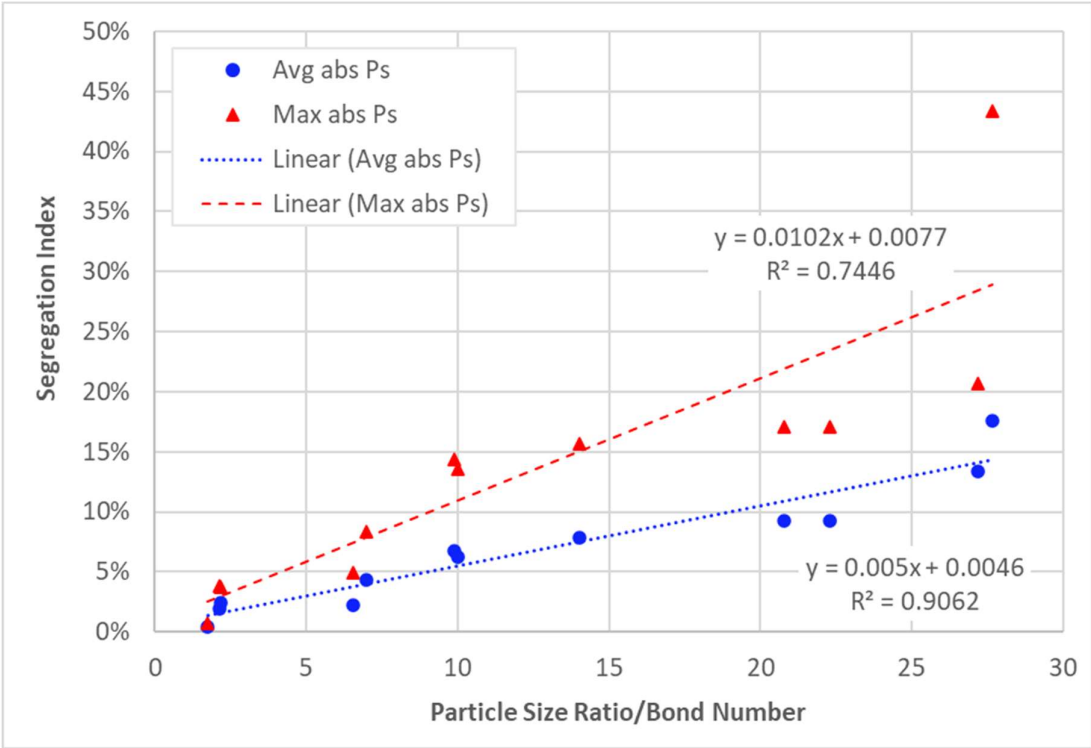


Figure 8: Segregation index versus the ratio of the particle size ratio to the bond number

3.4 Correlations between segregation and material properties

The results in Figure 8 show a strong correlation between segregation intensity of a powder and the material properties of the powder. Therefore, the $\text{abs } \bar{P}_s$ in Table 3 for the eight blends are plotted against the ratio of size ratio to the B_o and shown in Figure 9. Although the variations (as error bars) of the measurements are limited to the number of the measurements carried out, the results show a very good interpretation of R^2 value of 0.9677.

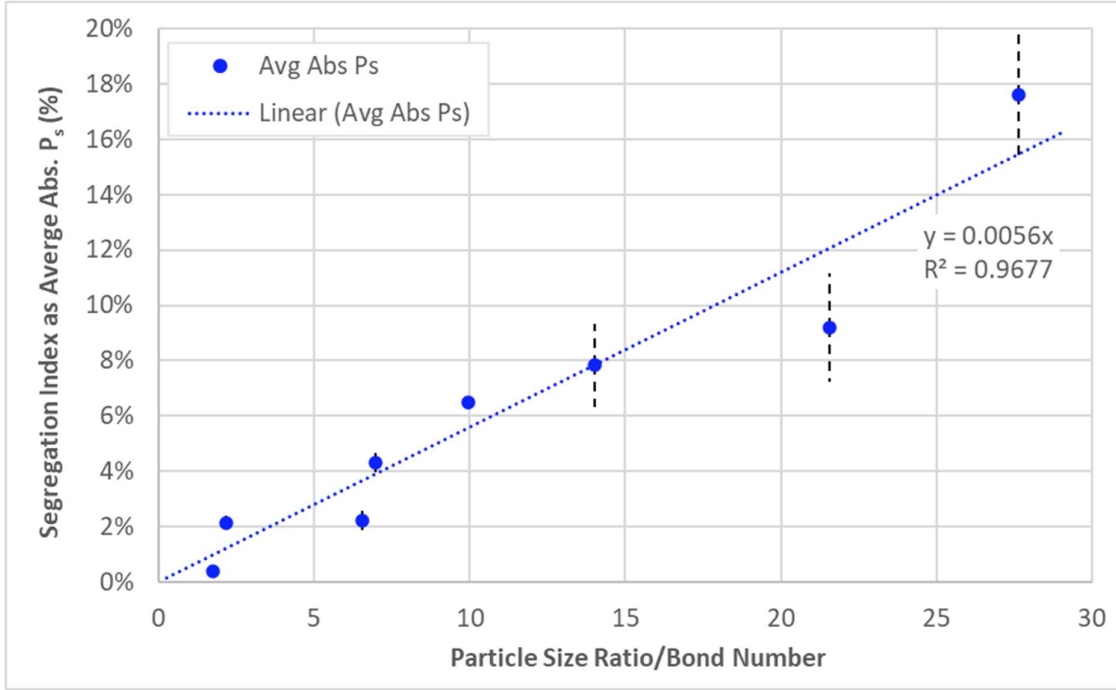


Figure 9: The correlation between the segregation and the ratio of size ratio to the B_o

For the results in Figure 9, it is realised that a correlation by segregation intensity of a powder and material properties such as particle size, size ratio, and the bond number can be established. If only based on particle size ratio ($R_s = D_{90}/D_{10}$) and the particle median size (D_{50}), segregation intensity of a powder can be expressed with different power weighting as:

$$P_s = k(D_{50})^m \left(\frac{D_{90}}{D_{10}}\right)^n = k(D_{50})^m (R_s)^n \quad (4)$$

The k , m , and n in Eq. (4) are constant for the material tested. The two sides of the equation are not equivalent in units because the D_{50} has a unit of *micron*. With the data in Table A1, the empirical parameters are determined, as $k = 0.0045$; $m = 0.15$ and $n = 0.4393$. The forecast by Eq. (4) is compared to the experiment results, and the comparison is given in Figure 10.

The results in Figure 10 show a good agreement between the Eq. (4) and the measurements as R^2 is about 0.964. This correlation is determined empirically by particle size, and not explained in physical meaning as both sides of the equation do not have the same units. It shows an advantage that the particle size and size ratio can be used to indicate the powder's segregation intensity if the empirical parameters for the powder can be identified in advance.

Additionally, a correlation between segregation intensity of a powder and material properties of particle size ratio (R_s) and the bond number (B_o) can be interpreted as:

$$P_s = k \left(\frac{R_s}{B_o} \right)^m = k(K)^m \quad (5)$$

The k and m in Eq. (5) are constant for the material tested. For this correlation, both sides of the equation are the dimensionless numbers and give more a meaningful explanation in physics. With the data in Table A1, the empirical parameters for the Eq. (5) are determined, as $k = 0.0015$ and $m = 1.3992$. The prediction results by Eq. (5) are compared to the experiments, and the comparison is shown in Figure 11.

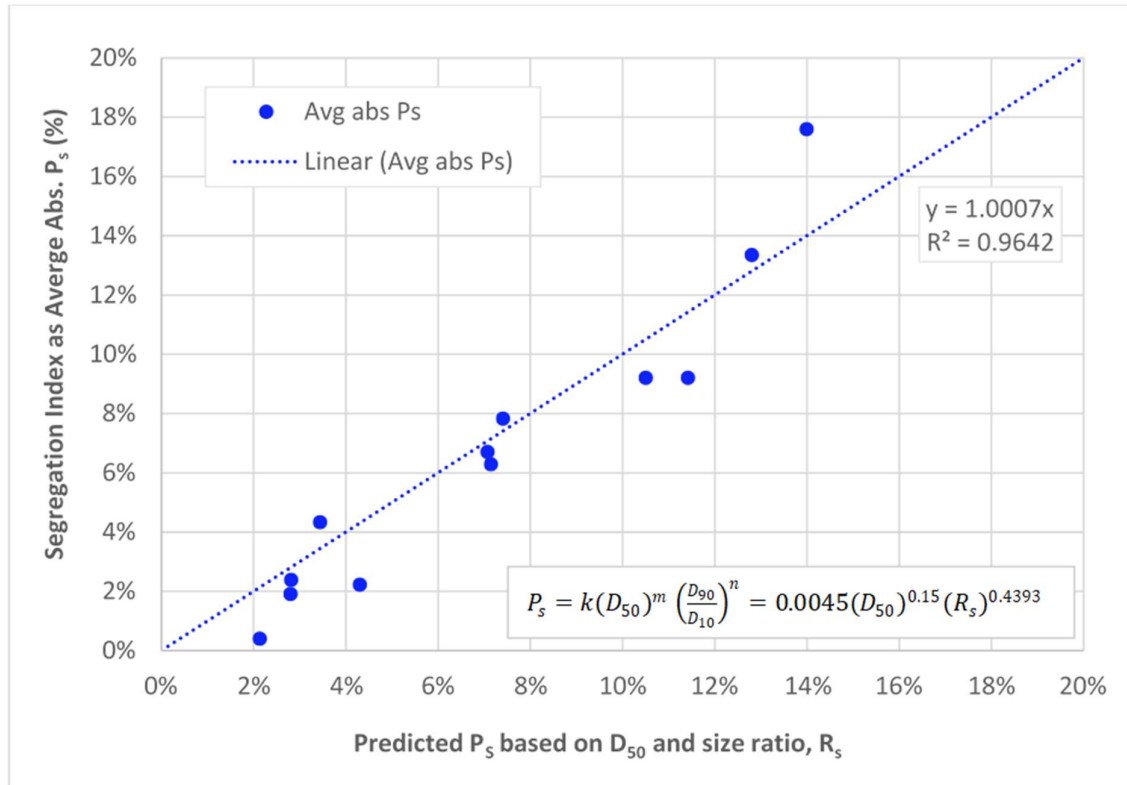


Figure 10: Comparison between the empirical model based on median size and size ratio to experimental measurements

The results in Figure 11 also show a good agreement between the Eq. (5) and the measurements as R^2 is about 0.948. However, the equation with the (R_s/B_o) ratio shows slightly under prediction at the low end and over-prediction at the high end of segregation. The Eq. (4) would give a slightly better fit than the Eq. (5) but generally, both correlations of material properties can be applied for assessing the segregation intensity of a powder if particle density and shape are not considered or have limited effects.

3.5 Empirical segregation models based on material properties

For the correlations established in the equation (4) and (5), segregation models based on material properties have been developed, the properties can be the particle size only or size ratios with the bond numbers. With the Eq. (4), a model based on particle size and size ratio is shown in Figure 12. In the size model, segregation intensity has been divided into several ranges identified by the Eq. (4). The dark blue areas show to have little segregation if the particle size ratio is less than about 15. With the reduced particle median size, the size ratio can increase to maintain the same segregation level. In the figure, experimental data are also presented. With the Eq. (5), another segregation model based

on bond numbers and particle size ratios is shown in Figure 13, while the bond number represents particle adhesion and flowability. A particle median size is not used directly in this model, but the bond number is detected for the median size. Therefore, the bond number model is correlated to more material properties including particle size, size range, flowability and particle adhesion.

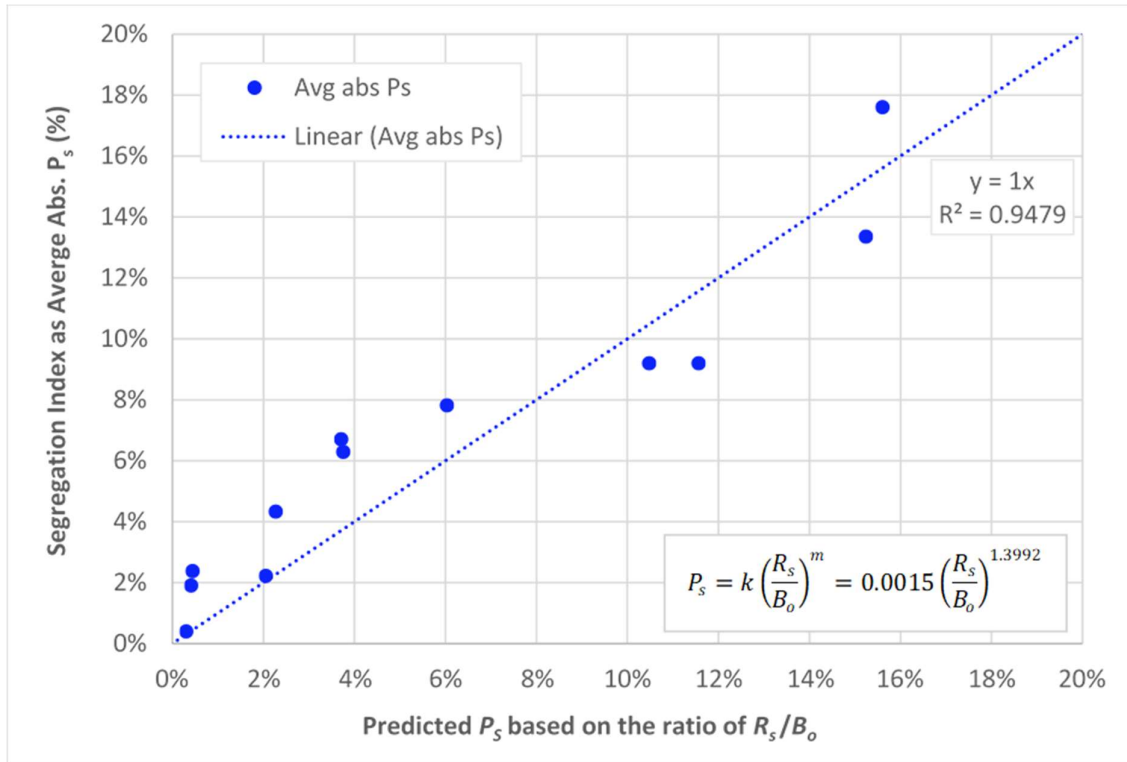


Figure 11: Comparison between the empirical model based on a ratio of particle size ratio to bond number to experimental measurements

The segregation models introduced here show strong potential for practical applications if the models are calibrated for more materials or blends in the future. Assessing surface segregation of a powder by only using the material properties such as particle size, size ratio and the bond number holds great advantage by reducing the number of the segregation tests significantly. The models can generate an indication of segregation intensity of a powder at early formulation stage when a large quantity of proposed blended materials is not available for further segregation tests.

4 Conclusions

In this study, correlations between the intensity for surface segregation and the material properties such as particle sizes, size ratios and particle adhesions were explored by using designed blends of different grade powders to give a wide range of particle size and adhesion but the same particle density and similar particle shape.

By the segregation index defined, the results showed that segregation intensity of a powder had a clear correlation with its particle size ratio, but not the particle size. There was also a possible increasing trend of segregation with increased particle sizes, but a specific correlation was challenging to define. A correlation of segregation intensity and particle size ratio was established, but a correlation between the blends' flow functions and the segregation intensity was not found. The

reason is believed to be that the flow function is measured under consolidation stress, but in surface segregation, consolidation stress is a minimum to deficient value. The bond number of a powder, a ratio of particle adhesion to its gravity of the particle, can be a better choice to describe particle adhesion influences on surface segregation. By introducing a new parameter, a ratio of the size ratio to the bond number, the correlation of the segregation intensity and the particle size ratios improved.

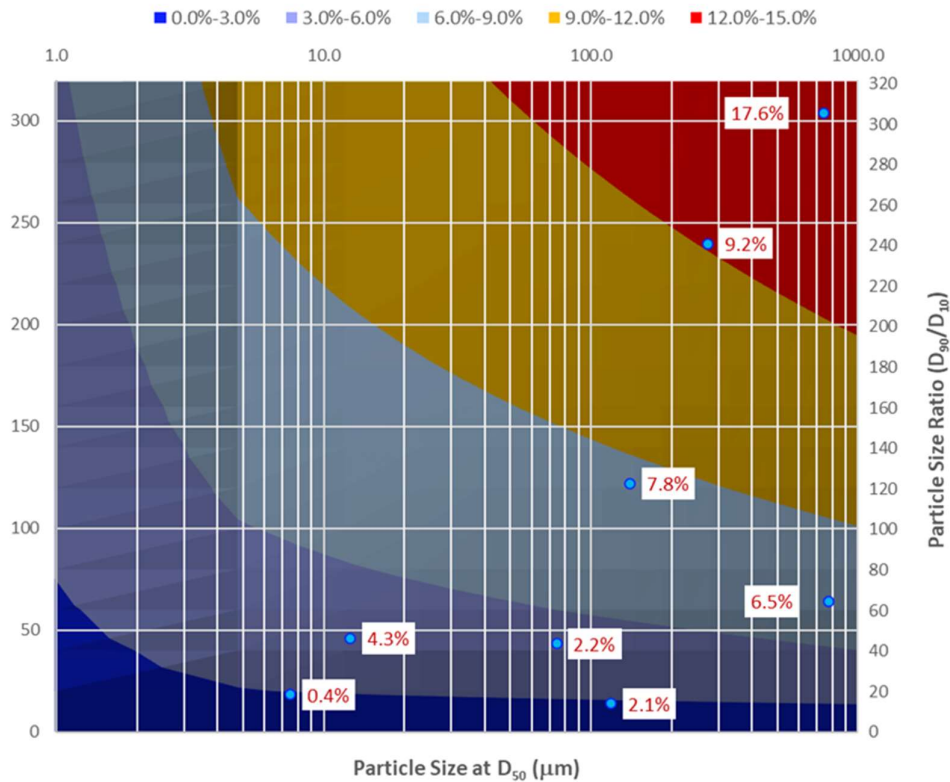


Figure 12: A segregation model based on particle size (D_{50}) and size ratio (D_{90}/D_{10})

A strong correlation between the experimental results and the ratio of the size ratio to the bond number was found with R^2 value of 0.967. The results demonstrated a strong possibility that surface segregation intensity of a powder can be quantified by the material properties. Two correlations were established, one was based on particle size and size ratio only, and the other one was based on particle size ratio and bond number. Both correlations gave good predictions with R^2 values of 0.964 and 0.948 to the experimental results.

With the correlations established, segregation assessment methods based on material properties were developed using either the particle size only or size ratio to the bond number. The novel methods show significant advantages in assessing material segregation intensity by using material properties if a large quantity of the materials is not available for many segregation tests. Although the current study is limited to the materials tested and the surface rolling segregation, it is possible to validate the methods further on different powders or even different blends of powders for different types of segregation mechanisms.

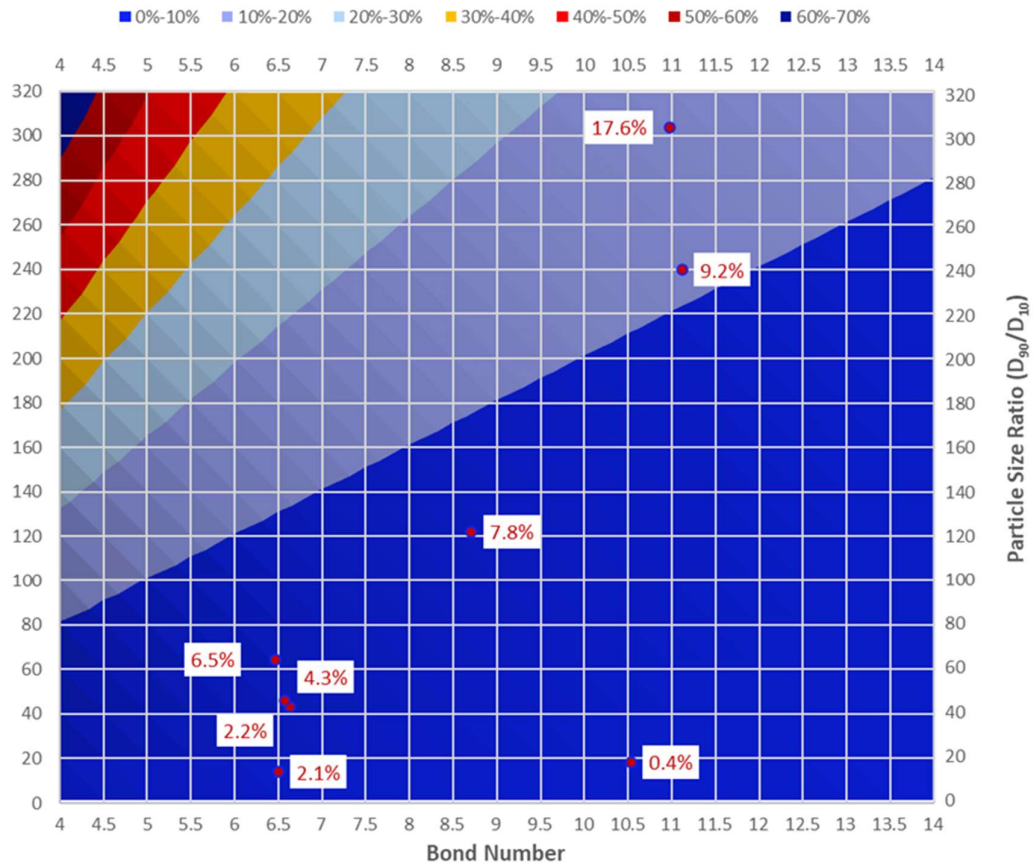


Figure 13: A segregation model based on bond number (B_o) and particle size ratio (D_{90}/D_{10})

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix:

Table A1: Segregation indices for the different size category of (D_{10} , D_{50} and D_{90}) of the blended samples and the repeated tests

Samples	Test Materials	Particle size (μm)			Seg. Index (P_s) at the size by volume	Average abs \bar{P}_s	Maximum abs $P_{s(max)}$
		Size Fraction	Top	Bottom			
Sample 1	E2-E80 (20-80)	D10	3.0	2.9	0.82%	2.2%	5.0%
		D50	75.5	73.1	-4.96%		
		D90	127.6	128.1	-0.88%		
Sample 2	E2-E0.1 (20-80)	D10	3.4	3.4	4.19%	9.2%	17.1%
		D50	277.5	168.3	17.05%		
		D90	897.2	695.1	6.37%		

Sample 2 R1	E2-E0.1 (20-80) R1	D10	4.8	3.7	-20.69%	13.4%	20.7%
		D50	436.3	354.6	-11.99%		
		D90	1382.8	1187.8	-7.38%		
Sample 2 R2	E2-E0.1 (20-80) R2	D10	3.4	3.6	4.19%	9.2%	17.1%
		D50	289.0	360.9	17.05%		
		D90	789.7	956.4	6.37%		
Sample 3	E4-E30 (50-50)	D10	2.1	2.1	-0.76%	0.4%	0.8%
		D50	7.5	7.5	-0.31%		
		D90	39.0	38.8	-0.15%		
Sample 4	E4-E0.5 (20-80)	D10	5.0	3.8	-43.35%	17.6%	43.3%
		D50	764.1	728.6	-9.08%		
		D90	1331.4	1341.6	0.38%		
Sample 5	E10-E80 (80-20)	D10	5.5	5.3	-8.38%	4.3%	8.4%
		D50	13.0	12.1	-3.86%		
		D90	394.3	101.8	-0.76%		
Sample 6	E10-E150 (20-80)	D10	12.1	11.9	-2.33%	2.4%	3.7%
		D50	119.7	117.8	-3.72%		
		D90	172.2	168.6	-1.12%		
Sample 6 R1	E10-E150 (20-80) R1	D10	12.6	12.1	-3.86%	1.9%	3.9%
		D50	119.6	118.7	-1.69%		
		D90	170.3	169.1	-0.17%		
Sample 7	E10-E0.1 (50-50)	D10	6.1	6.2	5.48%	7.8%	15.7%
		D50	134.4	145.8	2.28%		
		D90	491.1	1004.0	15.72%		
Sample 8	E30-E0.5 (50-50)	D10	21.7	23.3	13.54%	6.3%	13.5%
		D50	782.1	808.7	3.85%		
		D90	1439.1	1472.9	1.48%		
Sample 8 R1	E30-E0.5 (50-50) R1	D10	21.1	22.8	14.32%	6.7%	14.3%
		D50	742.5	768.9	5.63%		
		D90	1406.3	1401.7	-0.18%		