Measurement of the Axial Dispersion Coefficient of Powders in a Rotating Cylinder: Dependence on Bulk Flow Properties

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Abstract

Rotating drums are encountered in numerous industrial applications, including blenders, rotary calciners, impregnators, coaters, granulators, and cement mixers. In all of these devices, the rotation of the drum is used to engender mixing of the granular material in the radial direction. Axial mixing, because of its significantly lower rate, can also have an impact on the process performance, especially when control of residence time is important. Typically, the particle dynamics in rotating drums are quantified as a function of process conditions, such as rotation speed, fill level, and cylinder size. Particle properties are also important, but previous studies have largely been limited to the effects of particle size. In this work, the quantification of the axial particle dynamics has been expanded to include the effect of bulk flow properties by studying a number of cohesive powders. Fick's second law was found to describe the axial dispersion behavior of cohesive particles. Therefore, changes in behavior can be characterized using the axial dispersion coefficient. The effect of material flow properties was found to be statistically significant; the flowability of the material (as measured using bulk flow properties) correlated significantly to the axial dispersion coefficient. Partial least squares was used to determine that 95% of the variation observed in the axial dispersion coefficient measurement can be explained using particle size, compressibility, and shear cell measurements.

Keywords

Powder flow, cohesion, flowability, mixing, granular dispersion

1. Introduction

Predicting the flow behavior of powders and granular materials in a solids processing operation is difficult [1]. To date, a general, theoretical approach describing powder flow, mixing, and segregation has yet to be developed [2]. As a result, the results of numerous experimental measurements and empirical correlations are used to characterize the flow behavior of powders and granular material in specific geometries [3]. These results are often used as the basis for the design and scale-up of manufacturing processes.[4, 5] This approach is utilized across industries for many unit operations; e.g. agitated drying, fluidized bed coating, and blending in pharmaceutical manufacturing [6-11] and impregnation, drying, mulling, and calcinations in catalyst manufacturing [12, 13]. Many characterization techniques exist to attempt to describe the flow behavior of a powder in a processing step, each technique characterizes the powder flow behavior differently [14]. In addition, each unit operation presents a unique processing environment exposing the material to various levels of stress, altering the flowability of the material [15]. Despite these differences, which often lead practitioners to develop internal characterization standards, the different powder flow characterization methods can be highly collinear [16], which in turns leads to superfluous or redundant measurements. Therefore, the selection of the most useful technique(s) for a given process is often unclear and complex [17].

The horizontal rotating drum is commonly used for processing powders. It appears across industries and processes as rotary kilns, impregnators, cement mixers, coaters, blenders, etc.[18, 19] For batch operations, the drum or cylinder is placed in a horizontal position and rotated about its axis. For continuous operations the drum is often arranged in an inclined configuration, typically by a few degrees [20]. There are two main observations that have informed studies of axial mixing of powders and granular materials: that Fick's second law can be used to describe

the axial dispersive mixing [21], and that the axial dispersion coefficient can be related to the residence time distribution via the Peclet number, known as the Taylor dispersion model [22-24]. As a result, characterization of the axial dispersion coefficient in batch systems can be related to that in continuous flow systems. These observations have been utilized by many authors to study horizontal rotating drums. Rotary kilns have been the focus of many recent studies [25-29] as have continuous blenders [30-33].

The axial dispersion coefficient has been measured in rotating drums to characterize axial mixing using Fick's second law [34]. Experimentally, tracer particles are introduced and the spatial distribution of the tracer particles throughout the bed is monitored as a function of time, to yield a measurement of the axial dispersion coefficient. Several initial conditions can be used, requiring slightly different mathematical treatment of the results to estimate the dispersion coefficient. Parker *et al.* [35] used a single particle method, tracking the movements of a single radioactively labeled tracer particle. Publications by Hogg *et al.* [36], Rao *et al.* [37], Cahn and Fuerstenau [38], Carley-Macaulay and Donald [39], and Wightman and Muzzio [40] described studies where one half of the drum was initially filled with tracer particles. Hogg *et al.* [41] began experiments with a thin band of tracer particles at one end of the drum and Shoji *et al.* [42] used a thin band near the center of the drum. Singh [43] started with the tracer particles in a wide band at the center of the drum; this is also the initial condition used in the work reported in this article.

These publications have studied the effect of several process parameters on the axial dispersion coefficient in a rotating drum, including the rotation rate, fill level, and the incline of the drum. In batch systems, it has been found that the axial dispersion coefficient increases with increasing rotational speed [37, 44], and an inverse relationship between the fill level and axial dispersion

coefficient has been observed; the coefficient decreases with increasing fill level [44]. Abouzeid *et al.* [45], Hehl *et al.* [46], and Njeng *et al.* [47] have studied the effect of process parameters in continuous flow systems and found that the residence time decreases with an increase in rotational speed, drum incline, or feed rate (or, alternatively, an decrease in the fill level).

The effect of particle and bulk flow properties have been considered in several published works. Studies on the effect of particle size report conflicting results [33, 37, 43, 45]. Rutgers found that needle-like particles give lower axial dispersion coefficients than spherical particles [44]. Rao *et al.* [37] found that increasing surface roughness results in larger coefficients. The ratio of particle-particle friction to particle-wall friction was related to the mixing time by Woodle and Munro [48]. In mixing in high dilution systems with a tracer different from the bulk material, Orr and Shotten found that the ability to mix is dependent on the tensile strength of the bulk material [49]. There have also been equations developed based on continuous flow experiments that relate the axial dispersion coefficient to the angle of repose and the bulk density [50, 51]. However, these equations predict an axial dispersion coefficient of zero in “no-net-flow”, i.e. batch, systems; which is contrary to experimental observations. However, as of yet, there has not been a systematic study on the effect of cohesive forces, as characterized by bulk properties, on the axial dispersion coefficient.

This paper examines the effect of bulk flow properties on axial mixing. The axial dispersion coefficient was measured for five materials with a range of bulk flow properties. The rest of the paper is organized as follows. The materials characterized and the experimental method used to measure the axial dispersion coefficient are described in Section 2. Results and discussion are presented for the axial dispersion coefficient as a function of flow properties (Section 3). The discussion of results includes the use of statistical analysis to determine the statistical

significance of the material type. It was found that the material type was statistically significant. Using partial least squares regression, it was also found that, for the materials studied here, the variation in the axial dispersion coefficient can be mostly explained by measurements from three flow property characterization methods: particle size, compressibility, and the shear cell. Conclusions are presented in Section 4.

2. Materials and Methods

2.1 Materials

The axial dispersion coefficient of five powders was measured: Lactose Monohydrate N.F. Regular (Forremost, Rothschild, WI, USA), Vivapur 102 (JRS Pharma, Rosenberg, Germany), Compap L APAP (Mallinckrodt, Greensville, IL, USA), Zeolite Y (Zeolyst International, Conshohocken, PA, USA), and fluidized cracking catalyst (W.R. Grace & Co., Columbia, MD, USA). These powders were chosen as they are industrially relevant and represent a wide range in flow behavior. The materials and their properties are listed in Table 1. The particle size distribution was measured using a Beckman-Coulter LS 13 320 Series Tornado Dry Powder System laser diffraction particle size analyzer (Pasadena, CA, USA). The cohesion, flow function coefficient, compressibility, conditioned bulk density, and permeability were measured using the FT4 powder rheometer (Freeman Technology, Tewkesbury, Gloucestershire, UK). The FT4 methods are described in detail by Freeman [52, 53]. The cohesion and flow function coefficient were measured using the shear cell at an initial consolidation stress of 3kPa. The compressibility (percent volume change upon compression) and permeability (in terms of pressure drop) were measured using the vented piston at an applied normal stress of 15kPa, as is typical for these characterization techniques. The flow index was measured using a gravitational

displacement rheometer (GDR) as detailed by Alexander *et al.*[54] and Faqih *et al.*[55-57]. A smooth acrylic cylinder was used for each material, except for the fluidized cracking catalyst, where a ribbed cylinder was used to mitigate slumping (which is further discussed in Section 3).

2.2 Methods

2.2.1 Visible Light Spectroscopy

This work utilized a tracer material to measure the axial mixing behavior, as is typical of this type of study. In this case, the self-diffusion was of interest and so the material being studied, dyed to a darker color, served as the tracer. The concentration of the dyed material in a sample was measured using a color probe (X-Rite VeriColor Spectro 450, Grand Rapids, MI), i.e. visible light spectroscopy. This method is described in detail by Emady *et al* [58]; a brief summary is given here. The surface of a sample with an unknown concentration of dyed material was exposed to light in the visible spectrum. The intensity of the light reflected from the sample surface was proportional to the concentration of dyed material in the sample. Using a calibration curve, the concentration of dyed material in samples was measured. The calibration curve with error bars (the span of which is less than the span of the data point markers) for lactose monohydrate is shown in Figure 1. The experimental points were fitted with a 4th order polynomial to obtain the calibration curve. The following equation describes the calibration curve with a coefficient of determination of 0.9982 and p-value less than 0.01.

$$c = 5 \times 10^{-7} R^4 - 3 \times 10^{-4} R^3 + 6.4 \times 10^{-2} R^2 - 5.1 R + 153.2$$

where c was the mass concentration of dyed material in percent and R was the reflectance. Similar calibration curves were obtained for the other materials.

2.2.2 Experimental Setup

The particle mixing dynamics within a rotating cylinder was characterized in terms of the axial dispersion coefficient. An acrylic cylinder 30 cm long with an inner diameter of 9.5cm was used. The cylinder was loaded to a 35% fill level with a wide band initial condition, as illustrated in Figure 2. The cylinder was divided into three sections of similar width along the axial dimension using physical dividers. Subsequently, the first and third sections were loaded with undyed material, and the second (middle) section with dyed material (acting as the tracer). After filling to 35% by volume, the dividers were removed. Variability in the initial loading of the cylinder was a potential source of error in the experiment. Error in the placement of the physical dividers may translate to error in the width and lateral position of the band of dyed material. Further, the fill level may vary between sections, which could translate to error in the ratio of dyed to undyed material. Therefore for each material, three experimental replications were performed to consider both the within-group variability and between-group variability.

The cylinder was placed on rollers and rotated at 25rpm for a total of 30 minutes. Samples, 2.5mL in volume, were taken every 3 cm along the axial dimension at 5, 10, 15, 20, 25, and 30 minutes of mixing. The samples were collected by opening a portion of the end of the cylinder (so as to not disturb the powder) and then using a scoop to sample the powder from the bed surface. As samples were removed from the cylinder, the fill level was effectively decreased. Sampling ceased once 10% of the initial volume was removed, limiting the effect of the change in fill level. This removal limit was reached after three time points, therefore two independently filled runs were used for each experiment. The first run was sampled at 5, 10, and 15 minutes. A fresh run was then initially rotated for 20 minutes and sampled at 20, 25, and 30 minutes of mixing. This was repeated three times for each material. The visible light spectroscopy method

described in Section 2.2.1 was used to measure the concentration of dyed material in each sample. Concentration profiles as a function of axial position for each time point sampled were subsequently generated.

2.2.3 Axial dispersion coefficient calculation

As previously discussed, it has been suggested that the axial mixing of cohesion-less spheres in a rotating cylinder can be described using the functional form of Fick's second law [21].

$$\frac{\partial c}{\partial t} = D_{ax} \frac{\partial^2 c}{\partial x^2}$$

where c was the concentration of tracer (in this case, dyed material), t was the time, x was the axial position, and D_{ax} was the axial dispersion coefficient. The applicability of this approach to cohesive powders is investigated here. This equation was solved numerically using the wide band initial condition and no-flux boundary conditions using finite differences, as done by Singh [43]. To further mitigate some of the error introduced in the initial loading of the cylinder, the initial axial position and width of the wide band were allowed to vary. The wide band location and width that minimized the residual between the experimental concentration profile and numerical solution at 5 and 20 minutes of mixing, corresponding to the first time point in each cylinder, was found. Using this initial band location and width, the axial dispersion coefficient that minimizes the residual sum of squares between the experimental concentration profile and numerical solution (Eqn 2) was identified for each time point for which samples were taken (every 5 minutes up to 30 minutes of mixing). MATLAB R 2014b (The Mathworks Inc., US) was used to find the optimal numerical solution in this study. The residual between the experimental concentration profile and numerical solution was computed as follows:

$$ss_{res} = \sum_i (y_i - f_i)^2$$

where ss_{res} was the residual sum of squares, y_i were the experimental values, and f_i were the numerical values. Since each experiment was repeated three times, and each experiment included six concentration profiles from different time points, the estimated axial dispersion coefficient for each material was an average of 18 measurements. While we would expect that the axial dispersion coefficient would remain constant from time point to time point, our method computed a different axial dispersion coefficient for each time point in order to get an estimate of the variability of the axial dispersion measurement. We were therefore able to get a mean axial dispersion as well as an estimate of the standard error of the measurement.

3. Results and Discussion

3.1 Concentration profiles

Concentration profiles for each time point sampled from one of the three experimental runs are presented as an example in Figure 3 for lactose monohydrate. As shown in the Figure, the concentration of dyed material is higher in the center of the cylinder than at the edges. This was expected since the initial wide band of dyed material was centrally located along the axial dimension of the cylinder. This figure shows the evolution of mixing with time. As time progresses, the maximum concentration (located at about 15 cm) decreases while the concentration at the ends of the cylinder (0 and 30cm) increases. As additional time elapsed and the system approached a well-mixed state, the profile would approach a flat line at a concentration of 0.33. Qualitatively similar results are presented in Figure 4 for Compap L APAP. However, differences in mixing behavior were observed from the concentration profiles

as a function of time. For example, when comparing the lactose monohydrate (Figure 3) and Compap L APAP (Figure 4) results, the profiles for lactose monohydrate were flatter than those for Compap L APAP at the same time point. From this observation, it can be concluded that the lactose monohydrate would reach a well-mixed state faster than the Compap L APAP. As the axial dispersion coefficient describes the rate at which a system mixes, it would be expected that the axial dispersion coefficient for lactose monohydrate would be greater than that for Compap L APAP. As detailed below, this was indeed the case. Concentration profiles as a function of axial position for 5, 10, 15, 20, 25, and 30 minutes were measured three times for the three other materials (not shown). Similar features to those observed in Figures 3 and 4 were observed for the other materials.

3.2 Axial dispersion coefficient

The axial dispersion coefficient defined by Fick's second law: for the experimental concentration profiles of lactose monohydrate (Fig 3B) and Compap L APAP (Fig. 4B) are shown in Figures 5 and 6, respectively, as examples of typical results. The numerical fits are in reasonably good agreement with the experimental data describing the evolution of mixing over time. Similar observations were made for all experiments carried out for each material.

The D_{ax} values corresponding to the numerical fits for lactose monohydrate shown in Figure 5 are 0.023, 0.021, and 0.02 cm^2/s at 20, 25, and 30 minutes of mixing, respectively. The D_{ax} values corresponding to the numerical fits for Compap L APAP shown in Figure 6 are 0.012, 0.012, and 0.012 cm^2/s at 20, 25, and 30 minutes of mixing, respectively. As expected from comparing the concentration profiles of the lactose monohydrate and Compap L APAP, the D_{ax} values of the lactose monohydrate were higher than those for Compap L APAP, indicating faster

mixing. Similar results were obtained at 5, 10, and 15 minutes. It is concluded that Fick's second law can be used to describe the dispersive mixing in cohesive systems.

3.3 Axial dispersion coefficients for cohesive materials

The axial dispersion coefficient was estimated for each of the five materials studied here: Zeolite, lactose monohydrate, Compap L APAP, Vivapur 102, and FCC. The values obtained at each of the six time points for an experiment were averaged (each time point is considered a replicate measurement of the axial dispersion coefficient). Each experiment was repeated three times and an overall average axial dispersion coefficient for each material was computed as the average of 18 measurements. The average value of the dispersion coefficient, and its standard error for the 18 measurements (six time points, three repeats), are tabulated in Table 2. The axial dispersion coefficients range from 0.016 to 0.083 cm²/s, which is in agreement with values previously reported for granular materials. In the review by Sherritt *et al.* experimental results for the axial dispersion coefficient ranged from 0.001 to 10 cm²/s [34]. For comparison, it is noted that these coefficients are on the same order of magnitude of many gas-gas diffusion coefficients, and are larger than many liquid-liquid diffusion coefficients [59]. FCC had the smallest coefficient, 0.016 cm²/min, and Zeolite had the largest coefficient, 0.83 cm²/min, a five-fold increase over that for FCC.

The relationships between the axial dispersion coefficients and the various material properties in Table 1 were investigated. In Figure 7, the axial dispersion coefficient is plotted as a function of the flow function coefficient (FFC), measured using the shear cell. The flow function coefficient is often used to characterize the flowability of a material, where a larger flow function coefficient corresponds to a more free-flowing material. The materials can be ranked in terms of flow

function coefficient from lowest to highest as follows: Zeolite, lactose monohydrate, Compap L APAP, Vivapur 102, and FCC.

Contrary to intuition, Figure 7 shows that as the flow function coefficient increases, corresponding to an increase in flowability, the axial dispersion coefficient decreases. It can be seen that the error bar for the axial dispersion coefficient of the poorest flowing material (Zeolite) is the largest. The next material in terms of flowability is lactose monohydrate. A relatively large decrease in axial dispersion coefficient and standard error of the mean is observed. The next material in terms of flowability is Compap L APAP. There is less of a decrease in axial dispersion coefficient and standard error. The next two materials in terms of flowability are Vivapur 102 and FCC and these materials show a much smaller decrease in axial dispersion coefficient. The results for axial dispersion can be separated into two groups: the poorest flowing materials (Zeolite and lactose monohydrate) and the better flowing materials (Compap L APAP, Vivapur 102, and FCC). It is interesting that in spite of the fact that Compap L APAP, Vivapur 102, and FCC have quite different flow function coefficients, their axial dispersion coefficients are relatively similar.

In Figure 8 the axial dispersion coefficient is shown as a function of compressibility percent at an applied normal stress of 15kPa, measured using the FT4 compressibility test. Typically, a lower compressibility percent corresponds to a material with lower cohesion and better flowability. Behavior similar to that observed previously for the FCC in Figure 7 was observed for the compressibility percent in Figure 8, however, the relative order of Compap L APAP and Vivapur 102 reversed. In Figure 8, it can be seen that all the materials have significant differences in compressibility and again fall into two groups in terms of axial dispersion coefficient.

The particle size, in terms of median (d_{50}) and fines cut-off (d_{10}), also correlates with the axial dispersion coefficient, as seen in Figures 9 and 10. Smaller particle sizes often exhibit larger cohesive forces and worse flowability. It is observed in Figures 9 and 10 that the axial dispersion increases with decreasing values of d_{50} and d_{10} . Behavior similar to that observed previously for the FFC in Figure 7 and compressibility percent in Figure 8 was observed for the d_{10} and d_{50} . However, the relative order of Compap L APAP, Vivapur 102, and FCC varied. In Figures 9 and 10, it can be observed that all materials have significant differences in d_{10} and d_{50} and again fall into two groups in terms of axial dispersion coefficient. Zeolite and lactose monohydrate fall into the same group (in terms of axial dispersion coefficient) and appear in the same order for the flow function coefficient, compressibility percent, d_{10} , and d_{50} . Vivapur 102, Compap L APAP, and FCC belong to the same group but appear in different orders. Not every material property correlates with the axial dispersion coefficient, however. For example, the permeability does not exhibit a clear relationship with the axial dispersion coefficient as seen in Figure 11. While only some of the material properties have a relationship to the axial dispersion coefficient, the ones that do exhibit the same relationship. Summarily, the flow function coefficient, d_{10} , and d_{50} decrease with increasing axial dispersion coefficient while the compressibility percent increases with increasing axial dispersion coefficient. That is, as the flow properties worsen, the axial dispersion coefficient increases. Put another way, cohesive, poorly flowing materials mix more quickly than less cohesive materials.

This surprising observation, previously unreported to the best of our knowledge, is likely due to the manner in which cohesive granular materials flow in a rotating cylinder. The experimental conditions were chosen such that the type of bed motion exhibited in each of the experiments was in the cascading regime. As described by Mellman [60], powder bed motion in which the

295 bed does not slip at the drum wall, circulates continuously, and has an arched bed surface, shows
296 a flow pattern that can be classified as cascading motion. There is an active surface layer and a
297 relatively inactive core. Particles against the drum wall move with the wall until the maximum
298 height of the bed is reached. The particles then fall down the free surface of the bed along a
299 main flow pattern consisting of parallel “streamlines” orthogonal to the cylinder axis.
300 Superimposed to this “main flow” is a second, deviatory flow parallel to the cylinder axis,
301 composed of random deviations due to particle-particle collisions. Intuitively, the distance
302 travelled by a particle due to these collisions should be proportional to the particle size.

303 Cohesion-less particles tend to fall individually down the free surface. However, in cohesive
304 systems, the particles do not flow individually, but as assemblies or “chunks” of material. The
305 size of the particle assemblies is dependent on the cohesive forces; the larger the attraction
306 between particles, the larger the assemblies that can form. Therefore, the relevant characteristic
307 length for the deviatory flow may not be the particle diameter, but instead the size of the formed
308 assemblies. Since the cohesive, poorly flowing materials, which would form larger assemblies
309 and therefore have a larger effective length, exhibit larger axial dispersion coefficients, these
310 results follow the trend observed previously in the literature of surface velocities and axial
311 dispersion coefficient increasing with particle diameter [33, 61].

312 To verify the above concept, flow index measurements taken using the Gravitational
313 Displacement Rheometer (GDR) were compared to the axial dispersion coefficient
314 measurements. The GDR measures the size of the avalanches, or “chunks,” that fall during flow
315 of a granular material in a rotating drum [54]. A larger flow index indicates larger avalanches.
316 From the above discussion, it would be expected that the axial dispersion coefficient would
317 increase with increasing flow index. Moreover, the flow index in the GDR is measured for

powder that is flowing in the fully dilated state, i.e., in the same state as in the rotating cylinder. Behavior similar to that observed previously for the FFC, compressibility, d_{10} , and d_{50} (Figures 7-10) was observed for the flow index. In Figure 12, it can be observed that the materials have significant differences in flow index and again fall into two groups with respect to the axial dispersion coefficient. The three left-most points are Vivapur 102, Compap L APAP, and FCC, forming the same group as observed for the FFC, compressibility, d_{10} , and d_{50} . While their respective flow index values are different, they had similar axial dispersion coefficient values. The lactose monohydrate had a larger axial dispersion coefficient and belongs to the second group of poorly-flowing materials.

As mentioned previously, the flow index is typically measured in a smooth, acrylic cylinder. However, during the testing for the fluidized cracking catalyst, the particles flowed in the surging regime. Due to a low coefficient of wall friction, the powder bed would travel up with the cylinder wall to a critical angle and then slide down as a whole. This sliding movement registered as an avalanche. For this material, a ribbed cylinder was used to mitigate the slumping. The ribs increased the wall friction and prevented the bed as a whole sliding against the wall. The flow index of the fluidized cracking catalyst measured using the ribbed cylinder is shown in Figure 12.

The flow index measurement for the Zeolite, 16.34, was lower than expected and is not included in Figure 12. It was observed during Flow Index testing that the Zeolite particles would form spherical agglomerates, as seen in Figure 13. The material prior to testing is shown on the left and after testing on the right. The agglomerates likely formed due to electrostatic forces caused by friction with the Plexiglas cylinder.[62] Agglomerates were not observed during the axial dispersion measurement. Therefore, agglomeration is a phenomenon introduced by the flow

index measurement technique that does not occur during the other experiments. For the cases studied here, the flow index measurements support the concept that the relevant characteristic length for the axial mixing in cohesive systems should be the avalanche size as opposed to the particle diameter.

3.4 PLS regression

It is clear that the material “flowability” (as characterized by the different methods) has a statistically significant effect on the axial dispersion coefficient. It has also been observed that, for the materials studied here, some material properties correlate with the axial dispersion coefficient while some do not. A statistical approach to identifying the flow property or properties whose variation best predicts changes in the axial dispersion coefficient was explored. This approach may lead to a reduced number of material flow properties that are needed for the characterization of future materials and/or experimental conditions, saving time, money, and resources, helping the achievement of an improved understanding of the axial mixing dynamics in a rotating cylinder. The statistical approach used here was partial least squares (PLS) regression.

The PLS regression technique, also known as “projection to latent structures”, can be useful in situations where the goal is to identify and quantify relationships between predictors and observations. This is particularly useful in cases where the predictors are collinear and their number exceeds that of the observations. Here, there was a set of nine material flow properties acting as the predictors and one observation, the axial dispersion coefficient. While each property is measured in a different manner, they are all measurements of the cohesive forces. It would then be expected that a set of material properties would be highly collinear. Therefore,

363 PLS regression was a suitable method to employ here. Software Unscrambler X 10.2 (Camo, Oslo,
364 Norway) was used in this study for the multivariate analysis.

365 A two factor non-hierarchical PLS model was fitted to the x-block data consisting of nine material
366 properties for the five materials and the single y-variable, the axial dispersion coefficient. The
367 data were mean-centered and scaled. The cumulative percent variance explained by the two
368 factors was 94%, shown in Figure 14. The first factor explained 90% and the second factor
369 explained an additional 4%. Good parity was observed between the predicted and actual y-
370 values (axial dispersion coefficients), as shown in Figure 15. The root mean square error was
371 0.09. The regression coefficients are depicted in Figure 16. The coefficients represent the
372 contribution of each material flow property to the prediction of the axial dispersion coefficient,
373 with higher coefficients indicating larger contributions. Positive coefficients indicate a positive
374 correlation and negative coefficients indicate an inverse proportionality. Those with little or no
375 weight are not very useful in the prediction. The material property with the largest contribution
376 was the compressibility. The properties with the smallest contributions were the permeability
377 and the flow index. It was expected that the flow index would be a poor predictor of the axial
378 dispersion coefficient due to the smaller than expected measurement for the Zeolite. To verify
379 that these two independent measurements could be removed from the model without losing
380 predictive capability, a new PLS regression model was fitted using the remaining seven material
381 flow properties. The cumulative percent variance explained by two factors was 95%. The first
382 factor explained 91% and the second factor an additional 4%. The root mean square error of the
383 predicted versus actual y-values remained at 0.09. The coefficient weights, as depicted in Figure
384 17, also remained similar. The compressibility had the largest weight. The compressibility and
385 cohesion were positively correlated with the axial dispersion coefficient while the d_{10} , d_{50} , d_{90} ,

conditioned bulk density, and flow function coefficient were negatively correlated. Therefore, it can be concluded that the axial dispersion coefficient can be described using these seven material flow properties. These material flow properties are measured by three characterization methods. The d_{10} , d_{50} , and d_{90} are particle size parameters. The conditioned bulk density and compressibility at 15kPa are both measured using the FT4 compressibility test. The cohesion and flow function coefficient are measured by the FT4 shear cell. The parameters obtained using these three characterization methods can predict 95% of the variation observed in the axial dispersion coefficient.

4. Conclusions

In this study, the axial mixing behavior of cohesive systems was examined in a rotating drum. The self-mixing dynamics of five materials with a range of flow properties was characterized in terms of the axial dispersion coefficient. It was found that Fick's second law accurately describes dispersive mixing in both freely flowing and cohesive systems. The coefficients for the five materials varied five-fold and were within the range observed by previous work reported in the literature.

Keeping the experimental conditions constant, it was also found that the material type had a statistically significant effect on the axial dispersion coefficient. That is, the strength of the cohesive forces affects how quickly a cohesive system mixes. Analysis of the material properties revealed that the axial dispersion coefficient was correlated to the cohesive forces. Unexpectedly, more cohesive, poorly flowing materials mix more quickly than less cohesive materials. This is likely due to the "chunks" formed as the material flows (supported by the flow index measurements). For the materials studied here, measurements from three characterization

408 methods, particle size distribution, compressibility, and shear cell, can predict 95% of the
409 variation observed in the axial dispersion coefficient.

410 These results are based on five materials at a single experimental condition. Thus, further work
411 is needed at additional experimental conditions to generalize our results across multiple scales.
412 A better understanding of axial mixing behavior in rotating drums is important to the successful
413 design and scale up of processes that contain a mixing component, e.g. mixing, calcination,
414 coating, etc. This paper also demonstrates a statistical approach for identifying relevant material
415 flow properties for specific processes. The approach can easily be generalized and applied to
416 additional material properties and unit operations.

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558

559 Figure captions

560 Figure 1: Calibration curve with error bars for lactose monohydrate with a 4th-order polynomial
561 fit.

562 Figure 2: Initial loading condition of cylinder: 1/3 of the length loaded with undyed material, 1/3
563 with dyed material (tracer), and 1/3 with undyed material

564 Figure 3: Concentration profiles of dyed lactose monohydrate from one of the three experiment
565 replications at (A) 5, 10, and 15 minutes and (B) 20, 25, and 30minutes of mixing.

566 Figure 4: Concentration profiles of dyed Compap L from one of the three experiment replications
567 at (A) 5, 10, and 15 minutes and (B) 20, 25, and 30minutes of mixing.

568 Figure 5: An example of finding the numerical solution from the experimental results. The
569 experimental concentration profile from Figure 3B (shown as points) of dyed lactose
570 monohydrate was used to find the optimal numerical solution of Fick's Second Law (shown as
571 lines) at 20, 25, and 30min of mixing.

572 Figure 6: An example of finding the numerical solution from the experimental results. The
573 experimental concentration profile from Figure 4B (shown as points) of dyed Compap L APAP
574 was used to find the optimal numerical solution of Fick's Second Law (shown as lines) at 20, 25,
575 and 30min of mixing.

576 Figure 7: The axial dispersion coefficient as a function of the flow function coefficient as
577 measured by the FT4 shear cell.

578 Figure 8: The axial dispersion coefficient as a function of the compressibility at an applied
579 normal stress of 15kPa as measured by the FT4 powder rheometer.

580 Figure 9: The axial dispersion coefficient as a function of d_{10} .

581 Figure 10: The axial dispersion coefficient as a function of d_{50} .

582 Figure 11: The axial dispersion coefficient as a function of the permeability at an applied normal
583 stress of 15kPa as measured by the FT4 powder rheometer.

584 Figure 12: The axial dispersion coefficient as a function of the flow index as measured by the
585 GDR.

586 Figure 13: Zeolite before GDR testing (left) and after GDR testing (right).

587 Figure 14: Cumulative percent explained variance by PLS model.

588 Figure 15: Predicted versus reference parity plot for the axial dispersion coefficient

589 Figure 16: Regression coefficients of PLS model

590 Figure 17: Regression coefficients of reduced PLS model

Figure 1

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Figure 10

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Table 1: Particle size and flow properties

Material	d ₁₀ (μm)	d ₅₀ (μm)	d ₉₀ (μm)	Compressibility @ 15kPa	Permeability @ 15kPa	FFC	Flow Index
Zeolite	0.80	3.23	11.51	0.35	4.85	4.50	16.34
FCC	45.95	83.61	139.12	0.02	3.66	10.00	39.77
Compap L APAP	74.29	178.29	316.37	0.05	2.17	7.48	34.93
Vivapur 102	25.64	123.59	198.93	0.13	1.71	8.56	22.70
Lactose monohydrate	10.20	70.07	150.73	0.20	13.07	6.05	56.18

Table 2: Axial dispersion coefficients for each material tested

Material	Dax (cm ² /s)	Standard Error
FCC	0.016	0.004
Vivapur 102	0.022	0.003
Lactose Monohydrate	0.039	0.010
Zeolite	0.083	0.018
Compap L APAP	0.024	0.005