Modeling and Validation of Percolation Segregation of Fines from Coarse Mixtures during Shear Motion†

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Abstract

Segregation negatively impacts the product quality and depends on physical and mechanical properties of particulate materials. Size is the most dominant parameter contributing towards fines percolation segregation from mixtures with size distribution. A continuum theory-based convective and diffusive model was developed and validated to study time-dependent percolation segregation of fines. In addition, to scale-up the results, a mechanistic theory-based dimensional analysis approach was used to incorporate physical and mechanical properties of particulates in a time-independent model. In dimensional analysis model, size, shape, density, size ratio, mixing ratio, strain rate, strain, and bed depth were included.

The results showed that the time-dependent convective and diffusive model predicted the segregated mass of fines within the 95% confidence interval of measured fines for size ratios 2.4:1.0 and 1.7:1.0 at strains of 6% and 10%. Dimensional analysis results showed that the Coefficient of Variation (CoV) of the modeled values with respect to the experimental values were 18%, 15%, and 11%, respectively, for binary mixtures of urea and potash at strains of 2%, 6%, and 10% and strain rate of 0.25 Hz.

Keywords: Percolation segregation, constitutive models, dimensionless analysis, geometrical similitude, convective/diffusive, validation, scale-up

1. Introduction

Particulate materials are handled, stored, mixed, and processed in various industries such as agriculture, cosmetic, food, metal and metallurgy, nutraceutical, and pharmaceutical. Segregation occurs in particulates during the mentioned unit operations and negatively impacts the quality of products. Segregation in particulate materials occurs due to differences in the constituents’ physical and mechanical properties (Rosato et al., 2002). Although segregation is governed by several parameters; however, simultaneously studying the effects of all parameters is not feasible due to the lack of fundamental understanding of their complex and nonlinear interactions. To gain greater and deeper insights, dominant variables should be identified and studied for different process parameters. Bridgwater and co-workers were the pioneers in identifying the dominant parameters and mechanisms of segregation and reported that size is the dominant parameter (Bridgwater et al., 1978 and Bridgwater, 1994). In practice, segregation has been quantified using coefficients, described using mechanisms, and analyzed using models (Rosato and Blackmore, 2000). Of the three, to model the segregation process for a specific mechanism is preferred so that the model could be applied to different operating conditions. To date, thirteen segregation mechanisms have been identified based on different operating conditions, i.e., trajectory, air current, rolling, sieving and sifting, impact, embedding, angle of repose, push-way, displacement or floating, percolation, fluidization, agglomeration, and, concentration driven displacement (Mosby et al., 1996; Salter, 1998; de Silva et al., 2000). Of the thirteen, percolation segregation is widely observed when materials are han-
Percolation segregation in particulates is also affected by the mechanical conditions under which materials are handled (Tang and Puri, 2005 and Jha and Puri, 2009). Percolation segregation is defined as the migration of fine particles through a bed of coarse particles during gravity, shear, and vibration motions when subjected to dynamic conditions (Vallance and Savage, 2000). However, most of the studies were conducted for specific operating conditions using ideal materials (glass beads) and a few real-world materials (commonly used in industries) (Tang, 2004), few models were also developed for time-dependent process using ideal and real-world materials (Duffy and Puri, 2002). Tang and Puri, and Duffy and Puri used point feed and a layer of feed of fines in the coarse particle bed, respectively. Tang and Puri (2005) used point feed of fines in the coarse particle bed using real-world materials (poultry feed), which is not the common approach to handle particulates in industries. To overcome the limitations of previous studies, percolation segregation during shear motion was selected for modeling due to its wide application in industries. Segregation in particulates can be modeled by three approaches: continuum models, kinetic theory models, and discrete models (Moakher et al., 2000). Each modeling approach has advantages and disadvantages, continuum models consider the conservation principles of mass, momentum, and energy but neglect the discrete nature of particles; kinematic models consider the principle of interacting grains and colliding molecules in a dense gas but it is limited to the surface of an agitated granular mass; and discrete models consider the constituent grains to be distinct and apply the prescribed rules but limited to relatively small systems (Khakhar et al., 2001).

Tang (2004) developed a mechanistic theory which included the advantages of all the three mentioned modeling approaches. Mechanistic theory incorporated the continuum and discrete element theory by including the segregation potential of fines sizes and kinetic theory by studying the falling path of fines in larger size ratios.

Based on the literature review, application of the convective-diffusive model developed by Duffy and Puri is limited to large size ratios and ideal materials. To overcome this limitation, the model proposed and developed in this paper is for small size ratios, which includes resistance offered by the screen placed at the bottom; a common situation in modern day industrial practices. Dimensional analysis model developed by Tang (2004) is limited to binary mixtures and limited number of mechanical conditions. To improve the capability of mechanistic theory based dimensional analysis model, numbers of size components were increased and several operating conditions were included. As a result, the dimensional analysis model is more robust and applicable for scale-up of process from bench-scale to other relevant scales. The developed dimensional analysis model includes size, shape, density, size ratio, mixing ratio, strain rate, strain, and bed depth.

2. Materials and Methods

The Primary Segregation Shear Cell (PSSC-II) described sufficiently in Tang and Puri (2005 and 2007) and modified by Jha and Puri (2009) was used in this study. Since binary size mixture is considered to be the foundation of multi-size and continuous mixtures; a time-dependent convective and diffusive model is presented for binary mixtures (Jha, 2008). Nine different binary size ratios of potash and six different size ratios of urea in different mixing ratios were used in modeling studies. Mechanistic theory based time-independent model includes size, shape, density, size ratio, mixing ratio, strain rate, strain, and bed depth. Dimensional analysis model is suitable for process scale-up.

2.1 Test material selection and parameter determination

Urea and potash were selected for studying percolation segregation due to their extreme shape and density among the three major raw ingredients: urea, potash, and phosphate, used in the manufacture of different fertilizer blends (Jha and Puri, 2009 and Jha et al., 2008). For segregation study, three parameters including material bed depth, particle bed strain, and strain rate were selected for operating PSSC-II based on published results (Tang and Puri, 2005 and 2007). Bed depth of 85 mm (shear box height = 100 mm) was used to represent percolation of fines within bagged fertilizers in normal orientation, i.e., depth direction along gravity, during conveying, handling, and transportation. The selected strain of 2%, 6%, 10% and strain rates of 0.25, 0.5, 1.0 Hz represent the unfilled bag volume (≤15%) and intensity of motion, respectively, experienced by the blend in the bag during processing operations (<10 Hz) (Vursavus and Ozguven, 2004). The tests at strain rate of 1.0 Hz...
were conducted only at strain of 6% for binary mixtures of urea and potash due to limitation of operation of PSSC-II at high shear rate. Based on plant visits, strains higher than 6% at shear rate of 1.0 Hz seemed high for the operation of fertilizer blend manufacturing and conveying (Jha, 2008).

Different coarse and fine particle size ranges of the test material were obtained using US standard sieve of $(2)^{\frac{1}{4}}$ series. Potash and urea were received from local fertilizer blend plant facilities. Three size ranges (3,350-4,000; 2,800-3,350; and 2,360-2,800 µm) were designated as coarse and while three smaller size ranges (2,000-2,360; 1,700-2,000; and 1,400-1,700 µm) were designated as fines in the present study (Table 1). Since size spread of urea was small compared with potash, fines size range 1,400-1,700 µm were not found in sufficient quantity, therefore, this fine size was not included in the segregation study of urea. Size ratio of binary mixture was defined as the ratio of mean size of coarse particles to mean size of fine particles. For convective and diffusive model development using potash, two size ratios for each coarse sizes 3,675 µm, 3,075 µm, and 2,580 µm were used: 2.4:1.0, 1.7:1.0; 2.0:1.0, 1.4:1.0; and 1.7:1.0, 1.2:1.0, respectively (Table 1). For urea, two size ratios for coarse size 3,675 µm, 3,075 µm, and 2,580 µm were used: 2.0:1.0, 1.7:1.0; 1.7:1.0, 1.4:1.0; and 1.4:1.0, 1.2:1.0, respectively (Table 1). Multi-size mixtures were used for dimensional analysis model development in addition to the materials used for convective diffusive model (Tables 1 and 2). For validation of convective and diffusive model, the size ratio 2.0:1.0 of potash was used at strains of 2%, 6%, and 10% and two strain rates of 0.25 and 0.5 Hz. For urea, two size ratios for coarse size 3,675 µm, 3,075 µm, and 2,580 µm used were 2.0:1.0, 1.7:1.0; 1.7:1.0, 1.4:1.0; and 1.4:1.0, 1.2:1.0, respectively, at strains of 2%, 6%, and 10% and strain rate of 0.5 Hz (Table 3). Conditions used for validation of dimensional analysis model are given in Table 4. Different mixing ratios (MR) were used for different size ratios based on weight proportion of different size (Tables 1, 2, 3, and 4) distributions found in low analysis such as 10-10-10 fertilizer samples collected from Commonwealth of Pennsylvania blend plants (Jha, 2008).

### Table 1  Experimental design for binary size mixtures for potash and urea

| Material | Strain Rate (Hz) | Coarse size (µm) | Fine Size (µm) | Size Ratio | Mixing Ratio | Number |
|----------|-----------------|-----------------|---------------|------------|--------------|--------|
| Potash   | 0.25 1.00**     | 3,675           | 1,550         | 2.4:1.0    | 50:50        | 6      |
|          |                 |                 | 1,850         | 2.0:1.0    | 37:63        |        |
|          |                 |                 | 2,180         | 1.7:1.0    | 37:63        |        |
| Potash   | 0.25 1.00**     | 3,075           | 1,550         | 2.0:1.0    | 63:37        | 6      |
|          |                 |                 | 1,850         | 1.7:1.0    | 50:50        |        |
|          |                 |                 | 2,180         | 1.4:1.0    | 50:50        |        |
| Potash   | 0.25 1.00**     | 2,580           | 1,550         | 1.7:1.0    | 60:40        | 6      |
|          |                 |                 | 1,850         | 1.4:1.0    | 60:40        |        |
|          |                 |                 | 2,180         | 1.2:1.0    | 60:40        |        |
| Urea     | 0.25 1.00**     | 3,675           | 1,850         | 2.0:1.0    | 37:63        | 4      |
|          |                 |                 | 2,180         | 1.7:1.0    | 37:63        |        |
| Urea     | 0.25 1.00**     | 3,075           | 1,850         | 1.7:1.0    | 37:63        | 4      |
|          |                 |                 | 2,180         | 1.4:1.0    | 37:63        |        |
| Urea     | 0.25 1.00**     | 2,580           | 1,850         | 1.4:1.0    | 37:63        | 4      |
|          |                 |                 | 2,180         | 1.2:1.0    | 37:63        |        |
| Total (replications) | | | | | 30 × 3 × 6 = 540 |
### Table 2  Experimental design for multi-size mixtures for potash and urea

| Material | Strain Rate (Hz) | Coarse size (µm) | Fine Size (µm) | Size Ratio | Mixing Ratio | Number |
|----------|-----------------|------------------|----------------|------------|--------------|--------|
| Potash   | 0.25            | 3,675            | 1,850          | 2.0:1.0    | 28:44:28:28 | 6      |
|          | 0.50            | 3,075            | 2,180          | 1.7:1.0:1.0 | 23:39:39    |        |
|          | 0.50            | 3,075+2,580      | 1,850          | 2.0:1.7:1.0 | 33:46:21    |        |
|          | 0.50            | 3,075+2,580      | 2,180          | 1.4:1.4:1.0 | 29:42:29    |        |
| Urea     | 0.25            | 3,675            | 1,850          | 2.0:1.0    | 22:39:39    | 4      |
|          | 0.50            | 3,075            | 2,180          | 2.0:1.7:1.0 | 17:28:38:17 |        |
|          | 0.50            | 3,075+2,580      | 2,180          | 2.0:1.7:1.0 | 13:25:37:25 |        |

Total (six replications)  \(12 \times 6 \times 3 = 216\)

*Strains of 2%, 6%, and 10%

### Table 3  Validation design for binary size mixtures for potash and urea

| Material | Strain Rate (Hz) | Coarse size (µm) | Fine Size (µm) | Size Ratio | Mixing Ratio | Number |
|----------|-----------------|------------------|----------------|------------|--------------|--------|
| Potash   | 0.25            | 3,675            | 1,850          | 2.0:1.0    | 37:63        | 2      |
|          | 0.50            | 3,075            | 1,850          | 1.7:1.0:1.0| 50:50        |        |
|          | 0.50            | 3,075+2,580      | 1,850          | 1.4:1.0:1.0| 63:37        | 2      |
| Urea     | 0.25            | 3,675            | 1,850          | 2.0:1.0    | 37:63        | 2      |
|          | 1.00            | 3,075            | 1,850          | 2.0:1.0:1.0| 50:50        |        |
|          | 1.00            | 3,075+2,580      | 1,850          | 2.0:1.0:1.0| 60:40        | 2      |

Total (three replications) \(12 \times 3 \times 3 = 108\)

*At strains of 2%, 6%, and 10%

### Table 4  Validation design for binary size mixtures for potash and urea

| Material | Strain Rate (Hz) | Coarse size (µm) | Fine Size (µm) | Size Ratio | Mixing Ratio | Number |
|----------|-----------------|------------------|----------------|------------|--------------|--------|
| Potash   | 0.5             | 3,675            | 1,550          | 2.4:1.0:1.0| 50:50        | 3      |
|          | 0.5             | 3,075            | 1,550          | 2.0:1.0    | 37:63        | 3      |
|          | 0.5             | 2,580            | 1,550          | 1.7:1.0:1.0| 63:37        | 3      |
| Urea     | 0.5             | 3,675            | 1,850          | 2.0:1.0    | 37:63        | 2      |
|          | 0.5             | 3,075            | 1,850          | 1.7:1.0:1.0| 37:63        | 2      |
|          | 0.5             | 2,580            | 1,850          | 1.4:1.0:1.0| 60:40        | 2      |
|          | 0.5             | 2,580            | 2,180          | 1.2:1.0:1.0| 60:40        |        |

Total (six replications) \(15 \times 6 \times 3 = 270\)

*Strain of 6%
Within each block, all treatments (replicate 6 = 6) were randomly assigned. Convective and diffusive model was validated with binary mixtures of urea and potash (Table 3). Developed dimensional analysis model was validated for binary size mixtures of urea and potash (Table 4). Ternary and quaternary size ratios were not available in the sufficient quantity; therefore, these were not included in validation in the present study, i.e., deferred to a future study. All tests were conducted in an environment-controlled laboratory with average temperature of 22 degree C ± 3 degree C and relative humidity less than 40%.

2.3 Convective and diffusive model development

Binary mixtures of different size ratios of urea and potash were filled in the shear box up to 85 mm (the height of the shear box is 100 mm, dimension 150 mm × 76 mm × 100 mm). This is consistent with the height of bagged blended fertilizers (Jha, 2008). Based on size of coarse and fine particles, the bed of binary mixture of 85 mm height was divided into twelve slabs, i.e., 11 interfaces and two faces (1 to

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**Fig. 1** Binary mixtures in shear box showing 12 equal size layers for model development.

**Fig. 2** Three-dimensional mass balances in the shear box.
Mass balance of fines along the three principal directions is given in Fig. 2. The main driving forces acting on binary mixtures in the shear box were gravity and shear (to-and-fro) motion of the box. Based on the nature of driving forces, the following two assumptions were made:

1) velocity of fines along gravity (z-direction) can be represented by a mean effective value,  
2) there is negligible concentration and velocity gradients along x and y directions compared with z-direction.

Based on the above, the governing convective-diffusive model is:

\[
\frac{\partial m_f}{\partial t} = -\bar{v}\frac{\partial m_f}{\partial z} + \bar{D}\frac{\partial^2 m_f}{\partial z^2}, \quad 0 < z < h = 85 \tag{1}
\]

Here, \(\bar{v}\) and \(\bar{D}\) are the fundamental material parameters and represent the convective or demixing and diffusive or mixing components, respectively. The units of \(\bar{v}\) and \(\bar{D}\) are mm/min (dimension, L/T) and \(mm^2/min\) (dimension, \(L^2/T\)), respectively. The binary mixture in the shear box was filled to height of 85 mm.

Based on nature of the problem, it was assumed that binary mixtures were well mixed when placed in shear box (Jha and Puri, 2009). The following initial condition and two boundary conditions are used to describe segregation attributed to the percolation mechanism, i.e., solve differential equation (1):

\[
m_{f0}(z, t = 0) = m_{f0}, \quad 0 \leq z \leq 85 \tag{2}
\]

There is a screen of opening size 2,360 \(\mu m\) at the bottom of the shear box, the preliminary experiments showed that there is no mass accumulation on the screen but some resistance was offered by screen bottom. The resistance “R” was included in the boundary condition at the screen bottom given in equation (3). Although top surface of binary mixture was slightly convex during the test (largest noted mound angle was \(\leq 5^\circ\)); therefore, for simplicity it was assumed that the top surface remains flat. With the help of above two conditions, two boundary conditions were formulated as given in equations (3) and (4). Crank and Nicolson (1947) proposed method that combines implicit and explicit method was used. Central difference method was for solving the differential equation.

\[
\left( R \frac{\partial m_f}{\partial z} \right)_{z=1} = \left( m_f \right)_{z=1} \tag{3}
\]

\[
\vartheta = 85, t > 0, \text{ Resistance offered by screen} \tag{3}
\]

\[
\frac{\partial m_f}{\partial z} = 0 \quad z = 0, t > 0, \text{No mass flux out of free surface} \tag{4}
\]

Crank and Nicolson method reduces the number of calculations and results in a difference form that is valid (i.e., convergent and stable) for all finite values. For most stable results, Crank-Nicolson scheme takes average of explicit and implicit methods. The final form of equation using Crank and Nicolson method is given in equation 5. Details of solving equation is given in Jha (2008):

\[
m_{fi,j+1}^{i+1} - m_{fi,j}^{i+1} = -\frac{\Delta t}{2\Delta z} \left( m_{fi-1,j}^{i+1} - m_{fi+1,j}^{i+1} + m_{fi-1,j}^{i} - m_{fi+1,j}^{i} \right)
\]

\[
+ \frac{\Delta t}{\Delta z^2} \left( m_{fi-1,j}^{i+1} + 2m_{fi,j}^{i+1} + m_{fi+1,j}^{i+1} \right) \tag{5}
\]

Consider

\[
\mu = \frac{\bar{v}\Delta t}{2\Delta z}, \text{ a dimensionless parameter}
\]

\[
\delta = \frac{\bar{D}\Delta t}{2\Delta z}, \text{ a dimensionless parameter}
\]

The physical interpretations of \(\mu\) and \(\delta\) are mass balance and concentration-gradient related parameters that represents the fraction of material leaving a given location. Solving for implicit and explicit parameters after incorporating \(\mu\) and \(\delta\). Rearranging equation (5) leads to equation 6.

\[
2(m_{fi,j}^{i+1} - m_{fi,j}) = -\mu \left( m_{fi-1,j}^{i+1} - m_{fi+1,j}^{i+1} + m_{fi,j}^{i} - m_{fi-1,j}^{i} \right)
\]

\[
+ \delta \left( m_{fi-1,j}^{i+1} - 2m_{fi,j}^{i+1} + m_{fi+1,j}^{i+1} \right) \tag{6}
\]

The final form of equation (6) is given in equation 7.

\[
-(\mu + \delta)m_{fi-1,j}^{i+1} + (2 + \mu + 2\delta)m_{fi,j}^{i+1} - \delta m_{fi,j}^{i+1} = (\mu + \delta)m_{fi,j}^{i} + (2 - \mu - 2\delta)m_{fi,j}^{i+1} + \delta m_{fi+1,j}^{i+1} \tag{7}
\]

Incorporating the two boundary conditions, a program was written in MATLAB (Mathworks Inc, Natick, Massachusetts) to solve the finite difference equations with the given boundary and initial conditions (Jha, 2008). The precision of \(\mu\) and \(\delta\) was kept to two decimal places to be consistent with variations in experimental data. Equation (8) represents the
Root mean square error (RMSE) = \[ \sqrt{\frac{\sum_{i=1}^{n} (\hat{m}_i - m_i)^2}{n - df}} \] (8)

where, \( n \) = number of observations

2.4 Dimensional analysis model development

Based on the physics of the problem and to correctly apply Fourier’s principle of dimensional homogeneity without omitting significant variables (Streeter et al., 1996, Murdock, 1993), percolation segregation in bagged fertilizer was assumed to be affected by size, shape, density, and mixing ratio, relative movement (strain), intensity of movement (strain rate), and fill height of bagged blended fertilizers. As mechanistic theory states that a mathematical relationship exists between percolation segregation in particulate materials (e.g., fertilizer) and physical and mechanical properties of the particulates. In the case of fertilizer, physical property includes size (as Size Guide Number, SGN, which is defined as size in millimeters multiplied by 100), size ratio (as Uniformity Index, UI, which is defined as 100 times the (size of 10 percentile particle divided by 95 percentile particle), shape, density, and mixing ratio and mechanical property includes strain (displacement), strain rate (intensity of to-and-fro motion), and bed depth. However, there are other physical parameters that indirectly affect segregation such as surface texture, surface composition, and electrostatics; their effect being secondary compared with the gravitational force, were not included. The physical and mechanical parameters used in this model are defined below for the better understanding.

Buckingham Pi theorem was to be used to develop dimensional analysis model but the number of variables were not sufficient to make proper dimensionless grouping. Based on fertilizer blend plant visits, experimental data, and previous experiences of Tang and Puri (2007), the physical and mechanical parameters that significantly affect percolation segregation are included in the mechanistic theory and grouped in such a fashion that each term is dimensionless.

Based on these considerations, a dimensional analysis model was proposed and is given below in equation (9).

\[
\frac{NSR}{StrainRate} = c (Shp)^{l} (\text{Coarse size})^{m} (\text{Mixing Ratio})^{o} (\text{Size Ratio})^{p} \] (9)

where,

- \( NSR \) = Segregated fines/fines in the mixture/Total time of FSSC-II operation, kg/kg-s
- \( Strain Rate \) = intensity of movement of bagged fertilizer, Hz
- \( Shp \) = shape and density of fertilizer, dimensionless
- \( Coarse Size \) = size guide number (SGN) or size of particle, mm*100
- \( Bed Depth \) = depth of fertilizer in the bag, mm
- \( Mixing Ratio \) = ratio of mass of coarse to mass of fines*100, dimensionless
- \( Displacement \) = relative displacement two side walls (length wise) of bagged fertilizer, %
- \( Size Ratio \) = defined via Uniformity Index, UI, which is defined as 100 times the (size of 10 percentile particle divided by 95 percentile particle), dimensionless
- \( l = \) power, indicates the contribution of shape and density to NSR/Strain rate
- \( m = \) power, indicates the contribution of coarse size and number of layers of coarse in the bed depth to NSR/Strain rate
- \( n = \) power, indicates the contribution of mixing ratio to NSR/Strain rate
- \( o = \) power, indicates the contribution of strain to NSR/Strain rate
- \( p = \) power, indicates the contribution of size ratio to NSR/Strain rate

The constant \( c \) and exponents \( l, m, n, o, \) and \( p \) were calculated by taking logarithm of equation (6) followed by linear regression analysis. Their physical meaning is needed to understand the dimensional analysis model well. Segregation under shear motion is contributed by the difference in physical and mechanical properties of particulates. On the left side, the model has two parameters, normalized segregation rate (NSR) and strain rate. The segregation measuring parameter NSR was developed to make segregation rate independent of amount of initial fines in the material mixtures. The NSR contains two fundamental dimensions, mass (M) and time (T). The second parameter strain rate is the operating parameter of the FSSC-II and it has the unit of time.
Strain Rate = \ln \left( \frac{\text{Size Ratio}}{\text{Mixing Ratio} + m \ln \text{Shape}} \right) + n \ln \left( \frac{\text{Coarse size}}{\text{Bed Depth}} \right) + \ln(m \text{Strain}) + \ln(p \text{Strain Rate}) \quad (10)

The physical and mechanical parameters were obtained from the experimental data and NSR was also calculated individually for these parameters. The size ratio rounded off to the first decimal place was used for data collection. The root-mean square error (RMSE) and the coefficient of variation (CoV) were calculated to evaluate the accuracy of the model through equations (11) and (12).

\[ \text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (P_i - R_i)^2} \quad (11) \]

\[ \text{CoV} = \frac{\text{RMSE}}{\text{Experimental mean}} \times 100\% \quad (12) \]

3. Results and Discussion

Results of the convective and diffusive segregation model and dimensional analysis model development and validation are presented and discussed in the following sections.

3.1 Convective and diffusive segregation model development

The measured and modeled segregated mass versus time relationships for potash and urea are given in Fig. 3. Fig. 3a shows the segregated mass vs. time relationship for size ratio 2.4:1.0 of potash at strain of 6% and strain rate of 0.5 Hz. The cumulative segregated mass increased with time. For size ratio 2.4:1.0, convective and diffusive segregation model values were within the 95% confidence interval (CI) of measured values. For size ratio 1.7:1.0, the modeled segregated mass values were always lower than the measured segregated mass values but within the 95% confidence interval (Fig. 3b). Fig. 3c shows the segregated mass vs. time relationship for size ratio 2.4:1.0 of potash at strain of 10% and strain rate of 0.5 Hz. For size ratio 2.4:1.0, convective and diffusive segregation model represents the measured values are also within the 95% confidence interval. For size ratio 1.7:1.0, the modeled segregated mass values were initially higher and after 25 minutes lower than the measured segregated mass values (Fig. 3d). The initial over prediction and later under prediction of segregated mass were observed because at higher strain of 10%, time was not enough for fine particles to percolate through the void spaces of coarse particles and also due to bridges formed in the binary mixtures. At later stage (after 25 minutes), bridges very likely collapsed and fine particles found the way through void spaces of coarse particles because
of less fines in binary mixtures. The initial modeled segregated mass up to 10 minutes of PSSC-II operation was not within the 95% confidence interval of the measured segregated mass. For size ratio 2.4:1.0 and 1.7:1.0 of potash, convective and diffusive segregation model segregated mass values were not within the 95% confidence interval at strain of 2% and strain rate of 0.5 Hz. At strain of 2% and strain rate of 0.5 Hz, the input energy was not sufficient to create large void spaces so that fines could percolate and bridges might have formed within the binary size mixtures of potash. Similar results were obtained at three strains of 2%, 6%, and 10% and strain rate of 0.25 Hz for size ratios 2.4:1.0 and 1.7:1.0; also for size ratios 2.0:1.0 and 1.4:1.0 when the coarse size was 3,075 µm. The modeled segregated mass was not within the 95% confidence interval for size ratios 1.7:1.0 and 1.4:1.0 when the coarse size was 2,580 µm.

At strain rate of 0.25 Hz and strain of 6%, convective and diffusive segregation modeled segregated urea mass was not within the 95% confidence interval; however, model under-predicted in the initial phase up to 13 minutes and thereafter over-predicted. At strain rate of 1.0 Hz, the modeled segregated mass values were not within the 95% confidence interval and under-predicted in the initial phase up to 22 minutes and thereafter over-predicted. In the case of urea over and under prediction of were observed in binary mixtures for the size ratio 2.0:1.0 at different times. The shape of urea was spherical and density was lower than that of potash. Gravity is the dominant force for material separation in binary mixtures, the combined effect of size, shape, and density was more in the case of potash compared with urea. The bridges very likely formed and energy supplied by the shear box and dominant gravity force was not sufficient to break those bridges initially and also time was not sufficient for fines to percolate through void spaces of coarse particles bed.

If energy imparted by shear box was sufficient to break bridges, then fines particles percolated through coarse particles bed. With increasing time, proportion of fines decreased that increased the rate of segregation causing model to under-predict segregated mass. Similar results were obtained for size ratio 1.7:1.0 at strain rates of 0.25 Hz and 1.0 Hz and for size ratios 2.0:1.0 and 1.7:1.0 at strains of 10% and 2% for strain rates of 0.25 Hz and 0.5 Hz. The convective and diffusive model well represented the measured segregated mass values of binary size ratios when the size ratios were higher than 1.4:1.0 and coarse size was larger than 2,580 µm. The accuracy of the model was the highest at strain of 6% because void space created in the coarse particles bed and time available to percolate for fine particles was sufficient. The under and over predictions of modeled segregated mass were observed in binary mixtures for the size ratio 2.4:1.0 at strain of 6% and 10% and strain rate of 0.25 Hz and 1.0 Hz.
seggregation mass might also have been observed because of other physical properties involved such as surface texture, and surface property. These properties cannot be studied with the discussed continuum theory based model. To overcome the limitation of continuum theory model, a hybrid model comprising continuum and discrete element theories (including multiscale formulations) needs to be developed, i.e., combines the advantages of continuum and discrete element models (including multiscale formulations) to explain segregation at particle-particle level and secondary and tertiary level structures.

4. Dimensional Analysis Model

The exponents l, m, n, o, and p were determined using linear regression. Equation (10) was linearly regressed using data for binary mixtures of urea and potash and binary mixtures when urea and potash were taken together. Equation (10) was also regressed using data of ternary mixtures of urea and potash. Quaternary mixtures were not used individually for linear regression because only limited (three) number of quaternary mixtures were available. Finally equation (16) was linearly regressed when binary, ternary, and quaternary mixtures of urea and potash were taken together. For multi-size mixtures, the equation was regressed in two different ways. In the first case, the average shape or porosity (53%) was considered when the porosity of urea and potash mixtures was 51% and 55%, respectively so that porosity could be treated as constant to eliminate a term from the final equation. In the second approach, the actual porosity of urea and potash was used for determining the coefficient l, m, n, o, and p.

4.1 Determination of exponents l, m, n, o, and p for multi-size mixtures

Regression-based variance of analysis for binary mixtures of potash showed that all five terms, Constant, ln(Size Ratio), ln(Coarse Size/Bed Depth), ln(Mixing Ratio), and ln(Strain) had significant effect on NSR/Strain Rate (p<0.05). The values of these five exponents l, m, n, o, and p (14.6, 7.17, -8.02, -1.28, and 1.63, respectively) were obtained through regression analysis and equation is summarized below (13).

\[
\ln\left(\frac{\text{NSR}}{\text{Strain Rate}}\right) = 14.6 + 7.17\ln(\text{Size Ratio}) - 8.02\ln\left(\frac{\text{Coarse size}}{\text{Bed depth}}\right) - 1.28\ln(\text{Mixing Ratio}) + 1.63\ln(\text{Strain})
\]

The porosity of binary mixture of potash was constant (55%) and was incorporated into constant “c” in the linear regression analysis. The contributions of Size Ratio and Strain were proportional to NSR/Strain Rate, whereas contributions of Coarse Size/Bed Depth and Mixing Ratio were inversely proportional. Positive constant l and exponents p and o represent that the effect of porosity, Size Ratio, and Strain are proportional to NSR/Strain Rate, i.e., increase in porosity, Size Ratio, and Strain will increase the NSR/Strain Rate. The negative exponents m and n represent Coarse Size/Bed Depth and Mixing Ratio showed inverse relation with NSR/Strain Rate, i.e., increase in Coarse Size/Bed Depth and Mixing Ratio will decrease NSR/Strain Rate.

Regression-based variance of analysis for ternary potash mixtures showed that all five terms, Constant, ln(Size Ratio), ln(Coarse Size/Bed Depth), ln(Mixing Ratio), and ln(Strain) had significant effect on NSR/Strain Rate (p<0.05). Values of these five parameters l, m, n, o, and p (22.6, 8.64, -9.12, -2.47, and 1.88, respectively) were obtained through regression analysis and are given in Table 5 with \(R^2 = 0.943\). The R-square value for ternary size mixtures (0.943) was higher than the binary size mixture (0.90) because coarse size 2,580 µm was included only twice compared with 5 times in the binary mixtures.

The porosity of ternary mixture of potash was constant (55%) and incorporated into constant “c” in the linear regression analysis. The contribution of Constant, Size Ratio, Coarse Size/Bed Depth, Mixing Ratio, and Strain was larger for ternary mixtures vs. binary mixtures when the regression equation was developed. Experimental design for binary and ternary mixtures showed that ternary mixtures had used coarse size 2,580 µm used twice vs. three times with binary mixtures (Tables 1 and 2). Exponents of Size Ratio and Coarse Size/Bed Depth were higher compared with Mixing Ratio and Strain. It means Size Ratio and Coarse Size/Bed Depth contributed more to NSR/Strain Rate compared with Mixing Ratio and Strain. Also the effect of Coarse Size/Bed Depth was the highest among five variables followed by Size Ratio. Results showed that size was the most dominant variable contributing towards segregation of fines from well mixed systems.

4.2 Validation of convective and diffusive segregation model

The convective and diffusive model was validated for size ratios 2.0:1.0 at strains of 2%, 6%, and 10% and strain rates of 0.25 Hz and 0.5 Hz (Table 3). The
Convective and diffusive parameters were estimated using linear interpolation from size ratios 2.4:1.0 and 1.7:1.0 for potash and strain rates of 0.25 and 1.0 Hz for urea at strains of 2%, 6%, 10%. The goal of interpolation was to determine convective, diffusive, and resistance parameters to predict segregated mass for potash and urea of size ratio 2:1.0. Fig. 4 shows the graphical representation of modeled data and experimental data. With the help of interpolated convective, diffusive, and resistance values, the segregated mass values were calculated but the modeled segregated mass values under predicted the measured segregated mass values at strains of 2%, 6%, and 10%. For size ratio of 2.0:1.0 of potash, at the strain of 6% and strain rate of 0.5 Hz after 30 minutes, the actual modeled segregated mass was 7.3 g, which is higher than the experimental segregated mass. Whereas, the segregated mass calculated from the model operating parameters convective, diffusive, and resistance obtained from linear interpolation was 45.3 g higher than experimental segregated mass. The modeled segregated mass was within the 95% confidence interval of the experimental values. The modeled convective, diffusive, and resistance values were 0.28 mm/min, 74.18 mm²/min, and 17 mm, respectively. Whereas these parameters calculated from linear interpolation were 0.85 mm/min, 45.11 mm²/min, and 6.75 mm, respectively.

The convective parameter was higher in the case of linear interpolation but diffusive and resistance parameters were higher in the modeled case. In the case of linear interpolation convective parameter responsible for segregation was overestimated but the other two parameters responsible for mixing (diffusive) and resistance responsible for offering resistance in binary mixture due to bridging were underestimated. The convective parameter is dominant because of dominant gravity driving force on larger fines size that resulted in over estimation of the segregated mass. The interpolation of convective, diffusive, and resistance parameters is not the best approach because the NSR is not linearly dependent on (ln(NSR) = m ln(size ratio)). Where m is the exponent of size ratio and varies with size ratio and type of material.

For size ratio of 2.0:1.0 of potash, at strain of 10% and strain rate of 0.5 Hz after 30, the modeled segregated mass was 8.4 g (643.2g) higher than the experimental segregated mass. Whereas, the segregated mass calculated from the model operating parameters convective, diffusive, and resistance obtained from linear interpolation was 24.5 grams (643.2g) higher than experimental segregated mass. The modeled segregated mass was within the 95% confidence interval of the experimental values. The modeled convective, diffusive, and resistance parameters were 0.99 mm/min, 98.24 mm²/min, and 13.00 mm, respectively. Whereas these parameters calculated from linear interpolation were 1.20 mm/min, 42.61 mm²/min, and 2.75 mm, respectively.

For size ratio of 2.0:1.0 of urea, at the strain of 6% and strain rate of 0.5 Hz after 30 minutes, the modeled segregated mass was 2.3 g (469.0g) lower than experimental segregated mass. Whereas, the segregated mass calculated from the model operating parameters convective, diffusive, and resistance obtained from linear interpolation was 15.4 g (469.0g) lower than experimental segregated mass. The modeled segregated mass was not within the 95% confidence interval of the experimental values. The modeled convective, diffusive, and resistance parameters were 0.00 mm/min, 21.05 mm²/min, and 1 mm, respectively. Whereas these parameters calculated from linear interpolation were 0.00 mm/min, 16.54 mm²/min, and 1 mm, respectively. Sufficient number of size ratios was not available so that a definite relationship between NSR and size ratio
could be deduced. The segregation behavior of urea at strain of 6% and strain rates of 0.25 and 1.0 Hz could not be used to explain the segregation behavior at intermediate strain rate 0.5 Hz accurately. To explain the segregation behavior of fines in urea at strain intermediate strain of 0.5 Hz and smaller size ratios of potash (<1.7:1.0), a hybrid model (as mentioned previously) which combines the principles of continuum and discrete element theories needs to be used.

5. Validation of the dimensional analysis segregation model

Accuracy of the model developed using binary mixtures of potash was determined through the comparison of experimental values to modeled values for binary mixtures of potash and urea. When equation (10) was used for validation purposes, the CoV, RMSE, and overall mean of the modeled values when compared with the experimental values were 18%, 0.63, and 2.85, respectively, for binary mixtures of urea and potash at strain of 2% and strain rate of 0.25 Hz. The CoV, RMSE, and overall mean of the modeled values when compared with the experimental values were 15%, 0.82, and 4.64, respectively, for binary mixtures of urea and potash at strain of 6% and strain rate of 0.25 Hz. Whereas, at strain of 10% and strain rate of 0.25 Hz, the CoV, RMSE, and overall mean of modeled values when compared with experimental values were 11%, 0.58, and 5.47, respectively. However, the overall mean of the NSR for binary mixtures increased with the increase in strain from 2% to 10% although CoV for binary mixtures of urea and potash decreased with the increase in strain from 2% to 10% for the same size ratios.

To validate the model, comparison between the modeled data and experimental values was performed for the size ratios for which model was developed but at intermediate strain rate. When equation (7) was used for validation purposes, the CoV, RMSE, and overall mean of the modeled values to the experimental values were 9.1%, 0.47, and 4.48, respectively (Fig. 5). To validate the model, comparison between the modeled data and experimental values was performed for size ratios for which model was developed but at intermediate strain rate of 0.5 Hz.

6. Conclusions

Percolation segregation in binary, ternary, and quaternary mixtures of urea and potash was modeled by convective and diffusive segregation model and dimensional analysis scale-up enabling model. Convective and diffusive model included binary size ratios of urea and potash for both model development and validation. Model over-predicted for small size ratios and under-predicted large size ratios but data were within 95% confidence interval. Dimensional analysis model included the range of variables size ratio, shape, density, strain rate, and strain to predict NSR.

![Fig. 5](image-url) Validation of dimensional analysis model by comparison of modeled values to experimental values for potash at strain rate of 0.5 Hz, with ± SD as error bars (P-potash, first four number after P is mean size of coarse particles followed by binary size ratio corresponding to coarse mean size).
Developed dimensional analysis model was validated for binary mixtures of urea and potash. Furthermore, all the binary, ternary, and quaternary mixtures were combined with and without keeping porosity constant but found not be the representative of experimental data. Based on results, the following conclusion can be drawn:

1) For size ratio of 2.0:1.0 of urea, at the strain of 6% and strain rate of 0.5Hz after 30 minutes, the modeled segregated mass was 2.3g higher than the experimental segregated mass. Whereas, the segregated mass calculated from the model operating parameters convective, diffusive, and resistance obtained from linear interpolation was 15.4g lower than experimental segregated mass. The modeled segregated mass was not within the 95% confidence interval of the experimental values.

2) At the strain of 10% and strain rate of 0.5 Hz after 30 minutes, for size ratio 2.0:1.0 of urea, the modeled convective, diffusive, and resistance parameters were 0.00mm/min, 21.05mm²/min, and 1.00mm, respectively. Whereas these parameters calculated from linear interpolation were 0.00mm/min, 16.54mm²/min, and 1.00 mm, respectively.

3) For size ratio of 2.0:1.0 of potash, at the strain of 6% and strain rate of 0.5Hz after 30 minutes, the modeled segregated mass was 7.3g higher than the experimental segregated mass. Whereas, the segregated mass calculated from the model operating parameters convective, diffusive, and resistance obtained from linear interpolation was 45.3g higher than experimental segregated mass. The modeled segregated mass was not within the 95% confidence interval of the experimental values.

4) At the strain of 10% and strain rate of 0.5Hz after 30 minutes, for size ratio 2.0:1.0 of potash, the modeled convective, diffusive, and resistance parameters were 0.99mm/min, 98.24mm²/min, and 13.00mm, respectively. Whereas these parameters calculated from linear interpolation were 1.20mm/min, 42.61mm²/min, and 2.75mm, respectively.

5) When equation (5) was used for validation purposes, the CoV of the modeled values to the experimental values were 18%, 15%, and 11% for binary mixtures of urea and potash at strains of 2%, 6%, and 10% and strain rate of 0.25Hz.

6) The dimensional analysis model developed by the binary mixtures of potash was sufficient to represent the binary, ternary, and quaternary mixtures of urea and potash.

This study showed the strengths and revealed limitations of continuum theory-based segregation model. To overcome the limitations, multiscale formulations that combine the principles of continuum and discrete element theories (involving multi-scale formulations) need to be developed to better model the segregation of fines; for not only smaller size ratio, but one that spans all size ratios.

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Author's short biography

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Virendra M. Puri, University Distinguished Professor, has been involved in research in the field of powder science, engineering, and technology for over three decades. He has served as the Acting Director of the NSF/IUCRC (Industry University Cooperative Research Center) – the Particulate Materials Center. Professor Puri has co-authored numerous publications. In addition, he is co-inventor and holder of patents in the area of powder flow, deposition and compaction. Professor Puri has a Copyright for multi-purpose computational software dealing with powder processing applications. He has been invited to serve on Editorial Boards, International Advisory Boards, and Chairpersons of several bulk solids-related publications and professional activities. Professor Puri is Co-Editor-in-Chief of Particulate Science and Technology, An International Journal. He regularly offers seminars, courses, and workshops in powder mechanics to industry and academia. Professor Puri has received several teaching and research awards.

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