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DEM Simulation of Binary Blend Mixing of Cohesive Particles in a High Intensity Vibration System

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Abstract

The effects of processing intensity, time and particle surface energy on mixing of binary cohesive blends (size ratio 1:2, fine concentration at 10 %) in high intensity vibration system were investigated via DEM simulations. Results show that both increasing processing intensity from 50 to 100 Gs and reducing surface energy from 50 to 0.5 J/m² lead to a faster mixing rate. Mixing Bond number (Bo_m) was introduced to capture the *effective* mixing rate, R_m ; higher Bo_m corresponding to lower mixing rate. The coefficient of variation, C_v , formed the basis for the mixing quality and R_m , while the mixing action is quantified by the product of R_m and mixing time ($P_{r,t}$). Simulation results show that C_v values drop initially, and then rise with $P_{r,t}$. Hence, low $P_{r,t}$ indicates inadequate mixing intensity, while high $P_{r,t}$ most likely indicates mixture segregation, and therefore too high or too low $P_{r,t}$ values should be avoided.

Key words: Cohesive particle mixing, DEM simulation, Bond number, Mixing mechanism

1. Introduction

The mixing of powders is a common but important step in chemical, mining, pharmaceutical, food and other industries ¹⁻⁶. The aim of mixing is to produce a mixture with adequate homogeneity, despite variations in powder properties such as particle size, particle size distribution, density, flowability, surface characteristics ⁷⁻¹⁰ and processing conditions such as the equipment and operating conditions, as well as the upstream and downstream processing steps ^{4,9-12}. For example, in a continuous direct compression tableting process, blend and drug content uniformity after feeding and mixing are critical factors for meeting tablet product uniformity requirements ¹³⁻¹⁵. In addition to particle size disparities between the constituents, the flowability of the powders are expected to have a critical impact on degree of mixing or segregation ^{4,15-17}. Existing literature provides the phenomenological explanations as well as some model-based understanding of particle mixing behavior ^{1,3,18-20}. Factors such as the particle density, particle sizes, size ratio, surface roughness, surface energy and shear rate, have been considered in analyzing the interactions between particles ^{6,21-24}. In general, the current understanding suggests that as compared with non-cohesive powders, cohesive blends are less likely to segregate although they may pose some challenge in achieving the mixing homogeneity, possibly requiring devices with higher intensity or longer processing times. Unfortunately, there is limited understanding of device effect and/or the mixing intensity effect.

There are a variety of mixers available, and several different ones have been used for dry mixing of powders ^{4,25-30}. Most industrial mixing devices exert relatively low intensity in comparison to those used in creating interactive mixtures ^{4,15,24}. For example, in pharmaceutical industry, low intensity blenders such as a tumbling blender, V-blender, Turbula mixer, ribbon blender, etc., are used. For some of these, their effective mixing intensity may be considered high

intensity at larger industrial scale. However, for the purpose of achieving higher intensity at lab-scale, high intensity mixing devices may be required. For example, vibrational mixers may be used as they have attracted attention due to their fast-mixing rate and good mixing quality^{31,32}. In addition to processing conditions, the degree of mixing attained in a mixer is also likely to be significantly affected by particle properties such as size, shape, and cohesion, because the rate and degree of mixing of free-flowing spherical particles is much higher than that of cohesive irregular particles^{2,4,5,28}. Unfortunately, the selection of mixing devices and processing parameters based on particle cohesion remains an underexplored topic. Therefore, further research is required to develop better understanding of mixing mechanisms for the cohesive fine powders. Establishing the relationships between mixing quality and particle properties together with processing conditions may benefit academics and industry practitioners in predicting mixing performance and selecting processing conditions for their specific blend systems.

Since the details experimental analysis of particle dynamics is rather challenging, Discrete Element Method (DEM) modeling and simulations have been widely used because they allow for computation of translational and rotational motion of individual particles in the system for obtaining detailed diagnostics of a process^{6,11,33-35}. Likewise, DEM simulations have been used to uncover macroscopic behavior of particulate matter^{12,28,36,37}. DEM has also been used effectively to study the mixing process of free-flowing particles³⁸⁻⁴⁰. However, the investigation of the mixing mechanisms for cohesive particles through DEM simulations have been less explored; only few examples of DEM simulations can be found dealing with cohesive particles^{41,42}. In those reports, the selected range of cohesive forces was relatively narrow, making it difficult to gain general understanding of the effect of particle cohesion effect on the mixing process³⁵. Nonetheless, DEM simulations are attractive as compared to traditional physical

experiments, with which it is rather challenging to develop general understanding due to the different mixing mechanisms and energy inputs of various mixing devices. Therefore, DEM simulations are utilized in this work as an alternative to compare and understand the cohesion effect on the mixing process, which allows for adjustment different processing conditions.

In this paper, the cohesive powder mixing process in a high intensity vibrational mixing system was investigated via DEM simulation keeping pharmaceutical blending as the application of relevance. The objective is to develop better mechanistic understanding of the mixing process by analyzing the relative effects of cohesion originating from particle surface energy and shearing forces due to mixing intensity. Mixing intensity was varied by applying different vibrational conditions, and the particle cohesion was varied via surface energy of the particles. The mixing process was analyzed by computing collision shear force, number and nature of the collisions, as well as resulting powder bed porosity. The Mixing Bond number, (Bo_m), defined as the ratio of pull-off force representing the particle cohesion, and shear force resulting from the mixing intensity, was introduced to characterize the mixing behavior. Bo_m was used to approximate the *effective* mixing rate, moving forward just termed the mixing rate, with higher value corresponding to lower mixing rate. The coefficient of variation of the mixture, C_v , frequently used for the characterization of pharmaceutical blends, was used to represent the mixing quality whereas the mixing action was captured through the product of *effective* mixing rate (R_m) and mixing time ($P_{r,t}$). Simulations were performed to examine the effect of $P_{r,t}$ on the mixing quality, C_v , for the purpose of gaining improved understanding of the effect of mixing intensity and processing time on the cohesive particle mixture quality and mixing dynamics.

2. Simulated System and Blend Content Uniformity

2.1 DEM Simulation Approach

A commercial discrete element method (DEM) simulation package named EDEM (EDEM 2018, DEM Solutions) was used to investigate cohesive particle mixing process in a high intensity vibration system. Translational and rotational motion of individual particles are given by the following equations.

$$m_i \frac{d\vec{v}_i}{dt} = \vec{F}_i + m_i \vec{g} \quad (1)$$

$$I_i \frac{d\vec{\omega}_i}{dt} = \vec{T}_i \quad (2)$$

In Eq. (1), m_i , \vec{v}_i , t , \vec{g} , and \vec{F}_i are the mass of particle i , velocity vector of particle i , time, gravitational acceleration, and contact forces for the particle-particle and particle-geometry interactions, respectively. In Eq. (2), I_i , $\vec{\omega}_i$ and \vec{T}_i are the moment of inertia, angular velocity and torque acting on particle i , which are induced by tangential contact force and rolling friction, respectively. *Hertz-Mindlin* with *Johnson-Kendall-Roberts* (JKR) cohesion contact model was used to describe the interparticle interaction between particles and interaction between particle-vessel geometry^{43,44}.

In order to simulate the cohesive particle mixing process, two sets of mono-sized particles were generated in a cuboid container with length, width and height of 1, 1, and 3 cm, respectively. As shown in Figure 1, 3000 large particles (gray) with 500 μm diameter were randomly generated and settled down at the bottom of container first, and then 2664 small particles (yellow) with 250 μm diameter were randomly generated and allowed to settle down on the top of coarse particles. The ratio of particle sizes and numbers lead to 10% by weight of small particles. After coarse and fine particles were introduced into the container, the container

vibrated at a pre-set amplitude from 5 to 10 mm with a frequency of 60 Hz along the z direction, thus having effective accelerations of 50 times gravity (Gs) and 100 Gs⁴⁵, respectively.

When cohesive force is much higher than a particle's weight, which results from small particle sizes or high *van der Waals*, electrostatic, or capillary forces, the mixing performance will be significantly affected⁴. Such an effect may be captured through a dimensionless parameter called the granular Bond number, defined as the ratio of cohesive force to gravity, and widely used to quantify inter-particle cohesion^{21,36,46}. It is given by the following expression,

$$Bo_g = \frac{F_{cohesion}}{mg} \quad (3)$$

here, $F_{cohesion}$ is the inter-particle cohesive force acting on a particle, m is the mass of the particle and g is the acceleration of gravity. In the JKR model, the normal cohesive force depends on the overlap (δ) and the interaction parameter, surface energy (γ), given by the following equations,

$$F_{jkr} = \frac{4E^i}{3R_e} a^3 - 4\sqrt{\pi\gamma E^i} a^{\frac{3}{2}} \quad (4)$$

$$a^4 - 2R_e\delta a^2 - \frac{2\pi\gamma}{E^i} R_e^2 a + R_e^2 \delta^2 = 0 \quad (5)$$

where $\frac{1}{R_e} = \frac{1}{r_i} + \frac{1}{r_j}$ and $E^i = \left(\frac{1-\nu_i^2}{E_i} + \frac{1-\nu_j^2}{E_j} \right)^{-1}$ are equivalent radius and combined elastic modulus, respectively, and ν_i, ν_j, E_i, E_j represent the *Poisson's* ratios, and *Young's* moduli of particles i and j . a is the contact radius. The maximum value of cohesion force occurs when particles break physical contact between each other. The value of maximum cohesion force, called pull-off force, is given by

$$F_{pullout} = \frac{-3}{2} \pi \gamma R_e \quad (6)$$

Here, the pull-off force is considered as the representative cohesive force. Although this simulation employs larger particles for the sake of keeping the computational burden limited, it was intended to mimic the experimental system comprising of coarse acetaminophen and Avicel 101 with sizes of 20 and 45 μm and surface energies of 40.86 and 42.33 mJ/m^2 , respectively ³². Other than particle size, the material properties of all particles were held constant for each simulation, and are listed Table 1. Use of larger simulated sizes of the particles are expected to overcome the computational burden, while physics of the real particulate system are retained by keeping the Bond numbers of simulated particles similar to the real particulate system ⁴⁷. This strategy was followed to minimize the effect of scaling of cohesion due to larger simulated sizes. It is noted that as compared with previous work, ^{12,48} simulated particles sizes were smaller and surface energy was adjusted to impart the required level of cohesion. Specifically, two standard model particles with the particle size of 250 and 500 μm and surface energy values of 0.5 through 50 J/m^2 were used. The coarse and fine particle size ratio of 2 was fixed so that the size driven effect on mixing was neither too small nor too high ⁶.

2.2 Quantification of Content Uniformity

Powder blend homogeneity is a key factor for evaluating the mixing quality after processing. As shown in Figure 2, the mixture was initially allowed to settle down at the bottom of container and this region was divided into 10 zones along the vertical axis for the sake of mixing quality quantification. The concentrations of small particles (yellow), which was counted by small particle number, in each zone were calculated and the coefficient of variation of the dose (C_v),

defined as the ratio of the standard deviation of concentration of small particles in each zone to the average concentration of small particles in the mixture, was computed as below.

$$C_v = \frac{\text{standard deviation of concentration of small particles} \in \text{each zone}}{\text{average concentration of small particles} \in \text{the mixture}} \quad (7)$$

Johnson model, which is an excellent model for predicting theoretical C_v values of ideal mixing of two components based on dosage and particle size distribution, is considered ⁴⁹. The C_v value for the *Johnson* model is given by the following expression,

$$C_v = 100 y \left(\frac{500 \pi \rho}{3 G} \right)^{0.5} \left[\sum f_i \left(\frac{d_i}{10000} \right)^3 \right]^{0.5} \quad (8)$$

Here, y is the fraction of major component in the mixture, which is 0.9 in this study, ρ is density of the drug (g/mL), G is the particle dose per sample (mg), which is 36 mg in this study, d is the mean particle size (μm), f_i is the weight fraction of mean particle size d_i (μm). If the actual C_v value is close to or lower than the theoretical C_v value based on the *Johnson* model, the mixture is considered well mixed. Otherwise, the mixing quality is not good. It should be noted that the theoretical well-mixing C_v value is relatively high in this study which is due to the small dosage of sample and large particle size.

3. Numerical Results and Discussions

The effect of vibration intensity was investigated by varying displacement magnitude from 5 to 10 mm, while keeping the frequency (60 Hz) fixed. This allowed the mixing intensity to vary from 50 Gs to 100 Gs. Hence, in this study, amplitude was used to indicate mixing intensity. The higher displacement magnitude equates to higher processing intensity or effective acceleration. Interparticle cohesive force is highly dependent on the particle size, *van der Waals* force,

moisture, electrostatic, or capillary force, etc. Here, the cohesive force is assumed to be the pull-off force based on the JKR model, which is related to the surface energy. Thus, by changing the value of the surface energy, the effect of cohesive forces on mixing quality could be investigated.

3.1 Vibration Intensity Effect on Mixing Behavior

Mixing experiments of two sets of mono-sized particles (250 and 500 μm) were conducted using the same surface energy (5 J/m²), with processing amplitudes varied from 5 to 10 mm. Figure 3 depicts the mixing performances after processing 1s, 5s and 10s at different processing intensities. It is observed that, in all cases, the mixing degree increased with the increase of mixing time, which means longer the processing time, better the mixing quality. In addition, the mixing rate, when processed at high processing intensity, was much faster than that at low intensity. The *effective* mixing rate (R_m) was investigated to understand particle mixing behavior, and the equation to calculate *effective* mixing rate is listed below,

$$R_m = \frac{\Delta C_v}{t_h} \quad (9)$$

Where ΔC_v is change in the C_v value compared to $t = 0\text{s}$, and t_h is the time to reach homogeneity or the endpoint of the process (10 s in this study). After 1s, it is clear to see from Figure 3(a) that small particles (yellow) transferred from top to bottom. On the other hand, Figure 3(c) shows that most of small particles were still on the top of big ones, even after processing for 10s with 5 mm amplitude. While very qualitative, visual observation indicates that at higher amplitudes of 10 mm and 7.5 mm, a relatively high homogeneity was reached in 5s and 10s, respectively, as compared to the 5 mm case. This is quantified in Figure 4, which depicts the C_v values of small particles as a function of time at different processing intensities. In all cases, C_v values reduced with the increase of mixing time, which was expected. The actual C_v value reached the

theoretical well-mixing C_v value (4.7%) in 6 seconds, and maintained steady state for the next 4 seconds when using a processing amplitude of 10 mm, indicating particles were well mixed and no segregation or de-mixing phenomenon was observed. C_v value reduced from 156% to 18.2% when processed at 7.5 mm amplitude for 10s. However, for 5 mm amplitude case, the C_v value only declined from 154% to 115%, and at a much slower rate as compared to the other two cases. For the 5 and 7.5 mm amplitude mixing cases, although C_v values declined with the increase of processing time, 10s was still not long enough to properly mix the particles at such intensities. Hence, longer processing time is suggested. In conclusion, high processing intensity could significantly elevate the mixing speed for cohesive powders. Similar phenomenon has also been reported in another work ⁵⁰.

3.2 Effect of Vibration Intensity on Collision Shear Force, Collision Rate and Particle Bed Porosity

In order to better understand why high intensity led to better mixing rate and quality, the mixing mechanism was further investigated. For high intensity vibration-based mixing, particles randomly collide with each other. This type of random collision-driven rearrangements are reported as diffusion-like mixing, where the diffusion rate or diffusion coefficient depends on the shear rate and particle size ^{12,51}. Since the same amount and particles having identical properties were used in various cases, the ‘viscosities’ of mixtures are assumed to be the same. Mixing simulations for two sets of mono-sized particles (250 and 500 μm) were performed using the same surface energy (5 J/m^2) with processing amplitudes varied from 5 mm to 10 mm, and fixed processing time of 10 seconds. Collision shear force was obtained from EDEM, which depends on tangential overlap and tangential stiffness, and is derived from the *Mindlin-Deresiewicz* theory ⁵². Average collision number was evaluated for further assessing the mixing process.

Three types of particle collisions were considered: small-small, small-big and big-big. The particle collision force was calculated based on the collision force computed through EDEM via averaging. Since too many collisions occurred simultaneously while the collision shear force changes at different timesteps, averaging of particle collision shear force was required. Figure 5 shows the average collision shear force for collisions between small-small, small-big and big-big particles as a function of mixing intensity. As shown, the particle collision shear force for all three types of collisions shows increasing trends as mixing intensity increased from 5 to 10 mm. This may be due to high intensity mixing enhancing relative velocities between the particles ⁴⁵. Thus, the dynamical assessment of the particle bed porosity is important, and more details will be discussed in a later section. In addition, the average collision force between big particles was far larger than the small-small and small-big collision, as expected. Which means the system was dominated by the big-big particle collisions. An interesting finding is that the increase of collision shear force from 7.5 to 10 mm was much larger than that from 5 to 7.5 mm, which means the effect was non-linear. This is because the collision shear force is related to the collision normal force, which is directly proportional to the square of the relative velocity between the particles in high intensity vibration system ⁴⁵.

Average collision rate is another important parameter for evaluating the mixing performance, which is defined as

$$Collision_{rate} = \frac{\sum C_{n,i}}{t_c * N} \quad (10)$$

Where C_n is collision number for each particle, t_c is collision time and N is number of particles, Figure 6 presents the effect of mixing intensity on collision rate for collisions between small-small, small-big and big-big particles. As shown, collision rate rose with increasing processing

intensity for all three cases, thus indicating higher intensity would lead to faster mixing rate. An interesting finding is observed that small-small particle collision rate was much smaller than that of small-big and big-big collisions, indicating collisions involving big particles were dominant in high-intensity vibration systems.

In the high intensity mixing system simulated here, the bed of particles has a tendency to move together as a whole, hence the phenomena could be treated as a moving powder bed. Since porosity plays a significant role in the mixing process⁵³, processing intensity effect on powder bed porosity was evaluated. Here, powder bed porosity is defined as the ratio of total pore volume to apparent volume of the moving powder bed, and not the entire available vessel space. Figure 7 shows powder bed porosity as a function of intensity. The powder bed porosity rose from 0.40 to 0.48 with increasing intensity, from 5 to 10 mm. Large porosity could provide more voids for small particle diffusion. In addition, it could enhance the mean free path of particles which leads to higher relative velocity and collision force that could overcome pull-off force between particles as well. Both of these effects could help enhance the mixing rate. In conclusion, higher processing intensity could enhance particle mixing rate by enhancing particle collision shear force, collision numbers and powder bed porosities.

3.3 Effect of Surface Energy on Mixing Behavior

The effect of cohesion on particle mixing was evaluated by investigating the maximum cohesion force or pull-off force. Since pull-off force is related to surface energy, the effect of surface energy on particle mixing quality was investigated. Mixing experiments of two sets of mono-sized particles (250 and 500 μm) were conducted using the same processing intensity (10 mm) with surface energy varied by two orders of magnitude from 0.5 to 50 J/m^2 for 10 seconds. The

surface energy of 50 J/m² was regarded as very cohesive case and 0.5 J/m² was regarded as nearly non-cohesive case.

Figure 8 depicts the C_v values of small particles, changing as a function of time at different surface energy values, all processed at processing amplitude of 10 mm. It is observed that the C_v value decreased faster with decreasing surface energy. For the 0.5 J/m² case, the C_v value reduced to the theoretical well-mixing C_v value (4.7%) in 0.3 seconds. Unfortunately, it then elevated to 20% after 0.5 seconds, which was likely due to de-mixing or segregation of particles upon further shear. On the other hand, C_v value only reduced from 157% to 115% after 10 seconds when surface energy was very high at 50 J/m². Therefore, either too high or too low values of surface energy or cohesion force are not good for achieving very good mixing, which was also observed in previous research^{4,11,12}. High cohesion force reduces mixing rate while low cohesion force leads to particle segregation.

Figure 9 presents the concentration of small particles as a function of vertical axis at 10s under a processing amplitude of 10 mm. In this figure, the normalized distance (Z-axis) of 0 is the bottom of the container, and 100 is the top of powder bed. It is easily seen for the intermediate value of surface energy at 5 J/m², small particles were well mixed after 10s processing. However, the concentration of small particles reduced with increasing powder bed height at the low surface energy value of 0.5 J/m², which means more small particles transferred from the top to bottom and segregation of particles was observed. However, for the high surface energy of 50 J/m², the concentration of small particles rose with increasing powder bed height, which means most of the small particles remained on the top of the powder bed.

To further analyze the mixing mechanism, collision shear force, collision rate and powder bed porosity was computed as a function of surface energy. Since the mixing system is

dominated by big-big particle collisions, and the trend of collision shear force and collision rate are similar for the various particle collision types, it is sufficient to only examine the big-big particle collisions. The results are shown in Figure 10 for collision shear force, collision rate and porosity as a function of surface energy at the highest processing amplitude of 10 mm and processing time of 10s. It is observed that the big-big particle collision shear force rose from 2.4 to 15.7 mN with increase of surface energy from 0.5 to 50 J/m², which in principle could enhance the mixing rate and improve the homogeneity of the mixture. However, both the collision rate and particle bed porosity reduced from 253 to 19 1/s and 0.52 to 0.43, respectively, with increase of surface energy from 0.5 to 50 J/m², which was due to high surface energy or cohesion between particles. Therefore, even when the large collision shear force helps improve the mixing rate, it was not high enough to overcome the effect of low collision rate and low powder bed porosity. Such results were evident from Figures 9 and 10. In conclusion, very high cohesion force between particles is not good for powder mixing, this could explain why dry powder coating could enhance the mixing quality by reducing the cohesive force between fine particles ⁴.

3.4 Ratio of Cohesive Force to Collision Shear Force

As mentioned in pervious sections, mixing rate and mixing quality are significantly affected by particle collision shear force, number of collisions and particle bed porosity. However, results so far do not lend themselves to a simple way to predict mixing rate and mixing quality because of the multiple factors that are involved, including process intensity and time. To facilitate predictive estimation of the mixing behavior in the high intensity vibration system, a dimensionless mixing Bond number (Bo_m) is introduced here, which is defined as the ratio of the cohesive force (pull-off force) to the collision shear force.

$$Bo_g = \frac{F_{Pulloff}}{F_{Collision\ shear}} \quad (11)$$

If the average shear force is higher than the pull-off forces between particles, it leads to a faster mixing rate. Since the aim of this study is to investigate the mixing behavior of small and big particles, the pull-off force used here is pull-off between small-big particles. In addition, as mentioned in previous section, big-big particle collisions are the most important collision type in the system, the average big-big particle collision shear force is used in the equation. Mixing experiments of two sets of mono-sized particles (250 and 500 μm) were conducted using the processing intensity (varied from 5 to 10 mm) and surface energy (varied from 0.5 to 50 J/m^2) for 10 seconds.

Figure 11 presents effective mixing rate as a function of mixing Bond number. It is found that mixing rate declined with increasing mixing Bond number, indicating higher cohesive force to collision shear force ratio is not good for mixing. The highest mixing rate 499 %/s was observed when the particle mixture with surface energy of 0.5 J/m^2 was mixed at 10 mm amplitude. On the other hand, the lowest mixing rate was observed when the particle mixture with surface energy of 50 J/m^2 was mixed at 5 mm amplitude. This mixture was the lowest mixing Bond number as well, indicating that collision shear force is not large enough to overcome the cohesive force between particles. Both simulations validated the observation that low cohesion force and high processing intensity enhance the mixing rate. However, as high mixing rate does not always lead to good mixing quality, the mixing time is another parameter in the particle mixing process. Figure 12 describes the C_v values of small particles as a function of product of *effective* mixing rate and mixing time ($P_{r,t}$). It is found that C_v values first reduced and then rose with increase of product of *effective* mixing rate and mixing time ($P_{r,t}$). The minimum

C_v value was observed when $P_{r,t}$ is around 100% to 300%. Before mixing, the C_v value is around 150%, indicating that $P_{r,t}$ could be used to estimate mixing quality. As shown in Figure 12, if $P_{r,t}$ was too small, which means mixing was not sufficient and mixing quality is not as good as expected. On the other hand, if $P_{r,t}$ was too big, although mixing rate was very fast and time was long, segregation of particles would be observed during the mixing process. In conclusion, the product of *effective* mixing rate and time ($P_{r,t}$) is an effective parameter to characterize the mixing performance in the high vibration system. Both too high or too low values of $P_{r,t}$ lead to inadequate mixing. In conclusion, if the powder is very cohesive, high processing intensity and long processing time are suggested. Meanwhile, if the powder is free flowing, low processing intensity or short processing time is preferred to get good mixing quality.

4. Conclusion

The cohesive particle mixing process for binary blends in a high intensity vibration system was investigated via discrete element method (DEM) simulations. Analysis of the collision shear force, collision rate and powder bed porosity as a function of particle sizes, vibrational amplitude representing the mixing intensity, particle cohesion represented by surface energy as well as processing time helped reveal the mixing dynamics and mechanism. When processed at the high amplitude of 10 mm, the coefficient of variation, C_v , reached the theoretical well-mixed C_v value (4.7%) in 6s, indicating particles were well mixed, and maintained the steady state in the remainder of the simulation time. In contrast, at lower intensities of 5 and 7.5 mm amplitude, the C_v values reduced much slower within the simulation time of 10s, representing lower mixing rate. Such trends were identified based on the detailed analysis of the collision shear force, collision rate and powder bed porosity. It is found that the mixing process is dominated by big-big particle collisions. High processing intensity enhances collision shear forces and collision

rates, which increase powder bed porosity and result in a higher powder mixing rate. Thus, high processing intensity is recommended to enhance the mixing efficiency. In addition, the effect of cohesion on particle mixing was evaluated by varying surface energy. C_v value reduced to theoretical well-mixed C_v value (4.7%) in 0.3 second and then rose to 20% after 0.5 second with low surface energy of 0.5 J/m². On the other hand, C_v value only reduced from 157% to 115% at 10 seconds with a very high surface energy of 50 J/m². Compared to the C_v value with a surface energy of 5 J/m², it is found that too high or too low cohesion force or surface energy are both not good for mixing. Such complex mixing dynamics was captured through introducing a dimensionless parameter, termed the mixing Bond number (Bo_m), to predict the *effective* mixing rate, R_m . The *effective* mixing rate declined with increasing Bo_m . Finally, the product of the *effective* mixing rate and mixing time ($P_{r,t}$) was used to characterize the mixing behavior. It was found that ideal mixing occurs when $P_{r,t}$ was close to its original C_v value. However, both too high or too low $P_{r,t}$ values are not conducive to proper mixing. If $P_{r,t}$ is too low, mixing is insufficient. If $P_{r,t}$ is too high, although the mixing rate is fast and mixing time is long, segregation of particles could be observed during mixing. Overall, if the powder is very cohesive, high processing intensity and long processing time would be required to reach mixture homogeneity. Meanwhile, if the powder is free flowing, a lower processing intensity and shorter processing time would be preferred in order to get adequate mixing quality.

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