Mixing of Bidisperse Cohesive Granular Materials in Food Processes

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***Abstract*—Particulate mixing is one of the most common unit operations in food processes. Yet, mixing phenomena are still not fully understood. In this study, we investigate experimentally the ability of cohesion to enhance mixing in dry cohesive particulate systems. Extensive chemical silanization is used to produce cohesive dry glass particles and the cohesive force is measured using an in-house setup. The effect of the cohesive force on the flow and mixing is then explored using a rotating drum. We found that high cohesive forces clump small particles together, and hence reduce the segregation and improve mixing. These results have important implications for food industrial processes (e.g., flowability control, engineered mixing and blending of multicomponent particulate systems).**

***Index Terms*—food processes, granular materials, mixing, cohesion, rotating drum, segregation**

I. Introduction

In food processes, mixing is employed not only to combine multiple components, but also to modify the structure and rheology of food products [1], [2]. However, the wide variety of particulates with different properties can result in major segregation issues, reducing the homogeneity of the food product. Hence, there is a need to understand and control the mixing mechanism of particulate systems.

To improve mixing and prevent the segregation in particulate food procesees, a typical approach is by using wet particles that will produce agglomerates clumbed by cohesive capillary forces. Chou *et al*. [3] demonstrated that segregation index in wet granular materials decreases with an increase of the angle of the slope in a rotating drum, regardless of the volume or viscosity of the added liquid. Jarray *et al*. [4], [5] investigated the effect of liquid induced cohesion on the flow in a rotating drum and showed that capillary cohesion increases the angle of the slope and decreases the granular temperature in the free owing surface. Li and McCarthy [6] showed that segregation in granular systems can be controlled by adding moisture. Apart from studies on segregation in wet systems with cohesion due to capillary forces, there are few experimental investigations focusing on dry cohesive systems. This is mainly because the cohesion between particles is difficult to introduce and control in dry systems. One way to modify the glass surface and obtain dry cohesive particles is called chimical silanization [7].

In this work, we experimentally investigate the effect of tunable cohesive forces on the mixing of dry granular systems. We use extensive silanization to modify the contact cohesion of millimetric size glass particles in a controlled way. The cohesive strength of the particles depends on the reaction duration time of the silanization process. Particle-particle cohesion force is measured using an in-house device. We employ a rotating drum as a mixing apparatus, where the flow and mixing of granular materials can be characterized conveniently because of its simple geometry comparing to other mixing devices. Finally, the influence of the cohesive forces between the particles on the bulk flow and mixing is explored and discussed.

II. Materials and Methods

A. Glass Surface Pre-treatments

Chemical compounds used for extensive silanization are: silanization solution~5 % (V/V, 5% in volume of Dimethyldichlorosilane in Heptane, Selectophore), Hydrochloric acid (HCl, 0.1 mol), Acetone and Ethanol. The procedure for making the glass particles cohesive is as follows: First, glass particles, initially hydrophilic, are cleaned for at least one hour by immersion into freshly prepared HCl solution under agitation using a rotor-stator homogenizer. Then, they are rinsed thoroughly with deionized water and oven dried for 3 hours at 60 ℃. Afterwards, the freshly cleaned samples of glass particles are immersed in the silanization solution under low agitation speed at room temperature for different duration of time (1, 2 and 3 hours) to obtain samples of particles with different cohesive forces. Finally, the glass particles samples are allowed to air-dry under a fume hood for 24 hours for covalent linkages to form with the substrate. The cohesive force between the silanized particles depends on the chemical silanization reaction duration and the Silane concentration.

B. Setup for the Cohesive Force Measurement

The cohesive force between the particles was measured using an in-house setup that we specifically designed for this study. The setup consists of a micro-balance

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(Sartorius) and a micro-positioner as shown in Fig. 1. The micro-positioner is mounted above the micro-balance. The whole setup is installed on an optical table to reduce the mechanical vibration. One particle was fixed on the balance by a double-sided tape while the other particle was glued to a flexible thin rod connected to the micropositioner. After the first mechanical contact between the particles, the balance shows a positive weight of about several mg. The weight value obtained in the microbalance corresponds to the cohesive force between the particles.

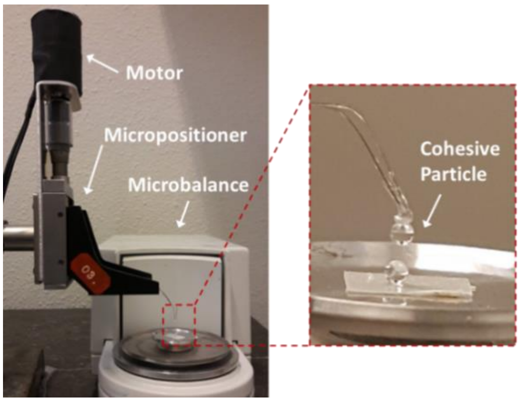

Figure 1. Setup used for measuring the cohesive force.

C. Rotating Drum Apparatus

To investigate the mixing of particles, a rotating drum is used. The drum is made by a cylinder of 60.5 mm inner radius and held between two circular Plexiglas (PMMA) plates of 5 mm thickness to allow optical access. For a quasi-two dimensional rotating drum, Jain et al. [1] argued that the width of the drum in the *z* direction should be larger than 6.4 *r*, with *r* the average radius of the particles, to neglect the front and back wall friction on the flow characteristics. The drum width in our study is 22 mm, larger than 6.4 *r* of all the particles used. The drum was placed on a horizontal rotating axis driven by a variable-speed motor, aligned along the *z* axis. Images of the rotating drum were recorded using a MotionBLITZ EoSens high speed camera working at 120 fps. Experiments were conducted using a selected set of Borosilicate glass particles of density 2500 kg/m3. The drum is less than half filled with the same mass of particles (i.e. 125 g of particles or a bed volume $V = 8.85\times10^{-5}$ m3).

D. Image Post-processing and Mixing Index Computation

The dynamic angle of repose and the mixing index were computed by analyzing the images acquired from the high speed camera. Particles were detected using the particle tracking package Trackmate within the FIJI ImageJ distribution [8]. Detected particles were converted into a sequence of tables and visualized by importation in ParaView [9]. The dynamic angle of repose is computed by linear regression of the positions of the particles using an in-house python code.

From the image software, binary images with different particle species were obtained. These binary images were converted into continuum density fields using a method similar Coarse Grain method (CG) [10]. To obtain the density field, we assumed the mass to be 1, which gives us a CG density function of:

$$\phi^n = \sum_{P=1}^{P} \Omega(x_p) \tag{1}$$

with ϕ^n the local density at position *n*, summed over all particles *P*, $\Omega(x_p)$, a Gaussian distribution with the mean placed on particle positions x_p and the standard deviation is the radius of the particles. The obtained particle density fields are then used to compute a mixing index. In this paper we chose to use the mixing index *MI* based on information entropy according to Schutyser et al. [11]. For our purpose, the calculation was slightly altered for a fully mixed system leading to *MI* and fully segregated system to be *MI*. For this we have to use the Boltzmann expression for entropy:

$$s^n = \phi_a^n \ln(\phi_a^n) + \phi_b^n \ln(\phi_b^n) \tag{2}$$

with ϕ_i^n the concentration of species *i* at position *n*. From this we calculate the system entropy by averaging the local sampled entropy:

$$S = \frac{1}{N}\sum_{1/n} \Phi^n s^n \tag{3}$$

Here, Φ^n is the local density of all particle species and *N* the total number of positions in the subdivided grid.

$$MI = \frac{S - S_s}{S_m - S_s} \tag{4}$$

For our geometry, to make the entropy scale from 0 to 1, we need to know S_s and S_m, a segregation of a known segregated and mixed system respectively. To get our mixing index *MI*, we use:

The mixing index is then calculated for all image frames to get the evolution of the mixing state of the system. For both the CG and the mixing index an in-house Matlab script was used.

III. Results and Discussion

A. Cohesive Force Measurment

Fig. 2 shows the cohesive force values between two particles as a function of silanization reaction time. This is determined using the microbalance setup. The cohesion forces increase with the silanization reaction time. This increased cohesive force can be attributed to the formation of more covalent bonds (-Si-O-Si-) as the reaction progresses.

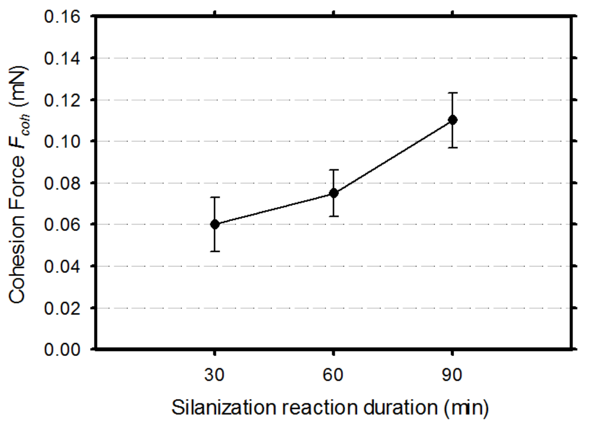

Figure 2. Cohesion force between two particles of radius 0.85 mm as a function of the silanization reaction time.

B. *Particulate Flow and Bulk Cohesion Characterization*

In addition to direct cohesive measurements, the cohesive force can be correlated with the angle of the flow (i.e. dynamic angle of repose) at the bulk level in a rotating drum. We show in Fig. 3 (a) the angle of the flow in a rotating drum as a function of time for non-cohesive and cohesive particles.

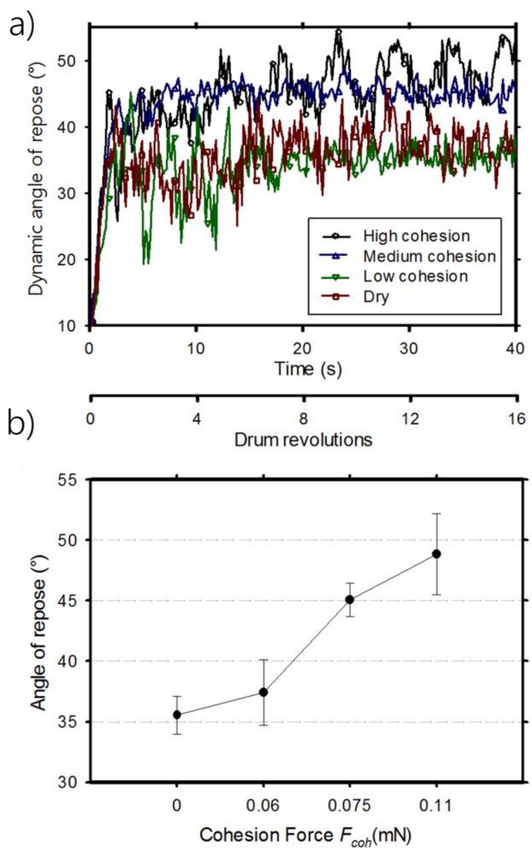

Figure 3. (a) Dynamic angle of repose θ as a function of time for dry and cohesive particles, b) Averaged dynamic angle of repose as a function of cohesive force.

The particles are of a radius 0.85 mm and the rotation speed of the drum is 25 rpm. Clearly, the cohesion has a significant effect on the bed flow motion. As the drum rotates, particles are lifted to the upper part of the bed and the angle of the slope increases until it reaches a maximum, then avalanches start to occur. We notice a variation on the avalanche amplitude and frequency depending on the cohesion force between the particles. This is similar to the effect of capillary forces on the flow of particles in a rotating drum [4], [5]. For medium cohesive particles the amplitude of the avalanches is the lowest indicating a liquid-like flow behavior. When the cohesion is low, the dynamic angle of repose is close to that of the non-cohesive case. Fig. 3 (a) shows that the dynamic angle of repose increases almost linearly with cohesive force confirming the results obtained by the micropositioner setup. As we increase the capillary force at a given rotation rate, the particles experience a greater capillary cohesion and they are dragged more to the upper part of the drum, causing an increase in the slope of the bed.

C. *Particles Mixing*

Next, by computing the mixing index (MI), we investigate the effect of cohesive forces on particles segregation. In all experiments, the drum contains a mixture of bidisperse 50-50% by weight (w/w) glass particles with the same density of 2500 kg/m^3.

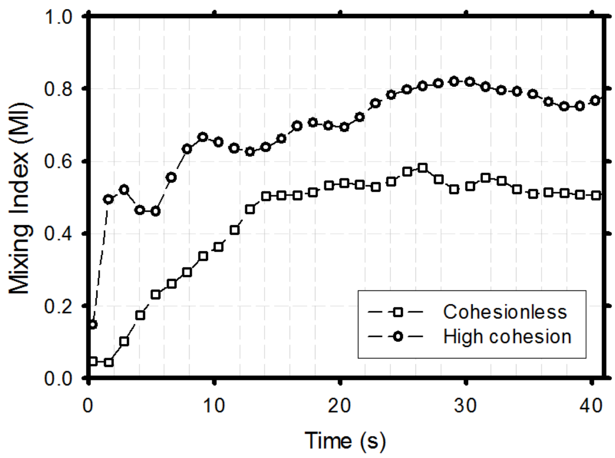

Figure 4. Mixing index as a function of time for non-cohesive system, and cohesive systems. The cohesive system is composed of cohesive particles of radius r_A = 0.85 mm and non-cohesive particles of radius r_B = 1.25 mm.

We show in Fig. 4 the mixing index (*MI)* as a function of time for non-cohesive particles of radius r_A = 0.85 mm mixed with non-cohesive particles of radius r_B = 1.25 mm. The mixing index curves are smoothed using Bisquare weighting smoothing to reduce the fluctuation of the mixing index. A mixing index close to 1 means the system is mixed, while an *MI* value close to 0 means a segregated system. For the cohesionless case, *MI* increases at first and then, after few rotation of the drum, remains relatively constant at around *MI* = 0.5 indicating segregation. When the same experiment is performed with cohesive particles, the results are different. Fig. 4 shows the mixing index *MI* of the case where particles of radius r_A = 0.85 mm are cohesive and mixed with non-cohesive larger particles of radius r_B = 1.25 mm. The value of the mixing index in this case is higher than for non-cohesive case, indicating better mixing with fewer large particles found in the outer region and more of them in the core of the bed. This demonstrates the ability of cohesion to improve particulate mixing in dry systems. The cohesive force clumps the small particles together and hence weakens the percolation segregation, which is in line with explanation given by Li and McCarthy [6] for the case of cohesion due to moisture between the particles.

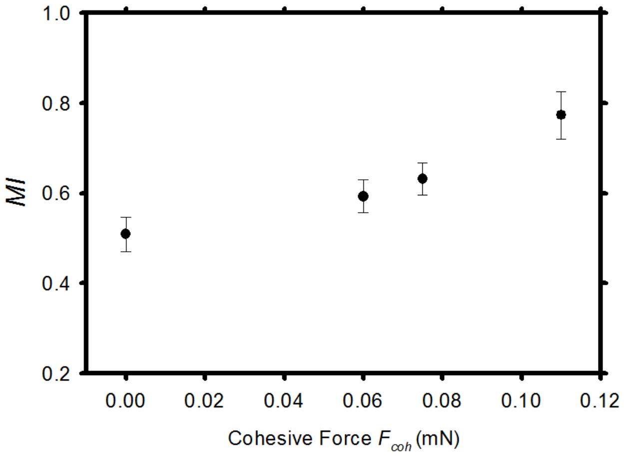

Figure 5. Mixing index versus cohesive force for a mixture of particles of radiuses r_A = 0.85 mm and r_B = 1.25 mm.

Fig. 5 shows the mixing index for a system composed of particles of radii r_A = 0.85 mm and r_B = 1.25 mm for different cohesive forces. As the cohesive force increases, segregation slows down. Cohesion plays a key role in improving the mixing of particulate systems through the formation of clusters. We infer that this makes the gaps between the large particles not wide enough for clumped cohesive particles to pass through, and hence improves mixing. Shinbrot *et al*. [12] addressed this mixing behaviour in the case of small cohesive particles (< 300 μm in diameter) and concluded that cohesion improves the mixing due to periodic stick–slip behaviour occurring in the flowing layer. These results show that the extent of the mobility of the non-cohesive particles depends strongly on cohesion, and, more importantly, the cohesion strength can be tuned to control segregation.

IV. Conclusion

We investigated the effect of cohesive forces between dry particles on the flow and mixing of particulate systems. We proposed an approach for tuning the cohesive force using extensive chemical salinization of the glass particles. Cohesive forces between the particles were measured using an in-house experimental device and characterized using the dynamic angle of repose in a rotating drum. We showed that the presence of cohesion increases the angle of the slope (i.e. angle of repose) and affects the avalanches amplitude and frequency. Introducing cohesive particles improves the mixing of the particulate system, suggesting that we could control the size segregation or the mixing of monosized particles simply by the right tuning of the cohesive forces. The work described here raises several perspectives for further research. For instance, a next step would be to explore the effect of cohesive force for larger particles and with different densities. Another interesting perspective is to use Discrete Element Method (DEM) to investigate the effect of cohesion on the mixing and on local clusters formation.

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