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Invited paper

## Validation of surface coating with nanoparticles to improve the flowability of fine cohesive powders

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### ABSTRACT

Fluidization of fine cohesive powders is seriously restricted by the strong interparticle cohesion. The rational combination of nanoparticles with fine cohesive powders is expected to obtain composite particles with improved flowability. In this work, we firstly reviewed the sandwich and three-point contact models regarding the fundamental principles of nano-additives in reducing cohesiveness. Based on these previous models, the effects of the size of nanoparticles, their agglomeration and coverage on the surface of cohesive powders in reducing interparticle forces were theoretically analyzed. To validate the theory effectiveness for the irregularly shaped cohesive powders, an extreme case of cubic powders coated with silica nanoparticles was fabricated, and the flowability of the composite particles was determined experimentally. Ultimately, based on force balance of a single particle, a semi-theoretical criterion for predicting the fluidization behavior of coated powders was developed to guide the practical applications of improving the flowability of cohesive powders through structural design and modulation.

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### Introduction

The flow behavior of particles depends on their intrinsic structures and morphologies. Based on the particle size and the density difference between solid material and fluidizing medium, Geldart has classified the powders into four groups: A, B, C, and D (Geldart, 1973). Group C powders, namely fine cohesive particles, have been broadly applied in pharmaceutical, food, chemical, and cosmetics industries. However, the strong interparticle adhesion force of these fine cohesive powders induces intense particle agglomeration, and further restricts their smooth fluidization and facile transportation (Wang, Zhu, & Beeckmans, 2000; Wang, Cheng, Jin, & Bi, 2007). Consequently, the handling of cohesive powders becomes a great scientific challenge and urgent technological issue for their bulk applications.

Group C cohesive powders, when subjected to fluidizing gas, tend to form cracks, channels, or even lift as slugs. Although agglomerate fluidization may be possible in some cases, due to the instability of the formed agglomerates, the bed preferentially dies in partial fluidization or even complete defluidization (van Ommen,

Valverde, & Pfeffer, 2012). To improve the flow quality of cohesive powders, two types of methods in terms of different mechanisms have been explored. The first approach intends to overcome the interparticle interactions by introducing external excitation, such as mechanical vibration (Kaliyaperumal, Barghi, Briens, Rohani, & Zhu, 2011; Nam, Pfeffer, Dave, & Sundaresan, 2004; Xu & Zhu, 2005, 2006), acoustic wave (Guo, Liu, Shen, Yan, & Jia, 2006; Liu, Guo, & Chen, 2007; Xu, Cheng, & Zhu, 2006), magnetic stirring (Yu, Dave, Zhu, Quevedo, & Pfeffer, 2005), rotating drum (Huang, Zhang, & Zhu, 2009; Huang, Zhang, & Zhu, 2010b), electric field disturbance (Lepek, Valverde, Pfeffer, & Dave, 2010), and pulsatile (Akhavan et al., 2009; Bizhaem & Tabrizi, 2013) or secondary gas flows (Quevedo, Omosebi, & Pfeffer, 2010; Quevedo & Pfeffer, 2010). However, additional processing steps and units, as well as high operation costs are inevitable in this case. The second method is the addition of flow aids (Meyer & Zimmermann, 2004; Zhou & Li, 1999), either coarser or finer particles, to break up the agglomerates or reduce the adhesion forces by optimizing surface properties. But the bed segregation occurs possibly due to the difference in particle size and density, thus reducing the efficiency in improving flowability.

Although nanoparticles serving as the flow aids can be introduced to improve the flowability of cohesive powders, the improvement depends on how well they mix with each other.

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**Nomenclature**

|             |  |
|-------------|--|
| $A$         | Hamaker constant, J  |
| $Bo_g$      | Granular Bond number   |
| $Co$        | Granular cohesive number                                       |
| $Co_{A-C}$  | Critical granular cohesive number                              |
| $Co'_{A-C}$ | Modified critical granular cohesive number                     |
| $C_D$       | Drag coefficient   |
| $d$         | Diameter of guest particle, m                                  |
| $d_{ag}$    | Diameter of guest particle agglomerate, m                      |
| $d_p$       | Particle diameter, m   |
| $D$         | Diameter of host particle, m                                   |
| $F_c$       | Interparticle adhesion force, N                                |
| $F_d$       | Drag force, N  |
| $F_{vdw}$   | van der Waals force, N   |
| $N$         | Number of guest particles on the surface of each host particle |
| $N_{ag}$    | Number of agglomerates on the surface of each host particle    |
| $\Delta P$  | Pressure drop across fluidized bed                             |
| $S$         | Cross-sectional area of fluidized bed                          |
| $SAC$       | Surface area coverage, %                                       |
| $SAC^*$     | Actual surface area coverage, %                                |
| $u$         | Superficial gas velocity, m/s                                  |
| $u_{mf}$    | Minimum fluidization velocity, m/s                             |
| $u_t$       | Terminal velocity, m/s   |
| $W$         | Weight percentage of guest particle, wt. %                     |
| $W_b$       | Buoyancy force, N  |
| $W_e$       | Effective weight, N  |
| $W_g$       | Gravity force, N   |
| $z_0$       | Minimum surface distance, m                                    |

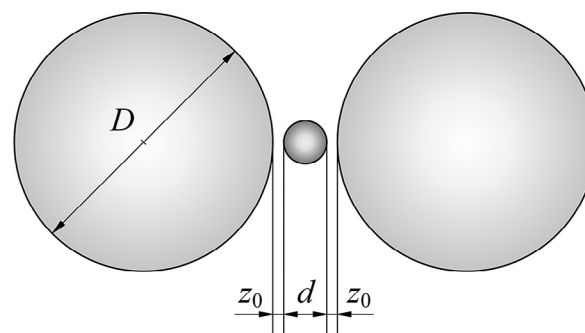
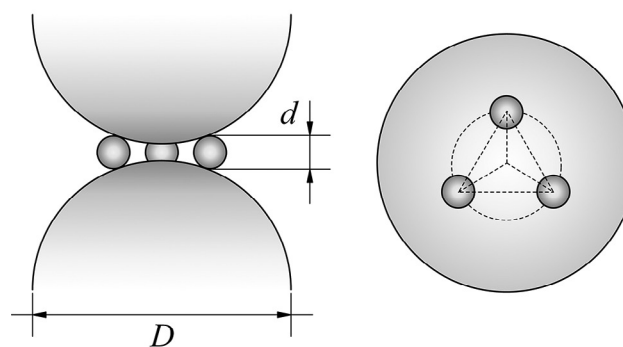
**Greek letters**

|                    |  |
|--------------------|--|
| $\varepsilon$      | Voidage of solids bed                        |
| $\varepsilon_{ag}$ | Voidage of guest particle agglomerate        |
| $\rho_d$           | Density of guest particle, kg/m <sup>3</sup> |
| $\rho_D$           | Density of host particle, kg/m <sup>3</sup>  |
| $\rho_f$           | Gas density, kg/m <sup>3</sup>               |
| $\rho_p$           | Particle density, kg/m <sup>3</sup>          |
| $\phi$             | Efficiency factor, %                         |

**Subscripts**

|             |   |
|-------------|---|
| d–D         | (Attraction) between guest and host particles |
| D–D         | (Attraction) between two host particles       |
| guest–guest | Guest–guest contact                           |
| guest–host  | Guest–host contact                            |
| host–host   | Host–host contact                             |

According to our previous experience, the nanoparticles are more easily to aggregate together than micron-sized cohesive powders because of their higher interparticle cohesion respect to gravity (Wang et al., 2007; Zhu, Zhang, Wang, & Wei, 2016). Some nanoparticles may even form a porous multi-stage agglomerate structure with the size up to several hundred microns (Huang, Wang, & Wei, 2008; Wang, Gu, Wei, & Wu, 2002). However, the particle agglomeration should be avoided for these nano-additives, given that it weakened their performance in reducing cohesiveness. Compared with simply blending the two kinds of powders together, coating the cohesive powders with nanoparticles can precisely deposit the nano-sized particles onto the surface of cohesive powders, reducing the interparticle adhesion force and improving the fluidization behavior more efficiently (Pfeffer, Dave, Wei, & Ramlakhani, 2001; Yang, Sliva, Banerjee, Dave, & Pfeffer, 2005). In addition, as the nanoparticles adhere to the surface of cohesive powders via van der

**(a) Sandwich contact model****(b) Three-point contact model**

**Fig. 1.** Van der Waals force models of two coated particles: (a) sandwich contact model (Xie, 1997) and (b) three-point contact model (Chen et al., 2008).

Waals attraction, which is up to a million times the powder gravity (Kendall & Stainton, 2001), the tendency of particle segregation is effectively inhibited.

In regard to the functional principles of nano-additives in improving powder flowability, it is widely accepted that van der Waals force decreases dramatically with increasing particle surface distance and dominates the interparticle forces for dry and neutral powders (Seville, Willett, & Knight, 2000; Visser, 1989). Therefore, most research groups believed that the nanoparticles acted as spacers, increased the surface distance between cohesive powders, and further reduced the interparticle adhesion force. To validate this theory, a series of research works have been conducted (Chen, Yang, Dave, & Pfeffer, 2008; Huang, Scicolone, Gurumuthy, & Davé, 2015; Xu, Zhang, & Zhu, 2009; Quintanilla, Valverde, & Castellanos, 2006). Especially, Xu et al. (2009) have successfully put this theory into practice by improving the flowability of powdered paints used in powder coating process (Huang, Zhang, & Zhu, 2010a). However, some researchers have questioned this mechanism and proposed different opinions, such as serving as lubricants to reduce the friction between cohesive powders (Podczeczek & Mia, 1996; Utermann, Aurin, Benderoth, Fischer, & Schröter, 2011), and functioning as neutralizers of electrostatic charges (Dutta & Dullea, 1990).

Up to now, a number of van der Waals adhesion force models have been established to elucidate the role of nano-additives as spacers, such as sandwich contact model (Xie, 1997; Xu et al., 2009) only probing the particle size effect, and three-point contact model (Chen, Quintanilla, Yang, Valverde, & Dave, 2009; Chen et al., 2008) further taking the surface area coverage (SAC) into account (see Fig. 1). However, both the contact models assumed a homogeneous coating of the guest nanoparticles onto the host powders, neglecting the possible agglomeration of nano-

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