Advanced Powder Technology 24 (2013) 560–564

Contents lists available at SciVerse ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Rapid Communication

Effects of the agglomerated states and the gap of coverage for admixed particles on particle-bed packing fractions

Mikio Yoshida *, Hiroaki Yamamoto, Jun Oshitani, Kuniaki Gotoh

Division of Chemical and Biological Technology, Graduate School of Natural Science and Technology, Okayama University, 3-1-1, Tsushima-naka, Kita-ku, Okayama-shi, Okayama 700-8530, Japan

ARTICLE INFO

Article history:
Received 21 August 2012
Received in revised form 5 December 2012
Accepted 6 January 2013
Available online 29 January 2013

Keywords:
Packing fraction
Admixture of nano-particle
Surface image analysis
Silica particle

ABSTRACT

One of the techniques used to decrease the cohesive force between particles is the admixing of nano-particles. However, the optimal conditions that will produce a minimum amount of force have not been established. In this study, we investigated the effects of the agglomerated state and the gap of coverage for admixed particles on particle-bed packing fractions in uni-axial compression. The main particles were made up of 397 nm silica particles. The admixed particles included 8, 21, 62 and 104 nm silica particles. The main and admixed particles were mixed using a mortar and pestle for 5 min for various mass ratios. SEM images were used to analyse the coverage diameter and the surface coverage ratio. As a result, the packing fractions with admixed particles of 8 and 21 nm were larger than when admixed particles were not used, and these admixed particles adhered onto the surface of the main particles as agglomerates. However, packing fractions of 62 and 104 nm were almost constant and were independent of the coverage states of admixed particles. Furthermore, these admixed particles with relatively larger diameters were adhered onto the surface as single particles. From the coverage diameter and actual surface coverage ratio obtained by the SEM image, the average gaps between agglomerates of 8 and 21 nm on the main particle were calculated. When the gap approached twice the size of the coverage diameter, packing fractions of 8 and 21 nm proved to be the maximum values. However, when the gap was less than the coverage diameter, the packing fractions deteriorated.

1. Introduction

Fine particles have a high reactivity and solubility due to a large specific surface area. Thus, they are widely used throughout the chemical, pharmaceutical, cosmetic, and food industries. Particles are known to become cohesive, however, when the diameter is decreased. The cohesiveness of particles causes trouble with handling processes such as feeding, mixing and packing. Therefore, techniques that decrease the cohesive forces are required for improving such handling processes of fine particles.

One of the techniques used to decrease the cohesive force is the admixing of nano-order particles. The admixed particles adhere onto the surface of a main particle, which results in an increased surface roughness [1–8]. Liu et al. [8] reported that a shear test showed that the combination of an ibuprofen particle and lactose nano-particles produced the minimum value for cohesiveness at an admixed mass ratio of about 50%. However, in a system comprised of a lactose particle and silica nano-particle, Ohta and Toyoshima [3] reported that one of the cohesive properties, the angle of repose, showed its minimum value at an admixed mass ratio of about 0.1%.

As shown in the literatures, the optimal admixed mass ratio for achieving a minimum cohesive property strongly depends on the combination of the main materials and that of the admixed particles. Furthermore, the van der Waals force, which is the dominating force of cohesive properties, depends on other particle characteristics, such as diameter, shape, hardness and elasticity [9]. Because of the complexity of the effects of these factors on cohesive properties, the optimal admixed ratio cannot be predicted, and then the optimal mixing conditions must be decided by trial and error. To predict the optimal mixing ratio, it is necessary to clarify the effect of each factor on the cohesive properties. However, there has been little investigation on the effect of each factor although there are many reports on the cohesive properties of various practical systems. Hence, we intended to investigate the effect of the diameters of main and admixed particles on cohesive properties. In order to clarify the effect of the diameter, we employed a simple system. We used spherical silica particles as both the main and the admixed particles and we focused on the cohesive properties that appeared in the packing process [10]. As a result, the details of the relationship between the coverage state

---

* Corresponding author. Tel./fax: +81 86 251 8085.
E-mail address: myoshida@cc.okayama-u.ac.jp (M. Yoshida).

0921-8831/$ - see front matter © 2013 The Society of Powder Technology Japan. Published by Elsevier B.V. and The Society of Powder Technology Japan. All rights reserved.
http://dx.doi.org/10.1016/j.apt.2013.01.004

Nomenclature

D	admixed particle diameter obtained by SEM (m)
D_{ad}	primary particle diameter of admixed particle (m)
G_{pp}	average gap between agglomerates of admixed particles adhered onto the main particles (m)
N	number of admixed particles on half surface area of a main particle (–)
R_m	mixing mass ratio (%)
R_{ac}	actual surface coverage ratio obtained by SEM (%)
R_{tc}	theoretical surface coverage ratio (%)
S_1	half surface area of a main particle (m^2)
S_2	product of N and S_a (m^2)
S_a	projected cross sectional area calculated using D (m^2)
$\Delta\phi$	difference of ϕ_{mix} subtracted from ϕ_{main} (%)
$\Delta\phi_{net}$	net packing fraction difference by admixture (%)
ϕ_{mix}	packing fraction with admixed particles (%)
ϕ_{main}	packing fraction when admixed particles were not used (%)

and the packing fraction were unclear, although we found that the admixed particle diameter was an important factor for increasing the packing fraction. Thus, in the present study, we measured the compressed packing fractions of the mixed particles for various admixed particle diameters and mixing mass ratios. In the experiments, we analyzed the 2-dimensional coverage diameter and the actual surface coverage ratio of admixed particles on main particles using SEM images. From these results, in particular, the effects of the agglomerated state and the gap of the coverage for admixed particles on the packing fraction were discussed.

2. Experiment

2.1. Mixing and compressing procedures

Spherical silica particles with a count median Martin diameter of 397 nm were employed as the main particles, while the same silica particles with diameters, D_{ad}, of 8, 21, 62 and 104 nm were used as admixed particles. Here, the sphere equivalent diameters were 8 and 21 nm, while the count median Martin diameters were 62 and 104 nm. These count median Martin diameters and sphere equivalent diameters were obtained by SEM image analysis and the BET method, respectively. The true density for all silica particles was 2.2×10^3 kg/m^3.

The admixed particles were mixed with the main particles using an alumina mortar and pestle for 5 min. The mixing mass ratio ranged from 0.5% to 33.3%. We placed 0.5 g of mixed particles into a cylindrical container with an inner diameter of 8.8 mm, and the mixed particles were then compressed from the top using 0.19 MPa. The packing fraction (=solid fraction), ϕ_{mix}, was calculated from an apparent volume of the compressed particle bed. As a control, only the main particle was stirred using a mortar and pestle for 5 min, then a packing fraction of 0.5 g of the main particle without the admixed particle, ϕ_{main}, was measured. Each experiment was repeated 3 times. From these results, we calculated the difference in packing fractions, $\Delta\phi = (\phi_{mix} - \phi_{main})$. Here, ϕ_{main} was 39.1 ± 0.7%.

The theoretical surface coverage ratio, R_{tc}, was calculated from mixing mass ratio, R_m. For the calculation, it was assumed that all of the admixed particles had adhered onto the main particles in a single layer. The coverage ratio was then defined as the ratio of the total projected area of admixed particles to the area required for the completion of a single layer with closely packed (triangle lattice) main particles. Hence, an R_{tc} of more than 100% indicated that the admixed particles had formed multiple layers on the main particle.

2.2. Definition of 2-dimensional coverage diameter and actual surface coverage ratio

The 2-dimensional coverage diameter of the admixed particles, D, and the actual surface coverage ratio of the admixed particles, R_{ac}, were evaluated by the following procedures. The mixed particles were observed using SEM (image magnification: about 100 K). In some of the experiments, the admixed particles adhered onto the main particles as agglomerates. Then, 80 of the agglomerates, or single admixed particles, onto a main particle were selected and their Martin diameters, D_i, were measured using image analysis software (i = 1–80). The 2-dimensional coverage diameter of an admixed particle, D, was defined by the average of the D_i.

Here, for simplification we assumed that the periphery shape of the agglomerates could be approximated by a circle. Then, a projected cross-sectional area of admixed particles, S_a ($=\pi D^2/4$), was calculated from D. On the other hand, 20 of the main particles were selected and their Martin diameters were measured. From these diameters, half of the surface area of the main particle, S_{1-j}, was calculated. The number of agglomerates of admixed particles on 20 of the main particles, N_j, was also counted (j = 1–20). The total projected area of admixed particles on each main particle, S_{2-j}, was calculated by multiplying N_j and S_a. Then, the actual surface coverage ratio of each main particle, R_{ac-j}, was calculated by dividing S_{2-j} by S_{1-j}. By averaging R_{ac-j}, the actual surface coverage ratio, R_{ac}, was calculated by the following equation.

$$R_{ac} = \frac{\sum_{j=1}^{20} \frac{S_{2-j}}{S_{1-j}}}{20} = \frac{\sum_{j=1}^{20} \frac{N_j S_a}{S_{1-j}}}{20} \tag{1}$$

3. Results and discussions

Fig. 1 shows the differences in packing fractions, $\Delta\phi$, as a function of the mixing mass ratio, R_m, at D_{ad} = 8 nm. Here, the error bars of white key indicate standard deviations for three independent

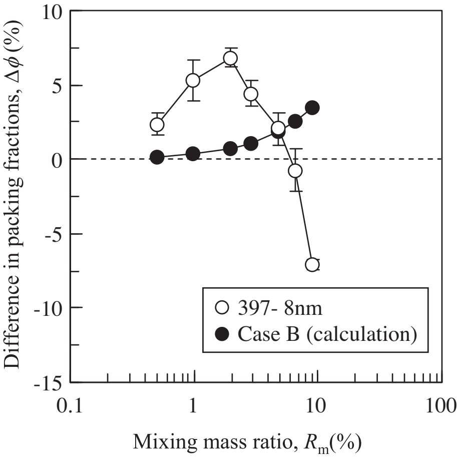

Fig. 1. Dependences of difference in packing fractions on mixing mass ratio (D_{ad} = 8 nm). (Case B: the admixed particles were simply placed in the void between the main particles with the same packing fraction as that of the control).