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Original Research Paper

Effects of main particle diameter on improving particle flowability for compressed packing fraction in a smaller particle admixing system

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ABSTRACT

Particle flowability can be improved by admixing particles smaller than the original particles (main particles). However, the mechanisms by which this technique improves flowability are not yet fully understood. In this study, we examined compressed packing in a particle bed, which is affected by particle flowability. To estimate the mechanism of improvement, we investigated the effects of the main particle diameter on the improvement of compressed packing fractions experimentally.

The main particles were 397 and 1460 nm in diameter and the admixed particles were 8, 21, 62, and 104 nm in diameter. The main and admixed particles were mixed in various mass ratios, and the compressed packing fractions of the mixtures were measured. SEM images were used to analyze the coverage diameter and the surface coverage ratio of the admixed particles on the main particles. The main particle packing fraction was improved as the diameter ratio (=main particles/admixed particles) increased. This was explained by a linked rigid-3-bodies model with leverage. Furthermore, the actual surface coverage ratio at which the most improved packing fraction was obtained decreased with increasing main particle diameter. This was explained by the difference in the curvature of the main particle surface.

1. Introduction

The specific surface area of particles generally increases with decreasing particle diameter, leading to higher reactivity and solubility. However, the adhesive force opposing the gravitational force for a particle increases with decreasing diameter [1]. Thus, fine particles have poor flowability causing problems with handling processes, such as compression, feeding, and transportation. In the production processes of pharmaceutical tablets, the poor flowability causes a non-uniform packing structure, which results in low tablet strength [2]. Hence, techniques for improving the flowability of fine particles are required.

The flowability can be improved by admixing particles smaller than the original particles (main particles) [2–9]. The admixed particles adhere to the surface of the main particles, which would decrease the adhesive force between the main particles. However, the flowability deteriorates in some conditions. It has not previously been possible to predict the properties of the admixed particles and admixing conditions that improve flowability. This is because the complex effects of various properties on the adhesive forces between main particles. The adhesive forces generally depend on a variety of particle characteristics, including the diameter, surface material (=surface free energy), shape, surface roughness, hardness, and elasticity [10]. The effects of each of the admixing factors must be investigated to establish a method for predicting the appropriate admixture particles and admixing conditions.

In previous experimental studies and simulations [11–13], we examined compressed packing as a process that is affected by the flowability, and investigated the effects of some characteristics of admixed particles (diameter, adhesive force, admixed mass ratio) on packing fractions by using silica particles as both the main and admixed particles. For example, in investigating the effects of admixed particle diameter experimentally [11], the 397/8 and 397/21 nm combinations improved the packing fraction of the main particles, and the increase in packing fraction depended on how the admixed particles adhered to the main particles, that is, the adhesion structure. However, the 397/62 and 397/104 nm combination did not show improvements. In the simulation [13],

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Nomenclature

d	separation distance between force point and fulcrum point in a leverage (m)
D	particle diameter (m)
D_c	admixed particle coverage diameter obtained by SEM (m)
F	force (N)
H_j	half the surface area of a main particle calculated by j-th main particle Martin diameter (m^2)
m	mass (kg)
N_j	number of admixed particle on half the surface area of j-th main particle (–)
N_m	number of main particles used for calculated R_{ac} values (–)
P	fulcrum point in a leverage (–)
r	rotational radius from fixed O' position (m)
R_{ac}	actual surface coverage ratio obtained by SEM (%)
R_m	mixing mass ratio (%)
R_{tc}	theoretical surface coverage ratio obtained by a calculation (%)
S_c	coverage area of admixed particle (m^2)
$\Delta\phi_{net}$	net difference in packing fraction of main particle by admixture (%)
ϕ	particle packing fraction (%)

Subscripts

AP	admixed particle
MIX	mixture particle (main and admixed particles)
MP	main particle

we proposed additional mechanism to the flowability improvements for the main-admixed particle system, which is based on changing the configuration to a linked rigid-3-bodies model.

In this study, we focused on the effects of main particle diameters on compressed packing fractions in a system of particles admixed with smaller particles. This is because changing many parameters, such as the curvature of the surface, void space size between main particles, and inertia force and moment, affects the appropriate admixed conditions at which obtained the greatest improvement in the flowability for packing fraction is observed. To investigate the effects of the main particle diameter easily, the morphological and mechanical characteristics should be kept the same. Thus, we used similar main-admixed particle combinations as in our previous study [11], in which 1460 nm silica particles (main particles) were admixed with 8, 21, 62, and 104 nm silica particles (admixed particles) in various mixing mass ratios. Then, the compressed packing fraction of the mixture was compared with the previous results for the 397 nm main particle diameter [11], and we discussed the effects of the main particle diameter on the improvement in the particle flowability for packing fraction.

2. Experiment

2.1. Samples

Spherical silica particles of diameter $D_{MP} = 397$ and 1460 nm were used as the main particles, whereas silica particles of $D_{AP} = 8$, 21, 62, and 104 nm were used as the admixed particles. Here, 8 and 21 nm were the sphere equivalent diameters determined by the BET method, whereas 62, 104, 397, and 1460 nm were the count median Martin diameters calculated from scanning electron microscopy (SEM) images. The true density for all silica particles was 2.2×10^3 kg/m^3.

2.2. Compression procedures and image analysis method

The compression procedures and image analysis methods were the same as those used in previous studies [11,12] and are only outlined here. The admixed particles were mixed with the main particles using a mortar and pestle for 5 min. The mixing mass ratio, R_m, ranged from 0.5% to 18.9%. A cylindrical container of 8.8 mm inner diameter with a filter was filled with the mixture particles. After leveling the particle layer surface by tapping the container, the mixture particles were compressed by applying 0.19 MPa of pressure on top of the layer. The packing fraction (= solid fraction) of the main particles, ϕ_{MP}, was calculated as follows:

$$\phi_{MP} = \phi_{MIX}\left(\frac{100 - R_m}{100}\right) \qquad (1)$$

Here, ϕ_{MIX} is the packing fraction of mixture particles in the compressed particle bed.

As a control, a packing fraction of the main particles without the admixed particles, $\phi_{MP}|_{R_m=0}$, was also measured. Each experiment was repeated three times. To discuss the effects of the main particle diameter, we calculated the net differences in the packing fractions of main particles caused by the admixed particles, $\Delta\phi_{net}$ [11,12]. $\Delta\phi_{net}$ was calculated by

$$\Delta\phi_{net} = \phi_{MP} - \phi_{MP}|_{R_m=0} \qquad (2)$$

$\Delta\phi_{net} > 0$ indicates an improvement in the packing fraction for the main particles. In this experiment, we used a fixed compression pressure, as described above. Thus, $\Delta\phi_{net} > 0$ also implies an improvement in the flowability of the main particle. Furthermore, to understand the admixed mass ratios, R_m, more easily, R_m was converted to a theoretical surface coverage ratio, R_{tc} [11]. For the calculations, it was assumed that the admixed particles would form a monolayer on the main particles when the total projected area of admixed particles was equal to the area of a single layer of closely packed main particles in a triangle lattice arrangement. Hence, an R_{tc} of more than 100% indicates that the admixed particles may form multiple layers on the main particles.

From the SEM images, 80 agglomerated or single admixed particles on the main particle were selected and their Martin diameters were measured by image analysis software. The average value of the Martin diameters was defined as the admixed particle coverage diameter, D_c. Furthermore, the number of agglomerates of admixed particles on the main particles was also counted. From these results and the Martin diameters of the main particles, the actual surface coverage ratio, R_{ac}, was calculated by [11,12]

$$R_{ac} = \frac{\sum_{j=1}^{N_m} \frac{N_j S_c}{H_j}}{N_m} \qquad (3)$$

where N_j is the number of admixed particles on half the surface area of a main particle, H_j is half the surface area of a main particle calculated by main particle Martin diameter, S_c is the coverage area of admixed particles ($=\pi(D_c)^2/4$), and N_m is the number of main particles used for this analysis.