Advanced Powder Technology 26 (2015) 626–631

Contents lists available at ScienceDirect

Advanced Powder Technology

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

Original Research Paper

Effects of an adhesive force of admixed particles on compressed packing fractions in a particle bed

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ARTICLE INFO

Article history:
Received 25 August 2014
Received in revised form 16 January 2015
Accepted 22 January 2015
Available online 7 February 2015

Keywords:
Packing fraction
Compressed particle bed
Admixture of nano-particles
Chemical surface modification

ABSTRACT

One of the techniques for a reduction in particle cohesiveness is the admixing of nano-particles. However, the mechanism of cohesiveness reduction has not yet been clarified. In this study we focused on a compressed packing fraction as one of the values reflecting the cohesiveness and flowability in a compressed particle flow. In order to estimate the mechanism of the reduction, the effects of an adhesive force of admixed particles on the packing fraction were investigated experimentally.

In the experiments, silica particles with 397 and 8 nm diameters were used as the main and the admixed particles, respectively. The surfaces of the admixed particles were modified chemically in order to vary their adhesive force with maintaining morphological and mechanical characteristics.

At more than 2.0% mixing mass ratios, the decreasing rate with increasing the mass ratio in net difference value of packing fraction for modified admixed particles was smaller than that for the raw particles. A calculation of the packing fraction of the admixed particles in the voids of main particles revealed that difference of the decreasing rates could be attributed to the difference of cohesivenesses between raw and modified admixed particles.

The maximum packing fraction achieved by the modified admixed particles was lower than that achieved by the raw admixed particles. However, when a mixture of main and admixed particles was packed by tapping, the packing fraction with modified admixed particles was higher than that with raw admixed particles. This implied that the adequate adhesive force of admixed particles is different by the applied compression force value.

1. Introduction

Fine particles have high reactivity and solubility due to a large specific surface area. However, fine particles also are highly cohesive due to an adhesive force that is high relative to the force of gravity [1]. This cohesiveness causes poor flowability during handling processes such as feeding or transportation. It also causes a low packing fraction during the compression process. These properties restrict the merits of fine particles. For example, fine aluminum particles are used as solid rocket fuels. However, high cohesiveness leads to a decrease in combustion efficiency [2]. In the production processes of tablet pharmaceuticals, high cohesiveness causes a non-uniform packing structure, which results in a shortage of tablet strength [3]. Therefore, there is a need for techniques that will reduce the cohesiveness of fine particles.

One of the techniques for a reduction in cohesiveness is admixing with nano-order particles [3–10]. It is believed that the adhesive force between main particles is decreased via the admixing of particles. The force generally depends on various particle characteristics such as diameter, surface material (=surface free energy), shape, surface roughness, hardness and elasticity [11]. Because of the complexity of the effects of these parameters on the adhesive force between main particles, adequate admixing particle characteristics and conditions have never been predicted. In order to establish a prediction method for adequate particles and conditions of admixing, the effect of each factor for the admixed particle must be investigated.

In our previous work [12], we focused on a compressed packing fraction as one of the values reflecting the cohesiveness and

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http://dx.doi.org/10.1016/j.apt.2015.01.014
0921-8831/

Nomenclature

D	admixed particle diameter obtained by SEM (m)
m_{mp}	the mass of the main particle in the compression test (kg)
N_j	the number of admixed particles on half surface area of a main particle (−)
N_m	the number of main particle used for calculating R_{ac} (−)
R_{ac}	actual surface coverage ratio obtained by SEM (%)
R_m	mixing mass ratio (%)
R_{oc}	occupied volume ratio of admixed particles (%)
S_{1-j}	half surface area of a main particle (m^2)
S_a	projected cross sectional area calculated using D value (m^2)
$V_{oc\text{-}ap}$	apparent volume of admixed particles at a R_m (m^3)
$V_{vo\text{-}mp}$	void volume of compressed main particle bed without admixed particles (m^3)
$\Delta\phi_{c-t}$	difference of compressed and tapped packing fractions (%)
$\Delta\phi_{net}$	net difference in packing fraction of main particle by admixture (%)
ϕ	packing fraction of mixture particles (%)
ϕ_{ap}	packing fraction of admixed particle at 0.19 MPa compression (%)
ϕ_{mp}	compressed-packing fraction of main particle without admixed particle (%)
ρ_p	true density of silica particle (kg/m^3)

flowability in a compressed particle flow, and we investigated the effect of the diameters of admixed particles on packing fractions by using silica nano-particles with diameters of 8, 21, 62 and 104 nm as admixed particles and a spherical silica particle of 397 nm as a main particle. During these investigations, the 8 and 21 nm admixed particles improved the packing fraction of the main silica particle. The admixed particles accomplished the improvement by adhering on the main particle as agglomerations. The highest packing fraction was obtained when the gap between adherent agglomerates was about twice the length of the diameter of the agglomerate.

In the next step, we focused on an adhesive force of admixing particles. One of the best ways to investigate this effect is using admixing particles with different adhesive force, and having the same morphological and mechanical characteristics (e.g. shape, diameter, hardness and elasticity). In order to come close to the ideal experimental situation, we employed an admixed particle with a surface that had been chemically modified. This is because an adhesive force depends on surface material. The surface-modified admixed particles were mixed with the main silica particles that had diameters of 397 nm, which was the same as in a previous study [12]. Then, the compressed packing fraction of the mixture was compared with the previous results for the mixing of main and raw admixed particles, and we discussed the effect of the adhesive force of admixed particles on the packing fraction.

2. Experiment

2.1. Samples and preparation of the modified admixed particles

As main and admixed particles, we used silica particles with a count Martin diameter of 397 nm and sphere equivalent diameters of 8 nm (by BET method), respectively. The true density for both types of silica particles was 2.2×10^3 kg/m^3.

In order to investigate the effect of the adhesive force of an admixed particle on improving the packing fraction, the surfaces of the admixed particles were modified chemically. The chemical modification was carried out as follows.

The admixed particles were sonicated using a supersonic sonicator for 10 min in an ethanol, acetone and 5 vol% hydrogen peroxide solution. After sonication, the particles were rinsed with pure water. Rinsed silica particles were immersed into a 0.2 vol% acetic acid solution. Then, 20 mmol of the silane coupling agent solutions were slowly added with stirring. Here, n-propyltrimethoxysilane was used as a silane coupling agent. After stirring for 1 h, the modified particles were rinsed with pure water and acetone and heated at 120 °C for at least 12 h.

Here, we intended to confirm how the surface modification had affected the adhesive force. It was, however, difficult to measure the adhesive force of an 8 nm particle. Therefore, we used Silicon plates that were chemically modified by the same procedures for the particles. For a measurement of the adhesive force, we used an AFM apparatus (MultiScan AFM, BMT). A cantilever made of Si_3N_4 (CSC38, MikroMasch) was used, and the maximum load between the cantilever and the plates was controlled at 15.6 nN. Fig. 1 shows the distributions of adhesive force for the raw and modified Silicon plates. The median forces for the raw and modified plates were 19.7 and 7.1 nN, respectively. This result implies that the adhesive force of the modified admixed particles were at least several-fold smaller than that of the raw admixed particles.

2.2. Compression procedures and image analysis methods

The compression procedures and image analysis methods were the same as used in previous work [12]. Hence, these methods are only outlined here. The raw or modified admixed particles were mixed with the main particles for 5 min. The mixing mass ratio, R_m, ranged from 0.5 to 9.1%. A polypropylene cylindrical container with an inner diameter of 8.8 mm was filled with 0.5 g of mixed particles. Then, the container was tapped to

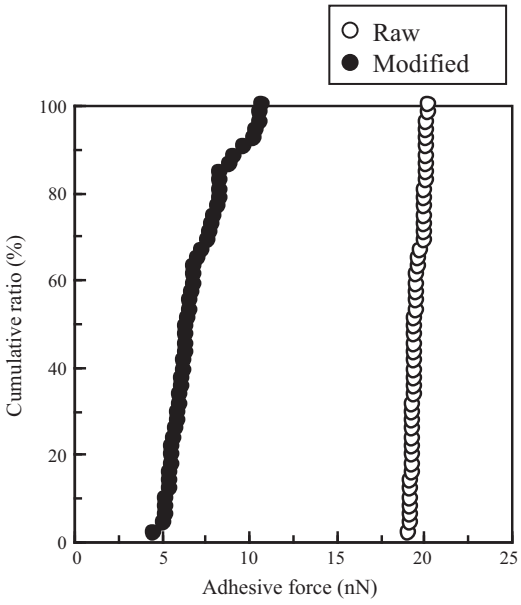

Fig. 1. Adhesive force distribution for raw and modified Si plates.