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Original Research Paper

Rebound behavior of nanoparticle-agglomerates

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ABSTRACT

In general, the rebound behavior of particles depends on the particle/substrate material combination and the particle size. In the present investigation the rebound behavior of nanoparticle agglomerates is investigated in a low pressure impactor and compared to single spherical particles. For agglomerates, their structure and mechanical strength will also affect the rebound behavior. The rebound of openly structured agglomerates (fractal dimension $D_f < 2$) is determined by the primary particle size and the particle-substrate combination. The impact velocity required for rebound (critical velocity) is independent of the agglomerate size and equal to the critical velocity of single spherical particles having the same size as the primary particles. In case of agglomerate fragmentation no rebound was observed for openly structured agglomerates. For denser agglomerates ($D_f > 2$), the critical impact velocity decreases with increasing agglomerate size, where the decrease is more accentuated for higher fractal dimensions, finally approaching the behavior of spheres.

1. Introduction

In the collision of particles with substrates rebound may occur even for nanoparticles thereby affecting measurement techniques such as cascade impactors [1] as well as deposition methods for nanostructured devices [2–5], where bouncing lowers the efficiency of the manufacturing process. Whether a particle sticks or rebounds from substrates depends on the balance between the kinetic energy of the impacting particle and the energy consuming processes, like adhesion, plastic deformation of the particle and/or the substrate and other processes as outlined by Dahneke [6]. The rebound of single spherical particles in the size range down to 0.8 μm has been experimentally investigated in several studies [7–12]. The observed critical impact velocities required for rebound increase with decreasing particle size with typical values in the range of 10 m/s for particle diameters of about 1 μm. An extrapolation from the measured values for micrometer particles leads to an expected critical impact velocity of about 1000 m/s for a 10 nm particle [10]. However, Arffman et al. [13] and Rennecke and Weber [14] extended recently the experimentally investigated size range down to 10 nm and found that the critical velocities for nanoparticle rebound are also in the range of 10 m/s –40 m/s. The discrepancy between the predicted critical impact velocities for nanoparticles from the early experimental investigations and the experimentally determined values can be explained by the different material combinations, which were used in the studies. Another effect, which may be superimposed to the material effect, is a possible pressure dependence of the escape probability [15]. Moreover, for individual nanoparticles a transition from elastic to plastic deformation with decreasing particle size was observed changing the slope of the critical impact velocity vs. particle size [14]. Awasthi et al. [16] carried out molecular dynamics simulations of the impaction of atomic cluster and observed also a transition between elastic and plastic deformation with increasing impact velocity leading to enhanced energy dissipation. The overall energy loss during rebound is represented with the coefficient of restitution, i.e. the ratio of rebound velocity and impact velocity, which itself depends on the impact velocity [17]. In general, the critical impact velocity required for rebound depends on the mechanical material properties of particle and substrate, which determine the deformation characteristics during the impaction, and on the adhesion energy between particle and substrate.

While the understanding of the rebound behavior of single spherical nanoparticles has progressed substantially over the last few years, the rebound characteristics of nanoparticle agglomerates, which are encountered in typical aerosol synthesis [18], are scarcely studied so far. Beside the already mentioned energy dissipation mechanisms, which affect the critical impact velocity, nanoparticle agglomerates can dissipate energy also due to internal restructuring, fragmentation [19–24] or mutual loading

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Nomenclature

Symbol	Meaning
A	Hamaker constant
C_c	slip correction
D_f	fractal dimension
E	particle surface interaction energy
E_{ad}	adhesion energy
E_{asp}	energy consumption due to plastic deformation of surface asperities
E_{CS}	adhesion energy
E_i	Youngs modulus of material i
E_P	energy consumption due to plastic deformation
e_n	normal coefficient of restitution
k_i	mechanical constant of material i
k	prefactor
m	mass
m_{aggl}	agglomerate mass
m_{PP}	primary particle mass
N_{PP}	number of primary particles in an agglomerate
p	exponent to describe the dependency of the normal coefficient of restitution on the impact velocity
S	slope
Stk	Stokes number
U_{gas}	gas velocity
v_{cr}	critical impact velocity
v_{0z}	impact velocity
v_y	onset impact velocity for yielding
x	particle diameter
x_m	mobility diameter
x_{PP}	primary particle diameter
Y	yield pressure
Z	minimum separation distance (0.4 nm)
β	exponent
δ_i	exponent
$\gamma_{i,j}$	surface energy at the interface i,j
υ_i	poisson ration of material i
ρ_{bulk}	bulk density
ρ_{eff}	effective density
ρ_P	particle density

between primary particles. Ihalainen et al. [25] observed rebound of agglomerates from substrates and possible fragmentation, where the total bounced mass fraction increased initially with increasing impact velocity but leveling off at higher impact velocities, which was attributed to the onset of fragmentation and plastic deformation.

Besides the interparticle forces also the coordination number, i.e. the average number of nearest neighbors of the primary particles, determines the mechanical strength and thereby the rebound behavior. Thus, restructuring of open agglomerates towards more compact morphologies, as characterized by an increasing fractal dimension D_f, is accompanied with an increase of the coordination number leading to improved agglomerate rigidity [26]. However, depending on the restructuring mechanism, interparticle bond strengths may stay constant (e.g. in spray-drying) or can increase during restructuring (e.g. sintering).

This article focuses on the influence of the fractal dimension on the rebound of nanoparticle agglomerates in the size range from 30 nm to 400 nm impacting perpendicularly onto surfaces. As the critical impact velocity for rebound also depends on the particle/-surface material combination, different particle materials (platinum and silica) and different targets (copper and mica) have been used. In addition, the bouncing behavior of agglomerates is compared to single spherical particles of the same material.

2. Theoretical section

2.1. Single particles bouncing

The bouncing behavior of single particles impacting on a surface is shown in Fig. 1 as a function of the impact velocity for different particle materials and sizes. It is generally observed that below a certain impact velocity (cf. inset in Fig. 1 regime I), i.e. the critical velocity v_{cr}, no rebound occurs. Above the critical velocity, the rebound efficiency, indicated by the normal coefficient of restitution e_n, increases rapidly with the impact velocity. In this regime (regime II) the energy dissipation channels are rather constant and the additional kinetic impact energy is transformed into kinetic energy of the rebounding particle [27]. However, for further increasing impact velocities, the plastic deformation of the particle or the substrate sets in and counteracts the efficient bouncing. For this regime (regime III), macroscopic models predict a relationship between coefficient e_n of restitution and normal impact velocity v_{0z} of the form $e_n \sim v_{0z}^{-p}$, where for a Hertzian contact p = 1/4 and for finite-element simulations p = 1/2 [17]. A p = 1/2 was also found by Schöner et al. [28] where the bouncing behavior of solid spherical silver particles of different sizes were investigated with molecular dynamics (MD) simulations and experimentally in a single stage low pressure impactor. MD-Simulations by Ayesh et al. [17] for solid and liquid bismuth (Bi) particles of about 3 nm show a nearly inverse relationship (p = 1), which was also determined experimentally for larger solid Bi particles (d_p = 32 nm). However, Ayesh et al. [17] investigated the bouncing behavior during oblique impaction. Due to the different loading caused by the tangential velocity component more energy is dissipated leading to a stronger decrease of the coefficient of restitution.

In the regime of plastic deformation, for Ag particles all values of e_n fall, within experimental and simulation uncertainties, onto one master curve, at least for particles larger than about 15 nm. For solid Bi nanoparticles a similar behavior is observed in experiments and simulations as also shown in Fig. 1, however, at different absolute values. The stronger dependence of the coefficient of restitution on the impact velocity for nanoparticles compared with macroscopic systems together with the known increase of the yield strength of nanoparticles [29], underline the special mechanical behavior of nanoparticles. Once the particles have reached the onset velocity for yielding (v_Y), the plastic deformation with impact velocity is much stronger than in the macroscopic case. In turn it means that nanoparticles behave elastically up to much higher loadings than macroscopic counterparts. The onset velocity for yielding (v_y) can be estimated with equation Eq. (1) derived by Wang and John [30].

$$v_Y = \sqrt{\left[\frac{\pi^8 \cdot Y^5 \cdot (k_s + k_p)^4}{40 \cdot \rho_P}\right]} \quad (1)$$

Here, Y denotes the yield pressure, k_i the mechanical constant of the substrate or the particle, which can be calculated with Eq. (5), where ν is the Poisson ratio and E the Young's modulus. ρ_P is the density of the particles.

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