Technical note

Effect of the surface energy of particle materials on the accommodation of gas molecules to the particle surfaces

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ABSTRACT

In this study, the slip correction factor is assessed from the measurements of the electrical mobilities of PSL, SiO_2 and Au nanoparticles under reduced pressure conditions. The asymptotic value ($\alpha+\beta$) of the slip correction factor, which is generally used for evaluating the particle drag force, are obtained for three different particle materials, and the values are used to assess the momentum accommodation function. Our experimental results show that the gas molecular slip is strongly dependent on the particle material characteristics. The higher the surface energy of the particle materials (PSL: 0.06 J/m², SiO_2: 0.30 J/m², and Au: 8.78 J/m²), the more diffusive reflections observed in gas molecular collisions with nanoparticles (PSL: 87.3%, SiO_2: 89.2%, and Au: 99.6%).

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1. Introduction

Nanoparticle motion in a gaseous medium has long been studied to understand nanoparticle behaviors in various operational conditions such as size measurement, particle sampling, low pressure synthesis, and semiconductor processing. In particular, the drag force of nanoparticles in the transition and free molecule regimes is an interesting topic in particle physics. Generally, the Stokes–Cunningham equation has been used to assess particle motions in a gas fluid (Cunningham, 1910).

$$F_D = \frac{-3\pi\mu v D_p}{C(Kn)}, \tag{1}$$

$$C(Kn) = 1 + AKn, \tag{2}$$

where D_p is a particle diameter, μ is the gas viscosity, v is the particle velocity relative to the fluid, C is the slip correction factor, Kn is the Knudsen number ($Kn = 2\lambda/D_p$), λ is the mean free path of gas molecules. A is the slip correction parameter, and it can be expressed as

$$A = \alpha + \beta\exp(-\gamma/Kn) \tag{3}$$

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where α, β, and γ are constants determined experimentally. Use of the slip correction parameter in assessing the particle drag force is critical for the high Kn regime, especially for the Kn higher than 1. Many results (Allen & Raabe, 1985; Eglin, 1923; Hutchins et al., 1995; Ishida, 1923) have been reported for the development of the slip correction parameter since Millikan (1923) first reported the constants from his classical oil-drop works. Recently, Kim et al. (2005) reported the slip correction constants by analyzing the electrical mobility of polystyrene latex (PSL) nanoparticles under reduced pressure conditions, and the constant values were further confirmed by Jung et al. (2012) and Kim et al. (2005) with the current smallest PSL spheres (SRM 1964).

In this study, the electrical mobility of PSL, SiO_2 and Au nanoparticles, which have different intermolecular potential energies, was measured using a nano-differential mobility analyzer (NDMA) and an electrospray system. As a control parameter to change Kn, the pressure of the NDMA system was controlled to the free molecule regime. From the experimental data, the values of $(\alpha+\beta)$ for these particle materials were determined. The values were further used for the drag force equation to interpret gas molecular accommodation on particle surfaces, and their discrepancy could be attributed to the difference of the gas–nanoparticle interactions. No prior experimental study that the surface energy of particle materials contributes critically to the gas molecular accommodation on particle surface due to the gas–particle interactions has been reported. Furthermore, the effect of momentum accommodation on the motion of nanoparticles is not well understood. In this article, we report that the gas molecular accommodation phenomenon increases with increasing binding energy (surface interaction) between gas molecules and particles based on experimental measurements.

2. Material and methods

2.1. Particle generation

Dehumidified air was utilized for the electrospray system (TSI 3480) incorporated with a soft x-ray charge neutralizer (HCT 4530). Suspension solutions of the PSL 19.62 nm (Duke Scientific 3020A), SiO_2 25.85 nm (Polysciences 24298), and Au 30.73 nm (NIST SRM 8012) nanoparticles were used for aerosolization. Before aerosolization by electrospraying the suspension solutions, one drop of the particle suspensions (PSL or SiO_2) was diluted to a 1 ml vial of ammonium acetate 20 mol/m^3 buffer solution with conductivity of 0.2 S/m. For the Au suspension solution, a salt removal process was conducted to remove the citrate stabilizer, and then ammonium acetate 2 mol/m^3 buffer solution (conductivity: ~0.02 s/m) was added to a 1 ml vial (Tsai et al., 2008a, 2008b) with a pH control to pH 8. Finally, the aerosolized nanoparticles were neutralized for the Boltzmann charge distribution through the soft x-ray neutralizer for the following DMA measurements.

2.2. Measurements

The measurement procedure consists of two sequential steps: (1) determining the particle diameter using electrical mobility measurements at ambient pressure, and (2) determining the slip correction factor (Eq. (2)) as a function of Kn at reduced pressures.

By a force balance between the Stokes drag and the electric field acting on a particle, one can obtain the following expression for the particle mobility Z_p (particle velocity in the field direction per unit electric field strength):

$$Z_{\mathrm{p}} = \frac{eC(Kn(D_{\mathrm{p}}))}{3\pi\mu D_{\mathrm{p}}},$$ (4)

where e is the electron charge, 1.602176×10^{-19} kg m^2 s^{-2} V^{-1}. In addition, the particle mobility for a spherical particle in a cylindrical DMA can also be expressed as follows:

$$Z_{\mathrm{p}} = \frac{Q}{2\pi VL}\ln(r_2/r_1),$$ (5)

where Q is sheath flow rate, V is center rod voltage, L is characteristic length of a DMA, and r_1, r_2 are inner and outer radius of a DMA, respectively. In Eq. (5), Q and V are only experimental parameters, and others are geometrical constants. Particle mobility distribution is measured with a DMA (HCT 4220) and a condensation particle counter (CPC, HCT 4312) by varying the center rod voltage at a fixed sheath flow rate and at atmospheric pressure. From the mobility distribution profile, the peak value is obtained, and it is further used for calibrating the particle size (D_{p}) using Eq. (4). In this study, we used the slip correction factor ($\alpha=1.165$, $\beta=0.480$, $\gamma=1.001$) reported by Jung et al. (2012) in the calibration because the particle size of interest is in the same range.

Once the particle size is calibrated at atmospheric pressure, we can get the slip correction factors measured for various reduced pressure conditions. By equating Eqs. (4) and (5), the slip correction factor in a DMA can be expressed as

$$C = \frac{QG_f 3\pi\mu D_{\mathrm{p}}}{Ve},$$ (6)

where G_{f} is $\ln(r_2/r_1)/2\pi L$, combination of geometric factors. From Eq. (6), the slip correction factor can be obtained by measuring peak voltages V for flow conditions Q and particle diameters D_{p} at specific pressure conditions. Then, values of the slip correction factors are analyzed as a function of Kn, and the slip correction factors are further processed for the correction parameter A by the nonlinear least square fitting method for α, β, and γ in Eq. (3).