DRAG OF NANO-PARTICLES IN THE TRANSITIONAL FLOW REGIME

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Abstract. This paper presents measurements of the aerodynamic drag on spherical polystyrene latex aerosol particles of 84.4nm and 104nm diameter in air at Knudsen numbers of 1.29 and 1.59. A differential mobility analyzer and condensation particle counter is used as an aerosol particle size spectrometer to measure the drag force on these particles. This measurement is compared with drag estimations using free molecular flow equations for both diffuse and specular boundary conditions. In both cases the experimental drag in the transitional regime was overestimated by the free molecular diffuse calculation, and underestimated by the specular calculation. It is concluded that the interaction of air molecules at near atmospheric pressure with 84.4 and 104 nm polystyrene latex spheres is not completely diffuse.

INTRODUCTION

Experiments were conducted to measure the drag on 84.4 and 104 nm diameter polystyrene latex (PSL) nano-spheres from Duke Scientific in air at near atmospheric pressure. The experimental apparatus consists of a TSI model 3076 Constant Output Atomizer, TSI 3062 Diffusion Dryer, TSI 3080 Electrostatic Classifier with 3081 Long Differential Mobility Analyzer (DMA), and TSI 3760A Condensation Particle Counter (CPC). The atomizer and diffusion dryer are used to generate a nearly monodisperse aerosol, while the electrostatic classifier, DMA, and CPC are used as an aerosol particle size spectrometer. The particle size spectrum allows the calculation of the aerodynamic drag force on the particle.

To generate the aerosol, the atomizer creates a fine water mist from a solution of distilled, de-ionized water mixed with solid PSL nanospheres. This creates a stable aerosol of water droplets that contain solid PSL spheres within them. The aerosol is then passed through a diffusion dryer that removes the water from the PSL spheres, resulting in a nearly monodisperse aerosol of solid particles. Deviations from monodispersity are a result of the drying of water droplets that contain multiple PSL spheres, causing agglomerated particles to form, and the drying of droplets containing impurities, forming very small aerosol particles.

TRANSITIONAL REGIME FLOW PAST A SPHERE

Using Stokes continuum flow, the drag on a sphere in a viscosity dominated flow field is

$$F_{Stokes} = 3\pi\mu VDp$$

Where μ is the gas viscosity, V is the sphere velocity, and Dp is the diameter of the sphere. For transitional small scale flows between the continuum and free molecular regimes, an experimentally determined correction factor is applied to Stokes flow for spheres. This Cunningham slip factor is a function of the mean free path of the gas molecules and the radius of the sphere[1]:

$$Cc = 1 + Kn\left[\alpha + \beta \exp\left(\frac{-\gamma}{Kn}\right)\right];\ \alpha = 1.142,\ \beta = 0.558,\ \gamma = 0.999 \qquad (1)$$

Where the Knudsen number *Kn* is the ratio of the mean free path of the gas molecules to the radius of the sphere. The constants in equation (1) apply to Knudsen numbers from 0.03 to 7.2, and were measured using polystyrene latex-divinylbenzene, polyvinyltoluene, and polystyrene latex particles. For a constant temperature and pressure experiment, *Cc* is a function of particle diameter only. Stokes drag, corrected for transitional effects, is now

$$F_{Stokes} = 3\pi\mu V \frac{Dp}{Cc(Dp)} \qquad (2)$$

DRAG MEASUREMENTS BY ELECTROSTATIC CLASSIFICATION

The TSI 3080 Electrostatic Classifier with 3081 Long Differential Mobility Analyzer (DMA) allows the calculation of the particle size exiting the DMA along with the drag on a submicrometer aerosol particle. This is accomplished by charging the particles with a Kr-85 aerosol neutralizer and making measurements of the particle's electronic mobility, that is, the ability of a charged particle to move in an electrostatic field when opposed by aerodynamic drag. See reference 2 for a detailed discussion of the DMA. In the DMA the charged particles are subject to an electrostatic field and quickly reach terminal velocity, resulting in the force balance

$$F_{Electric} = F_{Aerodynamic}$$

$$neE = ne\frac{\overline{V}}{r_2 - r_1} = 3\pi\mu V \frac{Dp}{Cc(Dp)}\ ;\ E = \frac{\overline{V}}{r_2 - r_1} \qquad (3)$$

Where *Dp* is the particle diameter, *n* is the number of charges on the particle, *e* is the elementary unit of electronic charge, $\overline{V}$ is the applied voltage, and r_1 and r_2 are the inner and outer radii of the DMA. The difference of the two radii is the distance that the voltage is applied across. This is also the distance that the particles must traverse in the direction of the electrostatic field. The electrical mobility *Zp* is defined as the ratio of the particle velocity to the electronic potential:

$$Zp = \frac{V}{E} = \frac{ne}{3\pi\mu} \cdot \frac{Cc(Dp)}{Dp} \qquad (4)$$

Knutsen and Whitby[2] determined the electrical mobility of the DMA using geometry, flow rate, and voltage as parameters. The equation is shown below:

$$Zp = \frac{Q_{sh}}{2\pi\overline{V}L}\ln(r_2/r_1) \qquad (5)$$

Where Q_{sh} is the sheath volumetric flow rate and L is a geometric parameter of the DMA. Equating the electrical mobilities in equations (4) and (5), the particle diameter can be determined by the fluid properties, the charge on an electron, DMA geometry, applied voltage, and sheath flowrate:

$$\frac{Dp}{Cc(Dp)} = \left[\frac{2e}{3\mu}\cdot\frac{L}{\ln(r_2/r_1)}\right]\cdot\frac{\overline{V}}{Q_{sh}}\cdot n \qquad (6)$$

For a constant temperature experiment, the terms in brackets are constant. See the figure below for a schematic of the 3081 Long DMA.

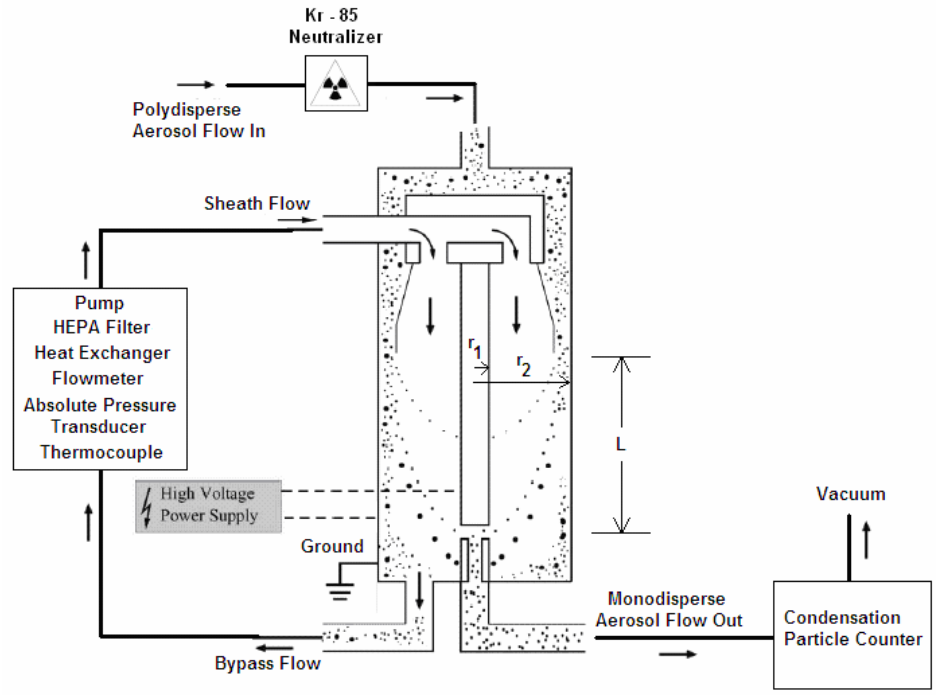

Figure 1: Schematic of particle size spectrometer system consisting of TSI 3081 Long DMA/TSI 3080 Electrostatic Classifier (TSI 3080 manual) and CPC.

By keeping the sheath flow rate constant and varying the voltage across its range from 0 to 10kV, a range of particle sizes will exit the DMA, predicted by equation (6). At each voltage increment, the aerosol particle concentration (particles/volume) is measured. From this data a particle size spectrum can be measured.

In order to calculate the drag on a certain size aerosol particle, we determine the voltage corresponding to the nanoparticle from the measured particle size spectrum. From equation [3], for a particle with n charges, the drag force is

$$F_{drag} = F_{electric} = ne\frac{\overline{V}}{r_2 - r_1} \qquad (7)$$

We are also interested in the velocity V of the particle. From equation (4)

$$V = \frac{ne}{3\pi\mu} \cdot \frac{Cc}{Dp} \cdot \frac{\overline{V}}{(r_2 - r_1)} \tag{8}$$

Solving equation (6) for $\overline{V}$:

$$\overline{V} = \frac{3\mu \ln(r_2/r_1)}{2eL} \cdot \frac{Q_{sh}}{n} \cdot \frac{Dp}{Cc} \tag{9}$$

Substituting (9) into (8)

$$V = Q_{sh} \frac{\ln(r_2/r_1)}{2\pi L(r_2 - r_1)} \tag{10}$$

FREE MOLECULAR FLOW PAST A SPHERE

The free molecular flow drag equations are now presented in order to compare with the transitional drag calculations. The following equations are derived from the kinetic theory by integrating around a sphere the force on an element of area[3]. Series expansions were employed to keep the function from diverging when S, the speed ratio, becomes very small. Assuming fully diffuse reflection and equivalent temperatures for incident and reflected molecules,

$$F_{Diffuse} \approx \frac{2\sqrt{\pi}}{3} SpA_{ref} \left(1 + \frac{3}{\sqrt{\pi}} \frac{erf(S)}{S}\right) \tag{11}$$

Where p is the absolute pressure, A_{ref} is the reference cross-sectional area of the sphere, and S is the speed ratio. Similarly, the sphere drag is calculated assuming specular reflection:

$$F_{Specular} \approx \frac{8}{3\sqrt{\pi}} SpA_{ref} \tag{12}$$

These equations are only valid when $S << 0$.

To calculate the drag using equations (11) and (12) the absolute pressure p and the molecular speed ratio S must be known. The absolute pressure is measured experimentally, while S is calculated using equation (10) and the definition of the speed ratio:

$$S = \frac{V}{c} = \frac{Q_{sh}}{2\pi} \frac{\ln(r_2/r_1)}{L(r_2 - r_1)} \cdot \frac{1}{\sqrt{2RT}} \tag{13}$$

Where R is the gas constant for air and T is the experimentally measured air temperature.

MEASUREMENT OF AERODYNAMIC DRAG

To measure the drag on aerosol nanoparticles, we generate an aerosol consisting of mainly one size particle (monodisperse aerosol). We then measure the particle size spectrum as a function of voltage to determine the voltage and diameter of the nanoparticle. The two experimental size spectrums are shown below:

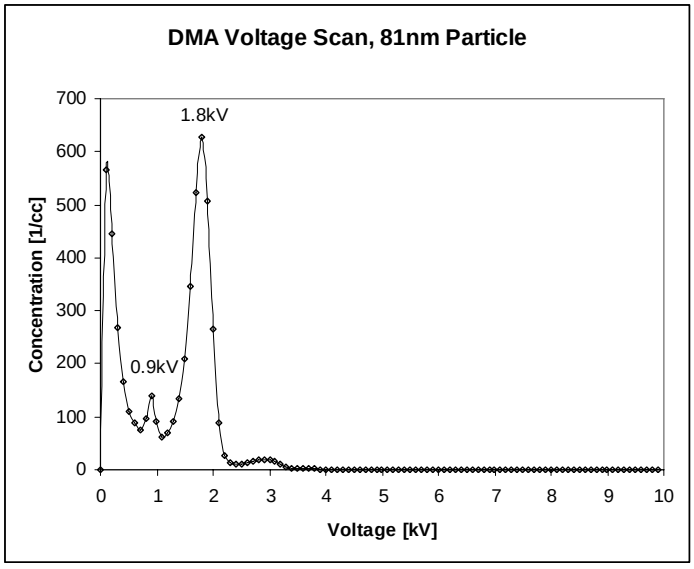

Figure 2: Size spectrum of 84.4nm aerosol.

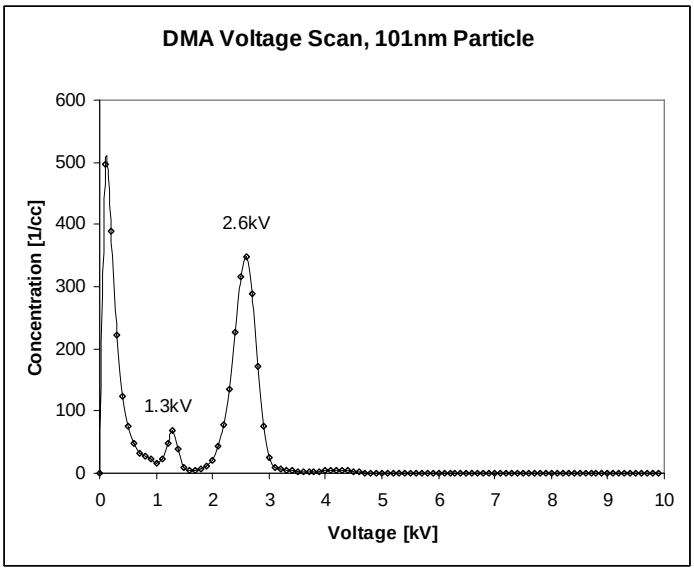

Figure 3: Size spectrum of 104nm aerosol.

The initial large peak consists of very small impurity particles resulting from the drying of water droplets containing impurities. The second smaller peak corresponds to the single solid particles containing a double positive charge. The third large peak corresponds to the single solid particles containing a single positive charge. Note that the voltages of these two peaks differ by a factor of 2, according to equation (6). Finally, the fourth, smallest peak indicates two solid spheres stuck together. This results from the drying of a water droplet containing two solid particles.

The diameter of the spherical particle is determined from equation (6), and the transitional drag on the particle is calculated from equation (7). The Knudsen number is estimated using the mean free path at the experimental temperature and pressure along with the radius of the particle. The speed ratio is calculated from equation (13). Finally, the aerodynamic drag on the particle by free molecular kinetic theory considerations for diffuse and specular boundary conditions is approximated by equations (11) and (12).

CONCLUSIONS

A table of experimental data is given below.

Gas Properties

R [J/kg K]	T [K]	μ [kg/m s]	p [Pa]	m.f.p [nm]
287	294	1.80E-05	96500	67

Particle Properties

		Drag Force [Newtons]		
V [Volts]	Dp [nm]	Transitional	Diffuse	Specular
1800	84.4	2.82E-14	2.92E-14	1.28E-14
2600	104	4.07E-14	4.44E-14	1.94E-14

The sheath flow rate was set at 15Lpm, yielding a speed ratio of 1.57e-5. In each case the free molecular estimate for drag using diffuse reflection overestimates the experimentally measured transitional drag. Furthermore, the experiment is conducted with Knudsen numbers of 1.29 and 1.59, well into the transitional regime. In nature, the aerodynamic drag on a body is larger in the transitional regime than in the free molecular regime. This further implies that the accommodation coefficients for 104nm and 84.4nm PSL are likely less than unity. The analysis of DMA data should take into account the effect of surface accommodation that is generally unknown for most materials of interest.

REFERENCES

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2. Knutson E.O., and K.T. Whitby [1975] “Aerosol Classification by Electric Mobility: Apparatus Theory and Applications.” *Journal of Aerosol Science*, 6:443.
3. Sentman, Lee. “Free Molecular Flow Theory and its Application to the Determination of Aerodynamic Forces.” Lockheed Missiles and Space Company, Armed Services Technical Information Agency, Arlington Hall Station, Arlington, Virginia, U.S.A. 01 October 1961.