Sizing Aerosol Particles between One and Three Nanometers

Thesis By

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Remember Love, and nothing else will matter. — Anon.

You cannot teach a man anything; you can only help him find it within himself.
— Galileo Galilei

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Abstract

The measurement of nanoparticle size is of primary importance to the fields of aerosol science and nanotechnology. Size affects the aerosol dynamics and impacts the optical, magnetic, and catalytic properties of nanoparticles. The size range from 1 to 10 nm is of particular interest because quantum effects and ambient aerosol nucleation occur in this range. Differential Mobility Analyzers are the primary instruments used currently to measure the size of aerosol particles, but diffusion impairs significantly their ability to size in the lower range (1–3 nm).

A primary focus of this thesis work has been to design, construct, and test a Radial Differential Mobility Analyzer, termed “nano-RDMA,” capable of measuring nanoparticles in the 1 to 12.5 nm size range with high resolution and transmission. The nano-RDMA was calibrated using electrospray techniques to aerosolize molecular ions for mobility analysis. The instrument was determined to have significantly improved resolution and transmission than the commercially available nano-DMA. Simulations of the nano-RDMA operation were performed to determine how the resolution could be improved.

The nano-RDMA has been employed to characterize *in-situ* size distributions of particles produced in an atmospheric pressure microplasma reactor. The operating parameters of the microplasma (i.e., plasma current, flow rate, precursor concentration, and precursor composition) were investigated to determine the effect on particle size distribution. The microplasma was further examined as a calibration source for narrow size distributions. Iron nanoparticles produced with the microplasma were used to grow nanotubes catalytically. A correlation was established between the size of the grown carbon nanotubes and the catalytic particles produced in the microplasma.

The nano-RDMA was also used as a size characterization and selection device in particle overgrowth experiments. Silicon nanoparticles produced in the microplasma were introduced into two different processing stages: a second microplasma and a furnace. The nano-RDMA permitted quick evaluation of how the operating conditions of the second processing stage affected overgrowth. The broad size distributions indicated that agglomeration was contributing to the measured size distribution, leading to the adoption of a Tandem DMA (TDMA) arrangement for overgrowth experiments. Limiting the size distribution with the first nano-RDMA permitted homogeneous overgrowth of silicon nanoparticles.

The microplasma was also investigated as a possible calibration source using the TDMA arrangement. While the microplasma produced a stable size distribution with a high concentration of nanoparticles, the measured resolution was lower than expected. The TDMA arrangement permitted studies of the thermal annealing of silicon nanoparticles. The particle size was observed to decrease with increasing temperature in a manner consistent with hydrogen evolution.

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b	Electrode gap spacing (mm)
E	Electric field ($V\ m^{-1}$)
k	Boltzmann constant ($1.38 * 10^{-23}\ J\ K^{-1}$)
m	Mass of background gas molecule (kg)
P	Pressure (pascal)
q	Elementary unit charge ($1.602 * 10^{-19}$ coulomb)
Q_a	Aerosol inlet flow rate (LPM, SLM or sccm)
Q_e	Excess outlet flow rate (LPM, SLM or sccm)
Q_s	Sample outlet flow rate (LPM, SLM or sccm)
Q_{sh}	Sheath inlet flow rate (LPM, SLM or sccm)
R_i	Inner radius of electrode (mm)
R_o	Outer radius of electrode; aerosol inlet radius (mm)
T	Temperature (K)
v_m	Migration velocity ($cm\ s^{-1}$)
Z_P	Electrophoretic mobility ($cm^2\ V^{-1}\ s^{-1}$)
Z_P^*	Ideal electrophoretic mobility ($cm^2\ V^{-1}\ s^{-1}$)
δ	Mobility shift (Z_P / Z_P^*)
η	Transmission