

Sizing Aerosol Particles between One and Three Nanometers

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Acknowledgements

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.

Table of Contents

List of Figures	xiv
List of Tables	xviii
List of Nomenclature	xix
Chapter 1: Introduction	1
1.1. Introduction and Background	1
1.2. Plasma Background	3
1.3. Aerosol Mobility Measurements Background	4
1.3.1. Particle Size	5
1.3.2. Particle Concentration	7
1.3.3. Log-Normal Distribution	8
1.3.4. Agglomeration	8
1.4. Dissertation Outline	10
References	12
Chapter 2: Radial Differential Mobility Analyzer for One Nanometer Particle Classification	14
2.1. Introduction	14
2.2. Experimental Method	17
2.2.1. Instrument Design	17
2.2.2. Instrument Calibration	20
2.2.3. Instrument Coupling with Mass Spectrometer (in Collaboration)	22
2.2.4. Instrument Comparison to Other DMAs (in Collaboration)	23
2.3. Results	25

2.3.1. Calibration.....	25
2.3.2. Results: Mass Spectrometry.....	26
2.3.3. Results: DMA Comparison.....	27
2.4. Discussion.....	28
2.4.1. Calibration.....	28
2.4.2. Mass Spectrometry.....	31
2.4.3. DMA Comparison.....	32
2.5. Summary.....	32
References.....	48
Chapter 3: Finite Element Analysis of the nano-RDMA Geometry.....	50
3.1. Introduction.....	50
3.2. Theoretical Considerations.....	54
3.3. Results and Discussion.....	57
3.3.1. Electrostatics.....	57
3.3.2. Incompressible Navier-Stokes.....	58
3.3.3. Electrokinetic Flow.....	58
3.3.4. Calibration Factor.....	59
3.3.5. Resolution.....	60
3.4. Summary.....	63
References.....	79
Chapter 4: Microhollow Cathode Discharge Operating Conditions Impact on Nanoparticle Size Production.....	81
4.1. Introduction.....	81

4.2. Experimental Method.....	82
4.3. Results and Discussion	84
4.4. Summary	86
References.....	95
Chapter 5: Nanoparticle Production from Cathode Sputtering in High-Pressure Microhollow Cathode and Arc Discharges.....	96
5.1. Introduction.....	96
5.2. Experimental Method.....	97
5.3. Results and Discussion	98
5.4. Summary	101
References.....	105
Chapter 6: Microdischarge Synthesis of Fe Nanoparticles for Diameter-Controlled Growth of Carbon Nanotubes	106
6.1. Introduction.....	106
6.2. Experimental Method.....	107
6.3. Results and Discussion	110
6.4. Summary	113
References.....	120
Chapter 7: Instrument Calibration Using Tandem Differential Mobility Analysis with a Microplasma Source	122
7.1. Introduction.....	122
7.2. Experimental Method.....	124
7.3. Results and Discussion	127

7.4. Summary	132
References.....	141
Chapter 8: Size Evolution of Annealed Silicon Nanoparticles.....	143
8.1. Introduction.....	143
8.2. Experimental Description	145
8.3. Results and Discussion	146
8.4. Summary	150
References.....	157
Chapter 9: Particle Size and Surface Modification of Aerosol Silicon Nanoparticles ...	158
9.1. Introduction.....	158
9.2. Experimental Method.....	160
9.2.1. Dual Microplasma Setup.....	160
9.2.2. Overgrowth	162
9.2.3. Size-Selected Overgrowth	163
9.3. Results and Discussion	164
9.3.1. Dual Microplasma.....	164
9.3.2. CVD Overgrowth.....	167
9.3.3 Size-Selected Overgrowth	168
9.4. Summary	169
References.....	181
Chapter 10: Future Work	182
10.0. Introduction.....	182
10.1. Improved Electrospray Apparatus	182

10.2. Conductivity of Nanoscale Solid Acid Fuel Cell.....	183
10.3. Tandem Differential Mobility Analyzer Growth of Carbon Nanotubes.....	185
10.4. Sensitive Faraday Cup Electrometer.....	186
10.5. Nanoparticle Calibration Source for Differential Mobility Analyzers	187
10.6. Nanoparticle Synthesis Using a Water Plasma Electrospray.....	188
References.....	192
Appendix I: Electrospray Sources	193
A1.1 Introduction.....	193
A1.2 Electrospray Apparatus: Detailed Assembly	194
A1.2.1 The Liquid Delivery System.....	194
A1.2.2 Electrospray for Volatilization of Molecular Ions	195
A1.2.3 Electrospray for Nano-structured Electrodes.....	197
Appendix II: Composite Nano-structured Solid Acid Fuel Cell Electrodes via Electrospray Deposition.....	202
A2.1. Introduction.....	202
A2.2. Experimental Method.....	204
A2.2.1. Deposition	204
A2.2.2. Solution Properties.....	205
A2.2.3. Oxygen Plasma Treatment.....	206
A2.2.4. AC Impedance Measurements	206
A2.3. Results and Discussion.....	207
A2.4. Summary	209
References.....	222

Appendix III: A Tool for Uniform Coating of 300 mm Wafers with Nanoparticles.....	223
A3.1. Introduction.....	223
A3.2. Design	227
A3.3. Experimental Apparatus and Methods.....	233
A3.4. Results.....	235
A3.5. Conclusions.....	237
References.....	255

List of Figures

Figure 2.1. Simplified Schematic of RDMA Operation.	34
Figure 2.2. Detailed Schematic of the nano-RDMA.....	35
Figure 2.3. Schematic of Electrospray.....	36
Figure 2.4. Schematic of nano-RDMA and Mass Spectrometer.	37
Figure 2.5. Schematic of TDMA.	38
Figure 2.6. Inverse Mobility Distributions.	39
Figure 2.7. Geometric Mean Mobilities.....	40
Figure 2.8. Inverse Mobility Distributions.	41
Figure 2.9. Inverse Mobility Distributions Using Mass Spectrometer as Detector.	42
Figure 2.10. Mobility Distribution of Monomer and Dimer Using MS.	43
Figure 2.11. Mobility Distribution Comparison between nano-RDMA and nano-DMA.	44
Figure 2.12. Mobility Distribution of TMAI.	45
Figure 2.13. nano-RDMA Resolution.....	46
Figure 3.1. Outline of the Axis-Symmetric Model.	64
Figure 3.2. Electrostatics Solution.....	65
Figure 3.3. Inset of Aerosol Inlet Region.	66
Figure 3.4. Electrostatics Solution without Mesh.....	67
Figure 3.5. Navier-Stokes Solution.....	68
Figure 3.6. Electrokinetic Flow Solution.....	69
Figure 3.7. Electrokinetic Flow Solution without Mesh.....	70
Figure 3.8. Calibration Factor for Electrode Spacing.....	71
Figure 3.9. Calibration Factor for Aerosol Inlet Gap.....	72
Figure 3.10. Resolution of nano-RDMA.	73
Figure 3.11. Resolution for Different Electrode Spacings.....	74
Figure 3.12. Resolution for Different Aerosol Outlet Gaps.....	75
Figure 3.13. Resolution for Different Aerosol Inlet Gaps.	76
Figure 3.14. Electrokinetic Flow Solution at Optimal Voltage.	77
Figure 3.15. Resolution Comparison Between the nano-RDMA and RDMA.	78
Figure 4.1. Schematic of Microplasma.	88

Figure 4.2. Schematic of Microplasma Deposition.	89
Figure 4.3. Size Distribution for Different Plasma Currents.	90
Figure 4.4. Size Distribution for Different Silane Concentrations.	91
Figure 4.5. Size Distribution for Different Plasma Flow Rates.	92
Figure 4.6. Size Distribution of Germanium Nanoparticles.	93
Figure 4.7. EDS of Germanium Nanoparticles.	94
Figure 5.1. Schematic of Sputtering Discharges.	102
Figure 5.2. Size Distributions of Positive Charged Particles.	103
Figure 5.3. Distribution of Negative Charged Particles.	104
Figure 6.1 Schematic of Microplasma for Fe Nanoparticles.	114
Figure 6.2. Size Distributions of Iron Nanoparticles.	115
Figure 6.3. AFM Images of CNTs.	116
Figure 6.4. Size Distribution of Nanotube Diameters.	117
Figure 6.5. Size Variation of Nanotubes and Nanoparticles.	118
Figure 7.1 Schematic of Tandem DMA.	133
Figure 7.2. Mobility Distribution from First nano-RDMA.	134
Figure 7.3. TDMA Mobility Distributions.	135
Figure 7.4. Normalized TDMA Mobility Distributions.	136
Figure 7.5. Shift of TDMA Mobility Distribution.	137
Figure 7.6. Transmission of the nano-RDMA.	138
Figure 7.7. Resolution Measured with the Tandem DMA.	139
Figure 8.1. Schematic of Tandem DMA Sintering Arrangement.	151
Figure 8.2. Size Distribution of First and Second nano-RDMA.	152
Figure 8.3. Size Variation of 3.1 nm Silicon Nanoparticles with Temperature.	153
Figure 8.4. Size Variation of 2.7 nm Silicon Nanoparticles with Temperature.	154
Figure 8.5. Size Variation of 1.7 and 1.3 nm Silicon Nanoparticles with Temperature.	155
Figure 8.6. Comparison of Size Variation with Temperature.	156
Figure 9.1. Schematic of Dual Microplasma.	170
Figure 9.2. Schematic of the Overgrowth Arrangement.	171
Figure 9.3. Schematic of the Tandem DMA Overgrowth Arrangement.	172
Figure 9.4. Size Distribution of the Dual Microplasma.	173

Figure 9.5. Size Distribution of Dual Microplasma with Additional Silane.	174
Figure 9.6. Size Distribution Produced with Dual Microplasma Varying First Gap.....	175
Figure 9.7. Size Distribution Produced with Dual Microplasma Varying Second Gap.	176
Figure 9.8. Size Distribution with Overgrowth with Diffusion Mixing.	177
Figure 9.9. Size Distribution with Overgrowth with Jet Mixing.	178
Figure 9.10. Size Distribution without Overgrowth from Single Microplasma.	179
Figure 9.11. Size Distribution with Overgrowth in the Tandem DMA Arrangement.	180
Figure 10.1. Electrospray Plasma	191
Figure A1.1. Schematic of the Electrospray Source.	199
Figure A1.2. Electrospray.	200
Figure A1.3. Schematic of Electrospray.	201
Figure A2.1. Fuel Cell Electrode Particle Size Comparison.	211
Figure A2.2. Schematic of the Electrospray.	212
Figure A2.3. Fuel Cell Assembly.	214
Figure A2.4. Nano-structured CDP.	215
Figure A2.5. X-ray Diffraction of CDP.	216
Figure A2.6. Dense CsH_2PO_4 Film.	217
Figure A2.7. Effect of Aging on Nano-structure.	218
Figure A2.8. Stabilized Electrode Nano-structure.	219
Figure A2.9. FTIR of Electrosprayed Electrode.	220
Figure A2.10. Impedance Plot	221
Figure A3.1. Schematic of Thermophoretic Depositor with Isotherms.	239
Figure A3.2. Modeled Temperature Profile.	240
Figure A3.3. Modeled Velocity Contours for Flow Rates of 1.5 and 15 SLM.	241
Figure A3.4. Modeled Velocity Contours for Flow Rates of 35 and 60 SLM.	242
Figure A3.5. Modeled Temperature Contours.	243
Figure A3.6. Modeled Stream Function Plots.	244
Figure A3.7. Modeled Deposition Profile.	245
Figure A3.8. Modeled Deposition Profile.	246
Figure A3.9. Modeled Deposition Profile.	247
Figure A3.10. Modeled Deposition Profile.	248

Figure A3.11. Plot of Deposition Profile.....	249
Figure A3.12. Particle Size Distribution and Wafer Coverage.....	250
Figure A3.13. AFM Images of Deposited Particles.....	251
Figure A3.14. Modeled Inlet Temperature Distribution without Inlet Heating.....	252
Figure A3.15. Deposition Profile without Inlet Heating.....	253
Figure A3.16. Particle Deposition without Inlet Heating.	254

List of Tables

Table 2.1. Mobility Data.....	47
Table 6.1 Measured Particle Size.....	119
Table 7.1. Fitting Parameters of First nano-RDMA Mobility Distributions	140
Table A2.1. Critical Electrospray Parameters.....	213

List of Nomenclature

b	Electrode gap spacing (mm)
E	Electric field (V m^{-1})
k	Boltzmann constant ($1.38 * 10^{-23} \text{ J K}^{-1}$)
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 ($\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)
Z_P^*	Ideal electrophoretic mobility ($\text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)
δ	Mobility shift (Z_P / Z_P^*)
η	Transmission