Nanomaterial Distribution Analysis via Liquid Nebulization Coupled with Ion Mobility Spectrometry (LN-IMS)

Seongho Jeon[1], Derek R. Oberreit[2], Gary Van Schooneveld[3], & Christopher J. Hogan Jr.[1,*]

[1]Department of Mechanical Engineering, University of Minnesota, Minneapolis, MN, USA
[2]Fluid Measurement Technologies, St Paul, MN USA
[3]CT Associates, Inc., Eden Prairie, MN USA

Supplemental Information

***To whom correspondence should be addressed: hogan108@ umn.edu, Tel: 1-612-626-8312, Fax: 1-612-625-6069.**

INFORMATION AVAILABLE

- Schematic diagrams of the liquid nebulizer and the LN-IMS system.
- A description of and results with the residue method of droplet size distribution function measurement.
- Size distribution functions for bovine serum albumin and ovalbumin.

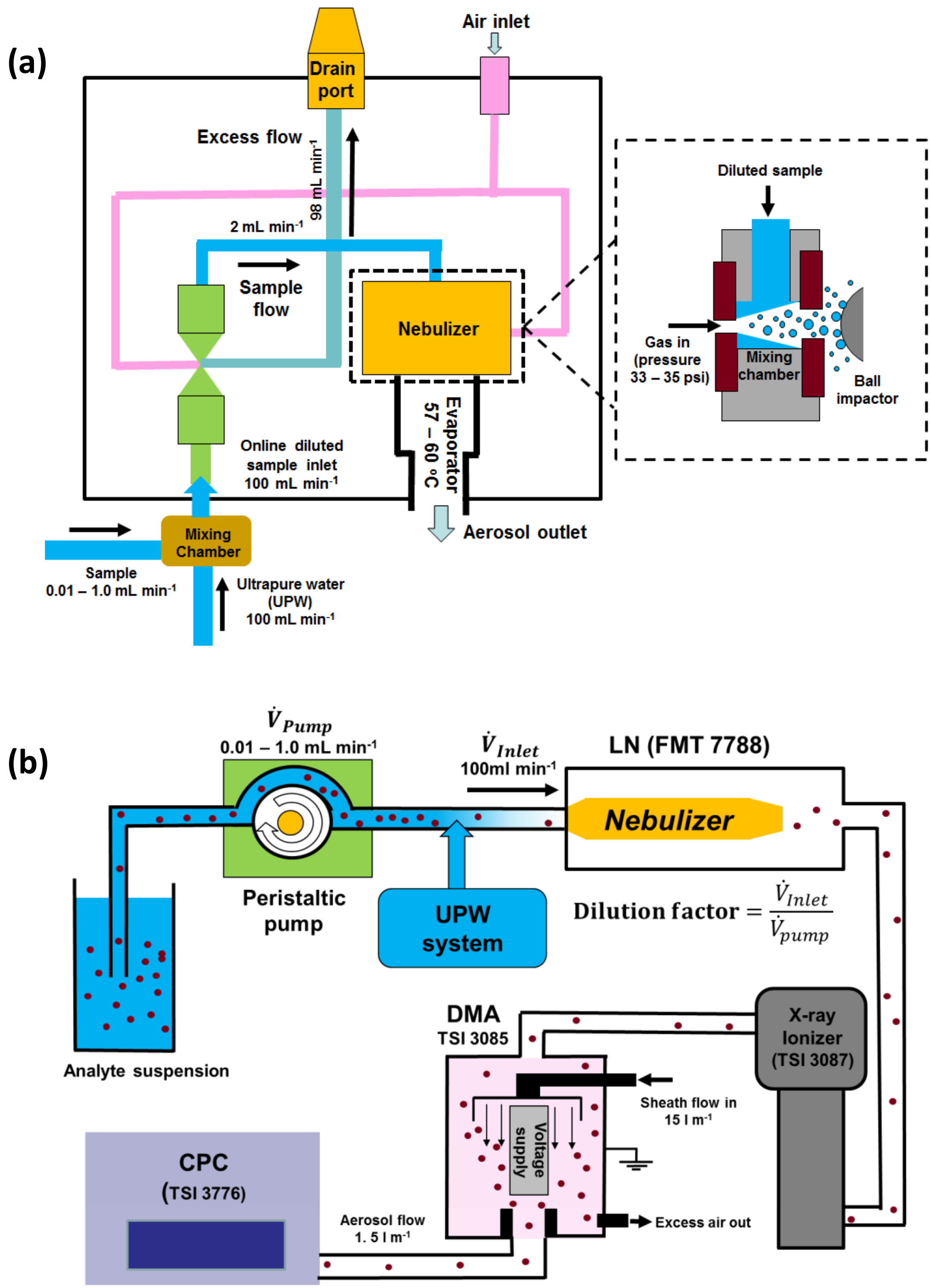

Figure S1. **(a.)** Schematic diagram of the liquid nebulizer (LN). **(b.)** Schematic diagram of the LN coupled with a differential mobility analyzer (DMA) and condensation particle counter (CPC) for IMS measurements.

Droplet Size Distribution Functions

Most pneumatic nebulizers create droplet size distribution functions which have mode diameters between 2 and 5 μm; such droplets are not well suited for aerosolization of nanoparticles without distortion of the particle size distribution function. To measure the droplet diameter, we applied the residue method,[1] in which we mixed sucrose with UPW at an original sucrose volume fraction (V_f) of 0.0654. This high concentration solution was then nebulized with DFs of 17.39 and 42.11, and after solvent evaporation, the size distribution functions of the residual sucrose particles were measured with the DMA-CPC. Droplet diameters (d_D) were linked to the measured sucrose particle diameters (d_S) through the relationship: $d_D = (\frac{DF}{V_f})^{1/3} d_S$. Inferred droplet size distribution functions are plotted in Figure S2. The good agreement between measurements with different DFs suggests that solutions are well mixed after dilution and prior to nebulization, and further suggests that the residue method enabled reliable estimation of the droplet size distribution function. The droplet size distribution function is found to be approximately lognormal, with a geometric mean diameter of 99.8 nm and a geometric standard deviation of 2.32. This geometric mean diameter is similar to many charge reduction electrospray sources[2]; however, the geometric standard deviation is much larger than cone-jet mode electrosprays (which are typically closer to 1.1).[1] Nonetheless, more than 95% of the droplets generated and exiting the nebulizer (i.e. transmitted around the impactor) are smaller in diameter than 350 nm, suggesting that the LN examined here will minimally distort nanoparticle size distributions during aerosolization.

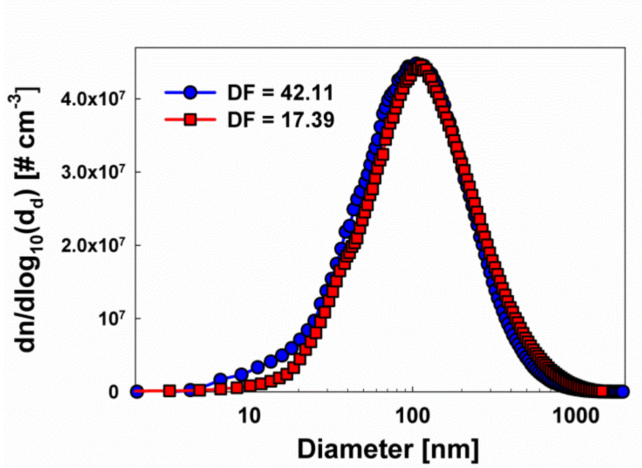

Figure S2. LN generated droplet size distribution functions, inferred using the residue method.

Table S1. The LN-IMS experiment conditions for 5, 7, and 10 nm gold nanospheres.

<table>
<tr><th>Material</th><th>Suspension Properties</th><th>particle # concentration (# mL^{-1})</th><th>Dilution factor</th></tr>
<tr><td rowspan="9">5 nm gold nanosphere</td><td rowspan="9">DI Water</td><td rowspan="2">4.80×10^{13}</td><td>2000</td></tr>
<tr><td>5000</td></tr>
<tr><td rowspan="2">8.00×10^{12} (mixture)</td><td>500</td></tr>
<tr><td>1000</td></tr>
<tr><td rowspan="3">4.80×10^{12}</td><td>500</td></tr>
<tr><td>1000</td></tr>
<tr><td>2000</td></tr>
<tr><td>4.80×10^{11}</td><td>500</td></tr>
<tr><td>4.80×10^{10}</td><td>200</td></tr>
<tr><td rowspan="16">7 nm gold nanosphere</td><td rowspan="16">DI Water</td><td rowspan="2">1.70×10^{13}</td><td>1000</td></tr>
<tr><td>2000</td></tr>
<tr><td>2.83×10^{12} (mixture)</td><td>500</td></tr>
<tr><td rowspan="2">2.83×10^{12} (mixture)</td><td>250</td></tr>
<tr><td>500</td></tr>
<tr><td>2.83×10^{12} (mixture)</td><td>250</td></tr>
<tr><td>8.50×10^{12}</td><td>2000</td></tr>
<tr><td rowspan="2">1.70×10^{12}</td><td>500</td></tr>
<tr><td>1000</td></tr>
<tr><td rowspan="2">8.50×10^{11}</td><td>500</td></tr>
<tr><td>1000</td></tr>
<tr><td rowspan="2">1.70×10^{11}</td><td>500</td></tr>
<tr><td>1000</td></tr>
<tr><td>8.50×10^{11}</td><td>200</td></tr>
<tr><td>8.50×10^{9}</td><td>125</td></tr>
<tr style="display:none"><td></td><td></td></tr>
<tr><td rowspan="13">10 nm gold nanosphere</td><td rowspan="9">DI Water</td><td rowspan="2">3.40×10^{12}</td><td>2000</td></tr>
<tr><td>1000</td></tr>
<tr><td rowspan="2">2.83×10^{12} (mixture)</td><td>500</td></tr>
<tr><td>1000</td></tr>
<tr><td>2.83×10^{12} (mixture)</td><td>500</td></tr>
<tr><td>3.40×10^{11}</td><td>1000</td></tr>
<tr><td rowspan="2">3.40×10^{10}</td><td>200</td></tr>
<tr><td>500</td></tr>
<tr><td>3.40×10^{9}</td><td>125</td></tr>
<tr><td>pH 6.0</td><td>3.04×10^{12}</td><td>2000</td></tr>
<tr><td>pH 9.7</td><td>3.04×10^{11}</td><td>500</td></tr>
<tr><td>PBS 0.01</td><td>3.04×10^{12}</td><td>2000</td></tr>
<tr><td>PBS 0.005</td><td>3.04×10^{12}</td><td>1000</td></tr>
</table>

Table S2. The LN-IMS experiment conditions of 15 and 30 nm gold nanospheres and the GNRs.

Materials	**Suspension Properties**	**particle # concentration (# mL^{-1})**	**Dilution factor**
15 nm gold nanosphere	DI Water	1.10×10^{12}	1000
		9.17×10^{11} (mixture)	250
			500
		1.83×10^{11} (mixture)	250
			500
		1.10×10^{11}	500
		1.10×10^{10}	125
	pH 6.0	1.00×10^{12}	2000
	pH 9.7	1.10×10^{11}	500
	PBS 0.005	9.90×10^{11}	1000
30 nm gold nanosphere	DI Water	2.00×10^{12}	125
			200
		1.67×10^{11} (mixture)	250
			500
		1.67×10^{11} (mixture)	250
		1.67×10^{11} (mixture)	250
			500
		2.00×10^{10}	200
			500
		2.00×10^{9}	125
Gold nanorod	DI Water	7.45×10^{10}	125
			200

Table S3. The LN-IMS experiment conditions of bovine serum albumin (BSA) and the ovalbumin (OVA).

Materials	Suspension Properties	particle # concentration (# mL^{-1})	Dilution factor
BSA	DI Water	4.53×10^{14}	10000
		2.26×10^{14}	10000
		4.53×10^{13}	2500
		1.13×10^{14}	5000
		8.48×10^{13}	2500
			5000
		5.65×10^{13}	2500
		2.26×10^{13}	2000
		2.26×10^{12}	1000
		2.26×10^{11}	250
		2.26×10^{10}	125
	PBS 0.01	2.00×10^{14}	10000
	PBS 0.05	4.04×10^{14}	10000
	pH 5.0	2.26×10^{14}	10000
	pH 9.7	2.26×10^{14}	10000
OVA	DI Water	6.12×10^{14}	10000
		6.12×10^{13}	1000
			2000
			2500
	PBS 0.01	5.54×10^{14}	10000
	pH 9.7	8.16×10^{14}	10000

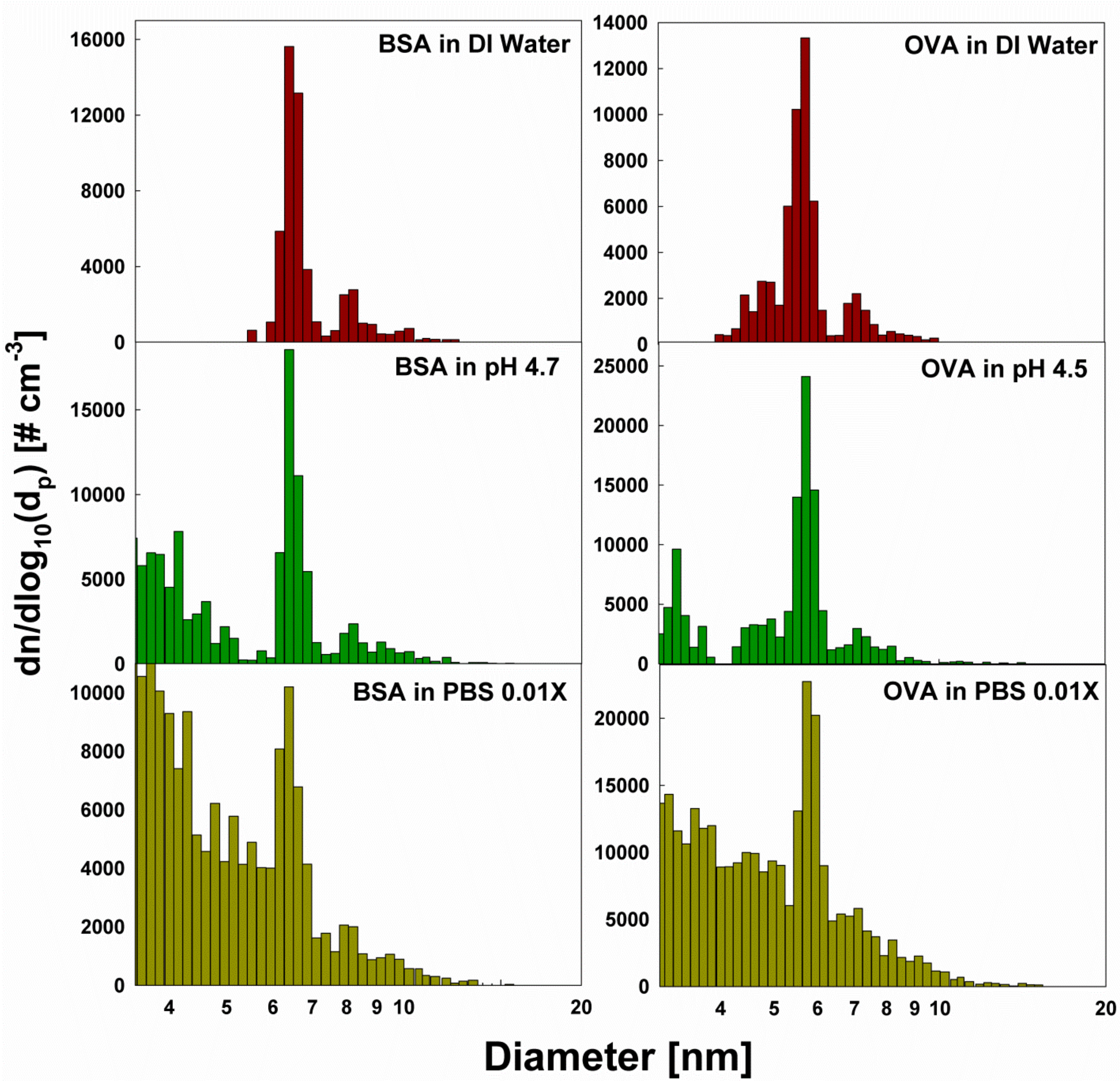

Figure S3. Size distribution functions of bovine serum albumin (BSA) and ovalbumin (OVA) in various suspensions. As with gold nanospheres, peaks below 5 nm in diameter correspond to non-volatile residue formed during the aerosolization process. Further details on the formation of residue particles is provided in the main text.

REFERENCES

1. D. R. Chen, D. Y. H. Pui and S. L. Kaufman, *J Aerosol Sci*, 1995, 26, 963-977.
2. S. L. Kaufman, *J Aerosol Sci*, 1998, 29, 537-552.