COMPARISON OF LIGHT SCATTERING AND MOBILITY SIZING TECHNIQUES FOR CHARACTERIZATION OF AGGREGATED PARTICLES

Yingwu Teng*, Umit O. Koylu*, and Fatih Dogan**
*Department of Mechanical and Aerospace Engineering
**Department of Materials Science and Engineering
University of Missouri-Rolla, Rolla, MO 65409, USA

The formation of carbonaceous particulates during the burning of fossil-based fuels significantly enhances the radiation heat transfer in internal combustion engines, natural fires, and industrial furnaces. When emitted to the atmosphere from various combustion sources (e.g. coal power plants and transportation vehicles), these aerosols modify the atmospheric radiation balance by scattering and absorbing solar light and therefore accelerate global climate change. Such sub-micron particles have long settling times, react with other atmospheric constituents, and increase melting rate of Arctic ice. These environmental concerns together with the negative effects on human health need to be assessed by accurately characterizing, monitoring, regulating and modeling particulates.

Because size is an important parameter to describe interactions of particulates with their surroundings (flame or atmosphere) through optical, chemical, transport, toxic, and deposition properties, there is a shift in measurement emphasis from mass to size (or concentration) basis [1]. Mobility particle sizing (MPS) is a widely employed technique to measure particulate matter due to commercial availability, real-time monitoring, and convenient control/data acquisition. Nevertheless, there are major ambiguities regarding the performance of these instruments for the characterization of non-spherical aggregated nano-particles. Primarily, the mobility data analysis assumes a spherical particle shape, which does not represent the common non-spherical morphology of fractal-like clusters (aggregates) formed by small spherical primary particles (spherules). Furthermore, the general sampling procedure used to remove the particles from their natural environment and transport them through these devices is extremely intrusive with lengthy lines and various pretreatment processes, which seem to potentially alter size and concentration distributions. Consequently, the correspondences between the mobility diameter measurements and the actual physical dimensions of various types of aggregates have been investigated in the past. In particular, the mobility diameters of flame-synthesized oxide [2] and diesel soot aggregates [3] were suggested to be proportional to their projected areas on transmission electron microscope (TEM) images. In contrast, recent studies on mainly diesel engine soot aggregates questioned not only this suggested relationship between the mobility and microscopy sizes [4] but also the ability of MPS to measure particle concentrations against the laser-induced incandescence technique.

In order to provide a reliable and unified database for atmospheric radiation balance, the present study extends the MPS and TEM measurements of Chandler et al. [4] at the exhaust of a diesel engine to soot aggregates above well-defined non-premixed laminar

flames. Such laboratory flames offer the flexibility to control and reproduce well-known sooting properties, unlike the diesel engines with transient and complex operating conditions. To help resolve the above-mentioned contradicting past results, laser scattering and extinction (LSE) technique is implemented as an independent diagnostic in addition to MPS and TEM measurements. LSE is a non-intrusive technique, unlike MPS and TEM, and the interpretation of LSE data is readily available for non-spherical aggregated particles [5], unlike MPS. Additionally, by furnishing soot concentrations in addition to sizes, the LSE method also yields particle surface areas that can be compared to those from MPS. Finally, polydisperse soot populations are considered so that particle sizing methods can directly be compared without any sample manipulations [2, 3].

A co-flow burner was used to support laminar non-premixed flames burning acetylene mixed with nitrogen and/or propane. Four different sooting conditions were generated above the flames: the first two cases were selected based on substantial size differences observed with the mobility instrument and the other two cases involved sooting conditions that were different according to the optical data.

Two mobility particle sizing (MPS) instruments were used in the present study. Performances of both devices were separately checked against known-size calibration latex spheres. Soot streams above the flames were sampled with a special microprobe that provided two-stage variable dilution rates. Particles first passed through a charger and then entered differential mobility analyzer (DMA) for classification according to their electrical mobility diameters. After exiting the DMA, particles were directed to a condensation particle counter (CPC) for measuring their concentrations.

A laser beam was focused at the same location of the MPS probe above the flame for the scattering and extinction (LSE) experiments. Photomultiplier tubes at 30 and 150 degrees collected the scattered light while a photodiode detector measured the transmitted light. The entire optical system was calibrated against latex spheres of known properties. By accounting for the aggregate fractal morphology in the data analysis, the measured ratio of scattering coefficient at 150 degrees to the extinction coefficient, dissymmetry ratio of scattering at 30 and 150 degrees, and light transmission data yielded the spherule diameter, aggregate radius of gyration, and soot volume fraction, respectively [5].

The first two experimental conditions (cases 1 and 2) were based on the distinct size distributions found from the MPS instrument as the mobility diameters for case 1 were much larger than those for case 2. In contrast, the raw optical measurements did not indicate a noticeable change from case 1 to case 2. Supplemental TEM measurements also supported this observation. Two new sooting conditions (cases 3 and 4) were therefore identified, this time, based on the optical measurements with substantially different scattering and extinction signals. While TEM measurements were also in agreement with the LSE data, the MPS measurements for conditions 3 and 4 displayed almost no change in the mobility diameter distributions. Mean values of spherule diameters, d_p, aggregate gyration diameters, d_g, mobility diameters, d_m, and specific particle surface areas, S_p, from the three independent diagnostics are conveniently summarized and compared in the table below.

Case	Method	d_p (nm)	d_g (nm)	d_m (nm)	S_p (m^2/m^3)
1	MPS	-	-	206	66
	LSE	44	414	-	1016
	TEM	30	316	-	
2	MPS	-	-	99	56
	LSE	36	380	-	1067
	TEM	27	326	-	
3	MPS	-	-	196	128
	LSE	33	465	-	982
	TEM	33	414	-	
4	MPS	-	-	200	115
	LSE	39	277	-	500
	TEM	33	211	-	

According to the mobility measurements, the soot size distributions were markedly different for the fist two experimental conditions, whereas the follow-up optical and microscopy measurements did not indicate any variations in either the spherule diameters or the aggregate sizes. For the next two experimental cases, both optical and microscopy measurements showed considerable variations in aggregate sizes in spite of almost the same mobility diameters. The results from the optical and microscopy techniques were consistent not only with each other but also with the expected soot properties in response to the variations in the flame conditions. The mobility measurements significantly underestimated the total specific particle surface areas compared the optically-inferred values due to erroneously combining the spherule and aggregate sizes in a single equivalent diameter. There was no evidence of a direct relationship between the mobility diameter and aggregate projected area that had been suggested in the past. These findings raised further questions about the performance of the mobility technique for determining the actual physical dimensions and surface areas of combustion-generated particulates.

Acknowledgments. University of Missouri Research Board and National Science Foundation sponsored this research.

REFERENCES

1. Kittelson,D. B., *J. Aerosol Sci.*, Vol. 29, pp 575-588, 1998.
2. Rogak, S. N., Flagan, R. C., and Nguyen, H. V., *Aerosol Sci. Technol.*, Vol. 18, pp 25-47, 1993.
3. Park, K., Kittelson, D. B., and McMurry, P. H., *Aerosol Sci. Technol.*, Vol. 38, pp 881-889, 2004.
4. Chandler, M. F., Teng, Y., and Koylu, U. O., *Proc. Combust. Inst.*, Vol. 31, pp 2971-2979, 2007.
5. Teng Y. and Koylu, U. O., *Appl. Opt.*, Vol. 45, pp 4396-4403, 2006.