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Measurement of combustion-generated nonvolatile nanoparticles

Gregory J. Smallwood*

ICPET, National Research Council Canada, 1200 Montreal Road, Ottawa, Ontario, K1W 1J7

Advances in optical diagnostics of nanoparticles have led to the recent emergence of capabilities to measure the concentration and size of nonvolatile aggregated nanoparticles emitted into the atmosphere during the production of energy through fossil fuel combustion. Advances and issues regarding the science of laser-induced incandescence (LII) for the measurement of the volume concentration of soot, the development of a high sensitivity LII (HS-LII) experimental apparatus, and the development of a combined LII and laser light scattering technique to determine the morphology of the soot nanoparticles are discussed.

1. Introduction

There have been major advances in the understanding of the physics and chemistry related to the nanoscale processes that occur as a result of the rapid heating and cooling of soot nanoparticles due to laser irradiation during the laser-induced incandescence (LII) process for measuring soot nanoparticles.

2. Laser-Induced Incandescence (LII)

The processes attracting the most attention are the heat and mass transfer processes that govern energy transfer in laser-induced incandescence. Recent research has highlighted the absorption function, $E(m)$, as the largest source of uncertainty in determining the soot volume fraction (SVF) and the accommodation coefficient, α , as a large source of uncertainty in determining the surface area and primary particle diameter. The use of coupled experimental and theoretical auto-compensating laser induced incandescence (AC-LII, employing two-wavelength pyrometry and low-fluence excitation [1]) approaches has led to improved determination of these properties. Further improvement in the determination of primary particle (the spherules that make up soot aggregates) diameter is achieved by accounting for the fractal aggregate nature of the soot nanoparticles.

3. High Sensitivity LII (HS-LII)

The development of a high sensitivity LII (HS-LII) experimental apparatus is discussed. The target was to improve the sensitivity of a laboratory AC-LII system by two orders of magnitude. The design considerations and implementation through a number of iterations of improvement are discussed. All aspects of the system were considered for optimization. The principle of the Lagrangian invariant was found to be most effective in designing the optical receiver subsystem. Application of the HS-LII instrument to measurements of soot concentration in the ambient atmosphere is presented, demonstrating a 500-fold improvement.

4. Combined LII and Light Scattering

For a complete morphological description of soot, the aggregate size (the number of primary particles making up the soot aggregate) is missing. Soot aggregate size distributions are described by a lognormal function whose distribution width is largely independent of the soot source and the (geometric) mean aggregate size. Given this relative insensitivity of the distribution width it is possible to determine mean aggregate size from a combination of absolute LII signals and near forward laser light scattering.

* Corresponding author: greg.smallwood@nrc.ca

A modified AC-LII system that allows simultaneous measurement with AC-LII and elastic light scattering employing Rayleigh-Debye-Gans Polydisperse Fractal Aggregate (RDG-PFA) theory [2] to interpret scattered laser light from the 532 nm excitation source is described. Thus a single measurement can result in estimates of soot volume fraction, the primary particle size and the geometric mean aggregate size. The application of this in situ technique to soot is described. The mean aggregate sizes derived in this way depend on a knowledge of the ratio of the soot absorption and scattering functions. The results obtained are compared favourably to those derived from the ratio of forward to backward scattering. This latter technique requires no knowledge of the absorption or scattering function values.

5. Future Research

Major new research projects for nanoparticle characterization have been recently announced in Canada. Research directions, including new scattering approaches and two-dimensional LII, will be discussed.

References

- [1] D. R. Snelling, G. J. Smallwood, F. Liu, Ö. L. Gülder, and W. D. Bachalo, *Appl. Opt.* 44 (2005) 6773-6785.
- [2] C. M. Sorensen, *Aer. Sci. Tech.* 35 (2001) 648-687.