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Optimization of measurement angles for soot aggregate sizing by elastic light scattering, through design-of-experiment theory

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ABSTRACT

In multiangle elastic light scattering (MAELS) experiments, the morphology of aerosolized particles is inferred by shining collimated radiation through the aerosol and then measuring the scattered light intensity over a set of angles. In the case of soot-laden-aerosols MAELS can, in principle, be used to recover the size distribution of soot aggregates, although this involves solving an ill-posed inverse problem. This paper presents a design-of-experiment methodology for identifying the set of angles that maximizes the information content of the angular scattering measurements, thereby minimizing the ill-posedness of the underlying inverse problem. While the optimized angles highlight the physical significance of the scattering regimes, they do not improve the accuracy of size distributions reconstructed from simulated experimental data.

1. Introduction

In most combustion processes pyrolized fuel molecules coalesce into nanospheres between 10 and 100 nm in diameter, called primary particles, which in turn agglomerate to form soot aggregates. Soot has long been a focus of combustion research, in large part because its formation within gas turbines and automotive engines is closely linked to the performance of these devices [1]. Accordingly, designing the next generation of clean and efficient combustion technology relies on improved soot models, which in turn are predicated on the availability of soot measurements carried out within these devices.

Soot also strongly impacts human health and the environment through mechanisms that depend strongly on aggregate size, which is often quantified by the number of primary particles per aggregate, N_p. For example, small soot particles penetrate more deeply into the lungs and can even cross the pleural membrane into the bloodstream [2]. Climatologists and atmospheric scientists have also assessed soot to be an extremely potent factor in climate change, second only to carbon dioxide [3]; soot deposited on glaciers increases their absorption of sunlight thereby hastening glacial melting [4], for example, and soot in the atmosphere promotes formation of clouds that shield the Earth from solar irradiation, which may drastically alter local climactic patterns [5]. Accordingly, instrumentation for measuring both the size and quantity of soot produced by a combustion device is crucial for assessing its impact on human health and the environment, and its compliance to emissions regulations.

While it is possible to infer the size distribution of aggregates within a soot-laden aerosol through electron microscopy of extracted soot aggregates, such as the image shown in Fig. 1, this process has a number of drawbacks. Perhaps foremost, characterizing the soot aggregate size distribution requires processing thousands of electron micrographs, an extremely time-intensive endeavor that effectively disallows analysis of transient processes. Also, inferring three-dimensional structural information from 2-D projections of soot aggregates induces certain biases into the results [6]. Finally,

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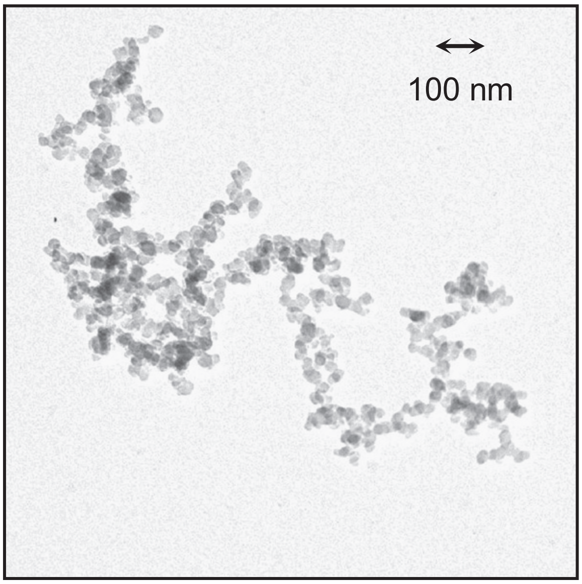

Fig. 1. Transition electron micrograph of a soot aggregate.

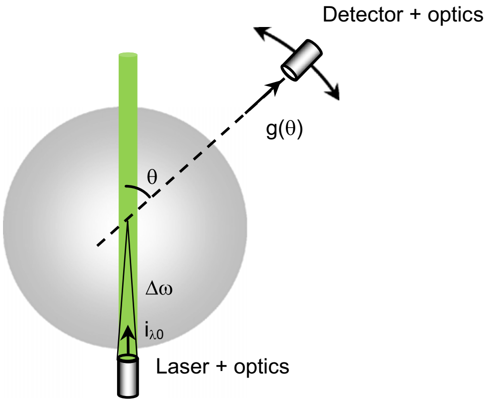

Fig. 2. Schematic of a MAELS experiment.

obtaining the physical access needed to probe the aerosol is often difficult (for example within a combustion chamber) and the probe itself may have a perturbing effect on the physical and chemical processes occurring in the aerosol.

Optical diagnostics overcome many of the above drawbacks. One such technique is multi-angle elastic light scattering (MAELS) (e.g. [7–11]), which is shown schematically in Fig. 2. In this experiment collimated light, usually from a laser, is shone through the aerosol, and the aggregate size distribution is then inferred from the angular distribution of scattered light. The angular distribution of the scattered light intensity, $g(\theta)$, and the aggregate size distribution, $P(N_p)$, are related by a Fredholm integral equation of the first-kind [11],

$$g(\theta) = C\int_{1}^{\infty} K(\theta,N_p)P(N_p)dN_p \tag{1}$$

where C is a scaling coefficient that depends on the experimental apparatus, the aggregate number density, and the optical properties of soot, $K(\theta, N_p)$ is the kernel function derived from electromagnetic theory, and $P(N_p)$ is the probability density for the number of primary particles per aggregate. Predicting the angular distribution of scattered light, $g(\theta)$, for a specified $P(N_p)$ can be done by simply carrying out the integration; this is called the *forward problem*. Solving the *inverse problem*, i.e. recovering $P(N_p)$ from $g(\theta)$, on the other hand, is mathematically ill-posed due to the smoothing action of $K(\theta, N_p)$. When carrying out the integral in Eq. (1), moderate variations in $P(N_p)$ are smoothed by $K(\theta, N_p)$ into comparatively smaller changes in $g(\theta)$; this is particularly the case for changes in $P(N_p)$ at small N_p, which are dominated by light scattered from large aggregates. As a consequence, small perturbations in $g(\theta)$, inevitable in an experimental setting, are amplified into large variations in $P(N_p)$ by the inversion process.

Until recently MAELS experiments were limited to few measurement angles, and consequently a distribution shape (often lognormal, e.g. [8–11], or self-preserving [12]) had to be assumed for the aggregate size distribution. The unknown distribution parameters are then found either through nonlinear regression, or by plotting the normalized scattering intensity as a function of the modulus of the scattering wave vector, $q(\theta)=4\pi\sin(\theta/2)/\lambda$ where λ is the detection wavelength and identifying features of this curve that correspond to different scattering regimes. This approach is far from ideal, however, since it biases the inferred distribution towards the distribution shape assumed by the analyst, which may not necessarily be correct.

Recent innovations in experimental apparatus [10,11,13] have led to the possibility of obtaining a much larger set of angular measurements, which in principle can be used to relax the need to assume a distribution shape. In a previous work [11], we demonstrated that $P(N_p)$ can be estimated by solving a matrix analogue of Eq. (1), $\boldsymbol{Ax}=\boldsymbol{b}$, where $\boldsymbol{b}$ contains the angular scattering measurements, $\boldsymbol{x}$ is a discrete form of $P(N_p)$ equivalent to a normalized histogram, and $\boldsymbol{A}$ is derived from $K(\theta, N_p)$. The smoothing property of $K(\theta, N_p)$ makes $\boldsymbol{A}$ ill-conditioned, which amplifies the noise contaminating the data into a very large error component in the recovered aggregate sizes. In Ref. [11] we showed that this issue can be addressed through regularization, in the form of Bayesian priors that promote smoothness, non-negativity, and distribution shapes. (Enforcing a distribution shape, as described above, is an extreme form of regularization.) While regularization stabilizes deconvolution of the MAELS data, it is undesirable since it biases the recovered solution towards the analyst's expectations.

While it has long been known that scattered light measurements made at different angles contain varying degrees of information about $P(N_p)$, a systematic and rigorous method for selecting measurement angles has yet to be proposed. Instead, most researchers either choose these angles heuristically by trial-and-error, or more simply use uniformly-spaced angles over the angular range afforded by the apparatus. By writing the MAELS problem in matrix form, however, it is clear that the ill-posedness of the underlying experiment can be quantified by the ill-conditioning of $\boldsymbol{A}$, which in turn depends on the