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Laser-induced incandescence: Particulate diagnostics for combustion, atmospheric, and industrial applications

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ABSTRACT

The understanding of soot formation in combustion processes and the optimization of practical combustion systems require *in situ* measurement techniques that can provide important characteristics, such as particle concentrations and sizes, under a variety of conditions. Of equal importance are techniques suitable for characterizing soot particles produced from incomplete combustion and emitted into the environment. Additionally, the production of engineered nanoparticles, such as carbon blacks, may benefit from techniques that allow for online monitoring of these processes.

In this paper, we review the fundamentals and applications of laser-induced incandescence (LII) for particulate diagnostics in a variety of fields. The review takes into account two variants of LII, one that is based on pulsed-laser excitation and has been mainly used in combustion diagnostics and emissions measurements, and an alternate approach that relies on continuous-wave lasers and has become increasingly popular for measuring black carbon in environmental applications. We also review the state of the art in the determination of physical parameters central to the processes that contribute to the non-equilibrium nanoscale heat and mass balances of laser-heated particles; these parameters are important for LII-signal analysis and simulation. Awareness of the significance of particle aggregation and coatings has increased recently, and the effects of these characteristics on the LII technique are discussed.

Because of the range of experimental constraints in the variety of applications for which laser-induced incandescence is suited, many implementation approaches have been developed. This review discusses considerations for selection of laser and detection characteristics to address application-specific needs. The benefits of using LII for measurements of a range of nanoparticles in the fields mentioned above are demonstrated with some typical examples, covering simple flames, internal-combustion engines, exhaust emissions, the ambient atmosphere, and nanoparticle production. We also remark on less well-known studies employing LII for particles suspended in liquids.

An important aspect of the paper is to critically assess the improvement in the understanding of the fundamental physical mechanisms at the nanoscale and the determination of underlying parameters; we also identify further research needs in these contexts. Building on this enhanced capability in describing the underlying complex processes, LII has become a workhorse of particulate measurement in a variety of fields, and its utility continues to be expanding. When coupled with complementary methods, such as light scattering, probe-sampling, molecular-beam techniques, and other nanoparticle instrumentation, new directions for research and applications with LII continue to materialize.

Summary of abbreviations and acronyms: AAC, aerodynamic aerosol classifier; AC-LII, auto-compensating LII; AMS, aerosol mass spectrometer; APM, aerosol particle mass (analyzer); CA, crank angle; CCD, charge-coupled device; CLiiME, community LII modeling environment; CMD, count median diameter; CPC, condensation particle counter; CPMA, centrifugal particle mass analyzer; CRDS, cavity ring-down spectroscopy; CVS, constant volume sampling; CW, continuous wave; DMA, differential mobility analyzer; EC, elemental carbon; EGR, exhaust-gas recirculation; ELS, elastic light scattering; HAB, height above burner; HDDV, heavy-duty Diesel vehicle; IR, infrared; LIDELS, laser-induced desorption with elastic light scattering; LIF, laser-induced fluorescence; LII, laser-induced incandescence; LIISim, LII simulation software; MD, molecular dynamics; NEXAFS, near-edge x-ray absorption fine-structure spectroscopy; nvPM, nonvolatile particulate matter; PAH, polycyclic aromatic hydrocarbon; PLIF, planar laser-induced fluorescence; PMS, particle-mass spectrometer; PMT, photomultiplier tube; RAYLIX, Rayleigh scattering, laser-induced incandesce, and extinction; SAXS, small-angle x-ray scattering; SEM, scanning electron microscopy; SMPS, scanning mobility particle sizer; SP2, single-particle soot photometer; TDC, top dead center; TEM, transmission electron microscopy; TiRe-LII, time-resolved LII; UV, ultraviolet; WAXS, wide-angle x-ray scattering; XPS, x-ray photoelectron spectroscopy.

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Contents

1. Introduction

The past several decades have been marked by a growing concern over the effects of combustion-generated particles on human health and the environment. Numerous scientific studies have indicated that soot particles, commonly produced during the incomplete combustion of hydrocarbons, pose a serious public health risk, *e.g.* Ref. 1. The effects of atmospheric soot emissions have gained a great deal of recent attention, stemming from growing concerns over the contribution of black-carbon particles to climate change. Most atmospheric particles reflect incoming solar radiation and tend to partially counteract global warming; soot particles, in contrast, strongly absorb in the visible and near infrared and are associated with significant positive radiative forcing leading to global warming. Models predict that the radiative forcing from atmospheric soot is ~65% that of CO_2, the most abundant greenhouse gas [2]. Relative to CO_2, however, soot has a short lifetime in the Earth's atmosphere, and reducing soot emissions into the atmosphere has been proposed as a near-term climate-change mitigation approach [3]. Black carbon can also settle on snow and ice, and thus decrease their reflectivity. In addition to increasing the radiative forcing because of this albedo effect, the additional surface absorption can accelerate melting of snow and ice, which, in turn, further enhances surface absorptivity and global warming [2,4].

Hydrocarbon combustion is the main source of atmospheric soot emissions, and, although soot chemistry has been studied extensively over decades, some of the primary mechanisms in soot formation, growth, and oxidation are uncertain [5–8]. Controlling soot emissions will require focused efforts to understand soot formation and oxidation under a variety of combustion conditions. Combustion is also the main source for industrially manufactured carbon nanostructures and provides various opportunities for the synthesis of non-carbon nanoparticulate materials [9]. Experimental studies of particulate formation in the context of both reduced emissions and targeted synthesis rely heavily on the ability to measure and characterize particles over a wide range of conditions.

Whereas incipient soot is close to spherical, small (1–10 nm), and composed of large organic species with high hydrogen-to-carbon ratios, mature soot is composed of primary particles of 10–50 nm in diameter with fine structures similar to polycrystalline graphite (low hydrogen-to-carbon ratios) [10–14]. These primary particles are covalently bound into branched-chain aggregates of tens to hundreds of nanometers in size. These aggregates are non-spherical and are characterized by fractal dimensions typically in the range of 1.7–1.9 [11,14–18]. The non-sphericity of these particles complicates optical measurements of them. In addition, these particles can be coated with semi-volatile coatings in the combustor, exhaust stream, or atmosphere, which can significantly change their physical