A combined laser induced incandescence, aerosol mass spectrometry, and scanning mobility particle sizing study of non-premixed ethylene flames

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Investigations into the chemical composition of soot particles often rely on samples extracted from the flame environment for subsequent analysis by mass spectrometry. These mass spectra have contributed to the general consensus that polycyclic aromatic hydrocarbons (PAHs) are involved in soot nucleation and surface growth processes. In the pioneering work of Dobbins et al.,[1,2] soot and soot precursor particles were extracted along the centerline of a non-premixed coflow flame by rapid-insertion thermophoretic sampling and subsequently analyzed by laser microprobe mass spectrometry (LMMS). Low in the flame, where TEM images suggested the particles were liquid-like, the mass spectrum was dominated by species between 200 and 300 amu in size. Higher in the flame, where TEM images indicated that carbonaceous aggregates were being formed, PAHs were no longer observed in the mass spectra. The laser-desorption ionization experiments of Bouvier et al.,[3] Lemaire et al.,[4] and Faccinetto et al.[5] complemented and expanded the findings of Dobbins et al.[1,2] Lemaire et al.[4] showed that the fuel composition strongly influences the soot composition. Faccenetto et al.[5] developed a new sampling method enabling some distinction between PAHs in the gas phase and those adsorbed onto the soot particles. Intrusive sampling techniques such as those used in the studies referenced above perturb the flame. In some instances such techniques may permit the agglomeration of existing particulates, condensation of low vapor pressure species onto the surface of soot nuclei, and nucleation of new clusters that could later be erroneously associated with nascent soot. Recently, we have coupled a non-premixed, opposed-flow flame system to an aerosol mass spectrometer to investigate the chemical composition of soot particles extracted from different regions of the flame. The present work provides insight into the effects of our intrusive sampling method on the observed soot composition and size distributions by combining laser-induced incandescence (LII) measurements with intrusive particle diagnostics.

In this work, we performed time-resolved LII measurements in conjunction with flame-sampling aerosol mass spectrometry (AMS) and scanning mobility particle sizing (SMPS) measurements to (1) investigate differences in particle size and composition as a function of temperature and position in the flame and (2) investigate processes occurring within the sampling system during sample extraction. We probed three non-premixed, opposed-flow, ethylene flames at conditions ranging from nearly sooting to moderately sooting. In situ time-resolved LII measurements revealed differences in the temporal response as a function of laser fluence and position in the flame. These differences may result from varying absorption coefficients due to particle composition and/or differences in the extent of particle surface coatings. Particles extracted from different regions of the flame also yielded varying temporal LII profiles, size distributions (as determined by SMPS), and chemical compositions (as determined by AMS). The use of a thermal denuder prior to the ex situ LII, SMPS, and AMS instruments also provided information on the extent of PAH condensation onto existing particles and the propensity for liquid- or tar-like droplet nucleation within the sampling system.

References

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