OPTICAL EXPERIMENTS IN LUMINOUS FLAMES AND IMPLICATIONS ON SOOT REFRACTIVE INDEX †

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The formation of soot particles reduces flame temperature due to the enhanced radiation heat loss, significantly affecting the efficiency and durability of many internal combustion engines. In addition, soot emissions from various combustion sources have been reported to have adverse effects on human health. Soot has also been identified as a major contributor to global warming, only second to CO_2. These technological and environmental implications motivate research for a complete understanding of the factors governing the complex soot processes in combustion systems.

In a previous study, we had proposed a simple diagnostic technique that is capable of characterizing soot physical properties in flames based on two-angle laser scattering and extinction measurements. This non-intrusive diagnostic technique drastically reduces the experimental efforts in the characterization of soot-containing flames by eliminating the time-consuming and complicated multi-angle scattering measurements. Furthermore, it separates the formation, growth, aggregation processes by yielding the soot volume fraction, spherule diameter, and mean aggregate size based on the proper consideration of actual soot morphology. In the present study, we implemented this simple laser diagnostic technique in laminar non-premixed flames burning ethylene and methane in air at atmospheric conditions. Scattering coefficients were measured using an argon-ion laser and calibrated optics at various axial and radial locations in a co-flowing burner. Local extinction coefficients were also obtained from the deconvoluted line-of-sight transmission measurements. The data interpretation procedure was based on the Rayleigh-Debye-Gans scattering theory, which has been shown to account for the aggregate morphology. Ex-situ thermophoretic sampling experiments (TS) followed by direct observations on a transmission electron microscope (TEM) also complemented the in-situ measurements independently. The present data analysis focused on the soot spherule diameter and volume fraction, which are the parameters of primary interest in soot-containing flames. The results were specifically presented here for various heights above the burner exit along the centerline and soot layer of the ethylene flame.

In order to reduce the experimental efforts significantly, assumptions were made in the analysis to compromise well established soot parameters. For example, extensive studies confirmed the fractal dimension and prefactor of soot aggregates to be 1.8 and 2.2, respectively, as these fractal properties were found to be remarkably insensitive to flame conditions and particle chemical composition. Furthermore, size distributions of many aerosols could be reasonably represented with a geometric width of 2.3, in agreement with the values reported for soot aggregates. Finally, an exponential scaling distribution was better suited for soot in comparison to the log normal distribution, which evidently fails for moments higher than the second one. The use of the above-mentioned values for

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fractal properties and width of aggregate size distribution eliminates the need to make multi-angle laser scattering experiments. Then, soot volume fraction, spherule diameter, and mean aggregate size at a particular flame location could be conveniently obtained from the measurements of scattering coefficients at two angles, here at 30 and 150 degrees, and extinction coefficient.

The results presented here focus on the soot volume fraction, f_v, and spherule diameter, d_p, which were inferred along the centerline (flame axis) and annular region of peak soot concentrations (soot layer) in the ethylene flame. In general, both scattering and extinction coefficients increased with increasing height along the flame centerline, whereas maximum values were reached at an axial location of about z = 40 mm along the soot layer. Although there was no light extinction at the center points below z = 25 mm, there was considerable amount of scattered light at this early soot nucleation region. This interesting feature, which had also been noticed by other researchers, evidently captured the formation of first soot precursor particles, which do not absorb light appreciably at visible wavelengths. A similar behavior was also observed in the methane flame.

Soot volume fractions obtained using two different refractive indices of m_1=1.62+0.66i and m_2=1.90+0.55i were shown in Figure 1 at the flame axis and radial locations of peak soot concentrations. Included in the figure were also the independent TS/TEM measurements, which were not affected by the uncertainties of soot refractive. Soot volume fractions inferred with m_2 were about 50% higher than those were found with m_1 (compare values for Center-m_2 to Center-m_1 and Peak-m_2 to Peak-m_1). This behavior was simply related to the difference in the refractive index function, $E(m) = Im[(m^2-1)/(m^2+2)]$, because $E(m_2)$ = 0.19 is smaller than $E(m_1)$ = 0.29. The optical results using the first refractive index, however, agreed much better with the independent TS/TEM experiments along the centerline.

Figure 1

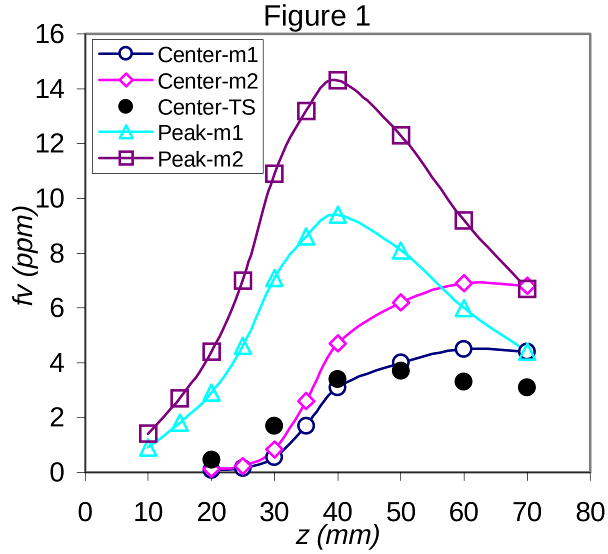
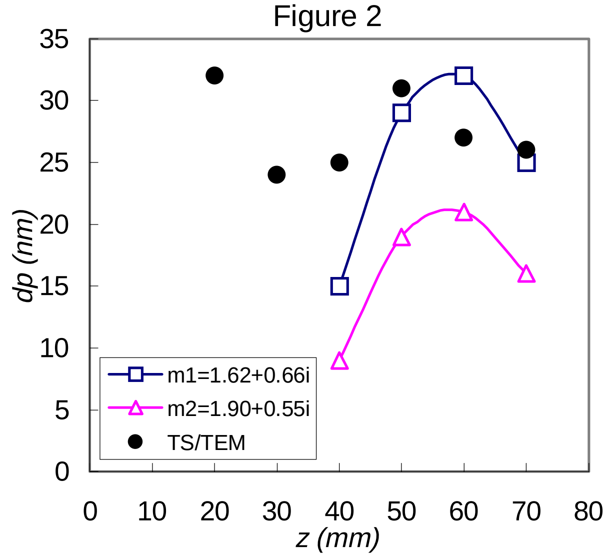


Figure 2 showed the spherule diameters along the centerline of the ethylene flame based on the analysis of optical measurements using the two refractive indices as well as the direct observations of soot samples from the flame. A comparison of results above z = 40 mm indicated that m_1=1.62+0.66i yielded primary particles sizes that were in good agreement with the TS/TEM measurements within experimental uncertainties. Thus, in-situ recoveries of both f_v and d_p appeared to improve when this first refractive index was used at the flame axis. The optically inferred soot particle sizes lower than z = 40 mm were

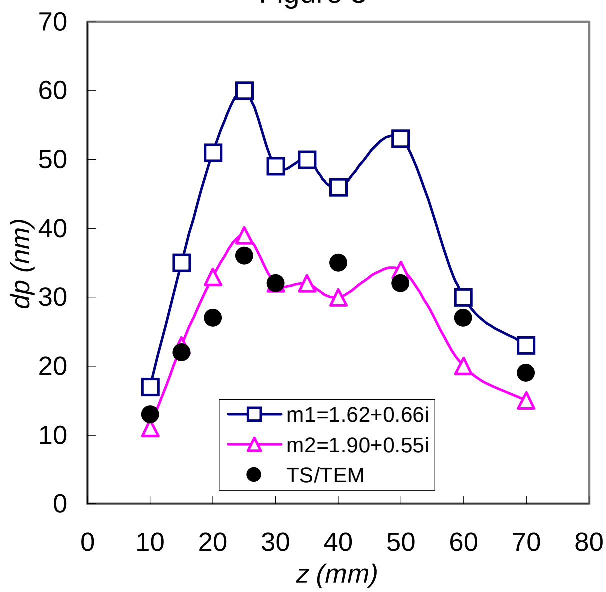
Figure 2



not included in the figure because they were almost less than 1 nm, irrespective of the refractive index value employed. This might be argued due to the failure of the aggregate scattering theory to predict the optics of small particles. Nevertheless, even the use of Rayleigh scattering theory consistently resulted in d_p's of about 13 nm at every such flame locations, failing to estimate the changes in soot spherule sizes. Therefore, this disagreement between the in-situ and ex-situ measurements was possibly due to the presence of a mixture of translucent and mature soot particles at the early stages of formation region that were seen on our TEM samples. This explanation appears to be more plausible in view of the fact that the composition, and consequently, refractive index of particles varies during the carbonization process of translucent soot precursor particles to mature aggregates. One may then ask if this evolution could be quantified although this problem was not the main objective of this study. A different analysis using the spherule size data as input to our optical measurements could yield the ratio of refractive index functions, *E/F*, where $F(m)=|(m^2-1)/(m^2+2)|^2$. Such an inverse consideration indicated that *E/F* ratio should increase from about 1.1 to 7 and then to 17 as the axial height, *z*, respectively decrease from 40 mm to 30 and then to 20 mm at the centerline of the ethylene flame.

The spherule sizes were also obtained at various radial locations corresponding to the soot layer in the ethylene flame, as seen in Figure 3. In contrast to the centerline results, optical measurements of d_p agreed with the independent TS/TEM experiments when the second refractive index of m_2=1.90+0.55i was used in the analysis. These results pointed out that soot refractive index, which is the dominant uncertainty in optical techniques, varies with not only the radial position but also the axial position even in our simple laminar non-premixed flames. Certainly, this observation tremendously complicates the radiation heat transfer predictions and laser-based experiments in fuel-rich conditions of various combustion devices. It also explains the source of controversy among numerous investigations on soot refractive index. Given the fact that soot refractive index is a fundamental property in radiative transfer calculations, it is essential to quantify its variations not only in laminar flames but also in other practical luminous flames involving turbulent conditions.

Figure 3



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