COMPARISONS OF SOOT VOLUME FRACTION MEASUREMENTS USING OPTICAL AND ISOKINETIC SAMPLING TECHNIQUES

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INTRODUCTION

In a previous study, the performance of a three-line optical probe technique for measuring soot volume fraction and temperature was assessed by conducting experiments in the path-invariant environment of a premixed flame [1]. In that study, the temperature and soot volume fraction (f_{va}, based on absorption measurements at 632.8 nm and f_{ve}, based on emission measurements at 900 nm and 1000 nm) measurements in a premixed ethylene/air flame were compared to the results of Harris and Weiner [2]. Although the temperatures and mean soot volume fractions compared favorably, the discrepancy between f_{va} and f_{ve} prompted new measurements to evaluate the importance of source wavelength on the f_{va} measurements, scattering by soot particles, light absorption by 'large' molecules and the use of different indices of refraction reported in the literature.

The experiments on the degree of soot scattering and light absorption by 'large' molecules indicated that these effects cannot reconcile the observed discrepancy in the soot volume fractions [1]. The measured soot volume fractions were sensitive, however to the absorption coefficient and therefore varied significantly when different sets of refractive indices were used. Furthermore, the agreement between f_{va} and f_{ve} was improved when extinction measurements were performed with longer wavelength light sources.

In recent years, the effects of the chemical properties of soot on the indices of refraction have been a source of controversy. The variations in the refractive indices measured by different investigators result in a large uncertainty in the soot volume fraction determined by optical techniques. In many of the previous studies [3-5], the variables of the Drude-Lorentz dispersion model were varied to fit the transmittance or the reflectance data. However, Felske and Ku [6] suggest that the use of the Drude-Lorentz dispersion relationship cannot be expected to produce accurate indices of refraction for a material such as soot (for which the properties are not well-characterized) when it can't predict the spectral features of a well-characterized material such as graphite. For this reason, it is desirable to calibrate the optically determined soot volume fractions by performing measurements that are independent of the soot refractive index. Isokinetic soot sampling experiments were performed in the flame in conjunction with the optical techniques for comparison. This technique does not rely on the refractive indices of soot and therefore provides an independent measure of the soot volume fraction. The soot volume fractions measured using this technique were typically within a factor of two of f_{va} and f_{ve} (calculated using various indices of refraction).

In the high temperature environments, there is uncertainty associated with the isokinetic technique (i.e., the uncertainties in the temperature at the probe entrance, density of soot and the assumption of quenching of further soot reactions). However, these uncertainties can be significantly reduced by performing experiments under lower-temperature conditions. For these reasons, additional optical/sampling experiments were performed in regions far downstream of the reaction zone.

ISOKINETIC SOOT SAMPLING EXPERIMENTS

In prior studies, isokinetic sampling experiments have been used primarily for analyzing particle/gas mixtures for conditions that are not far removed from the ambient. However, few studies have been performed in the high temperature environment of a premixed or diffusion flame [7]. In this study, isokinetic soot sampling experiments were performed using a premixed acetylene/air flame for conditions identical to the experiments performed in a previous study [1]. **Figure 1** displays the schematic drawing of the experimental arrangement. The probe consists of a 9.6 mm (O.D.) stainless steel pipe fitted with a teflon filter assembly that has a collection efficiency greater than 99% for particles larger than 0.1μ [8]. The probe was water-cooled with a 6.4 mm (O.D.) diameter copper tube (starting at approximately 3 mm from the tip of the probe) to quench further reactions of the soot as it traversed the length of the probe. Inevitably, there were substantial deposits of soot on the inner

surface of the cold probe. The deposited soot was scrubbed from the probe and was included in the determination of the total soot mass collected within the 10 minute sampling period. Since the gas flow was measured at room temperature, the flow at the probe entrance was calculated from the ideal gas law by multiplying it by the ratio of the temperature at the inlet of the probe (from emission measurements) to the ambient temperature. The flow was corrected for water condensation in the sampling probe. This increased the actual flow by only a few percent. The soot volume fraction was calculated from the following formula:

$$f_v = \frac{\dfrac{m_s \, T_\infty}{\rho_s \, T_{pe}}}{t \dfrac{dV}{dt}} \tag{1}$$

where m_s is the dry mass of the collected soot (after being placed in an oven at 110°C for 15 hours), T_{pe} is the temperature at the probe entrance, T_∞ is the ambient temperature, t is the collection period, dV/dt is the measured volumetric flow rate and ρ_s is the density of soot (1.84 ± 0.1 g/cm^3). The density of soot was measured using the Helium Pycnometry technique with soot collected on a stagnation plate placed 30 cm above the burner surface.

Soot sampling measurements were performed at a position of 13 mm above the burner surface (*corresponds to h = 8 mm*) to determine the reproducibility of this technique. The optically-determined soot volume fraction was 0.40 ppm (using Dalzell-Sarofim [3] dispersion equation to calculate the absorption coefficient) compared to the isokinetically measured soot volume fraction of 0.48 ppm. Soot sampling experiments were also performed at a position 30 mm above the burner surface, where the optically determined soot volume fraction was 1.41 ppm compared to the soot sampling value of 0.60. ·

Isokinetic experiments were also performed in regions far downstream of the reaction zone. The schematic diagram of the experiment is shown in **Figure 2**. Nitrogen was injected from two ports on the side of the quartz tube to reduce the temperature of the soot dispersion measured at location A. The transit time from the flame to the sampling point was about 2.5 seconds. Experiments were performed for three separate equivalence ratios, ϕ, of 2.3, 2.5 and 2.7. The measured temperatures were approximately 500 K for all three cases. At position A, two holes were drilled in the quartz tube to allow for passage of the 632.8 nm He-Ne laser beam for the absorption measurements. For each experiment, absorption measurements were performed in conjunction with the isokinetic soot sampling experiments. The f_{va} values were calculated from the following equation:

$$f_{va} = -\frac{\ln\left(\dfrac{I_\lambda}{I_{\lambda o}}\right)\lambda}{K_\lambda L}$$

where K_λ is the absorption coefficient, λ is the wavelength of the incident light ($I_{\lambda o}$) and transmitted light (I_λ) and L is the path length. The absorption coefficient was derived using the values of Dalzell and Sarofim [4]. **Figure 3** displays average soot volume fractions measured using both techniques. The repeated measurements agreed within 10% for ϕ = 2.3 and 2.5; while for ϕ=2.7, the results were less repeatable.

DISCUSSION

Our isokinetic measurements of soot volume fraction indicate that optical measurements using the Dalzell-Sarofim refractive index together with the Rayleigh-limit expression for the absorption coefficient overestimates the soot volume fraction by about 40% for the diluted soot. Approximate agreement between the two methods is obtained if the absorption coefficient is taken to be 7.32 (instead of 4.87). This would correspond to a refractive index of 1.55 +0.86i. Of course, the refractive index is not uniquely determined. We have fixed the real part to provide an indication of the change in the imaginary part required to provide agreement between the optical measurement and the sampling measurement.

Optical measurements in the flame indicated that the degree of light scattering by soot agglomerates was

negligible when compared to the absorption measurements. However, this may not be the case for the diluted soot, since the 2.5 second transit time allows for significant particle agglomeration. Other studies indicate that the ratio of total scattering to extinction is on the order of 25% for large agglomerates. Additional measurements are planned to quantify the total scattering and also to measure the wavelength dependence of the absorption coefficient.

ACKNOWLEDGEMENTS

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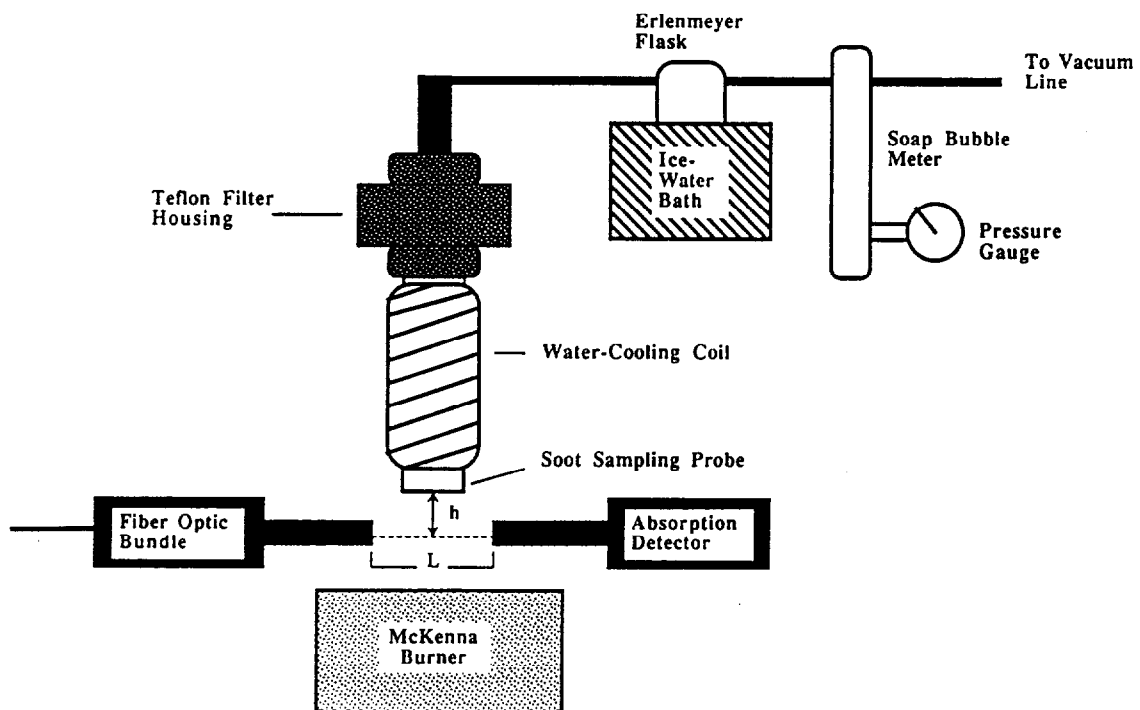
Fig. 1: Flame isokinetic sampling experiment

Fig. 2: Diluted soot isokinetic sampling experiment

To Vacuum

Filter Assembly

Detector

Laser

Position A
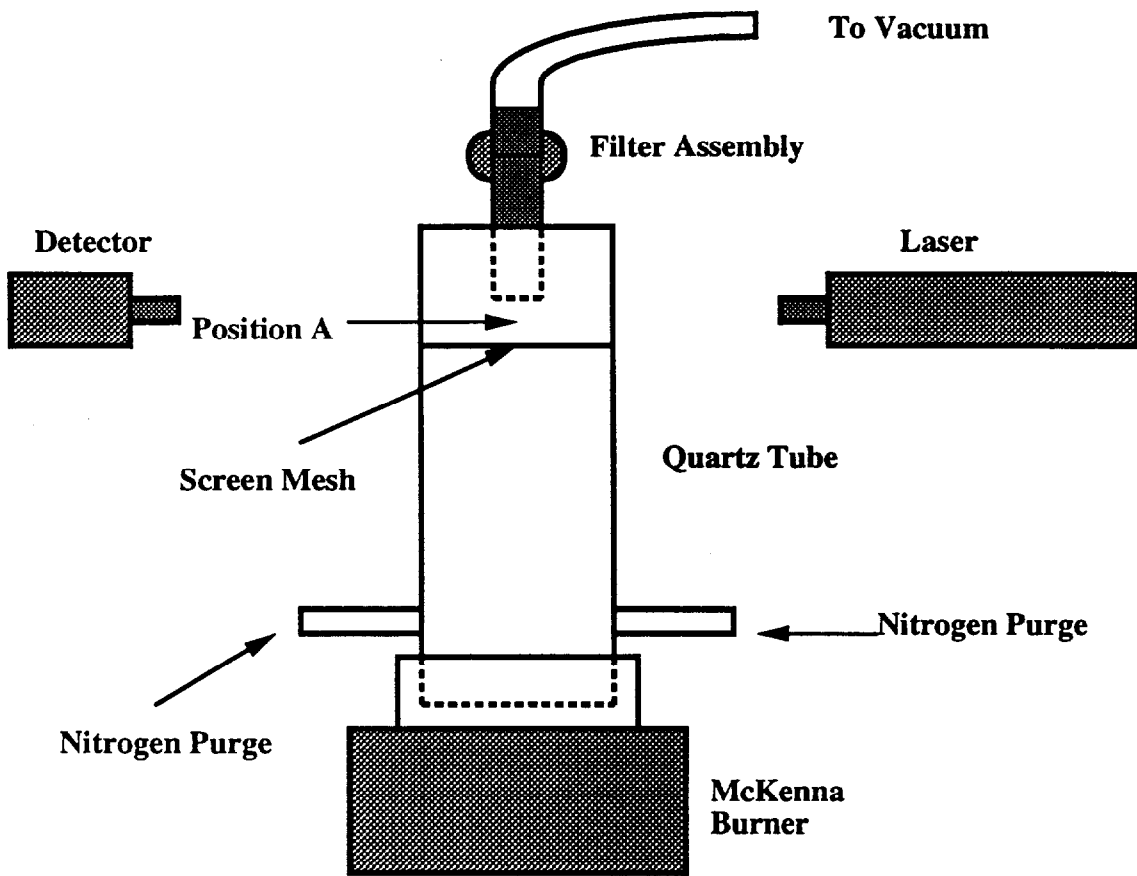
Screen Mesh

Quartz Tube

Nitrogen Purge

Nitrogen Purge

McKenna
Burner

Fig. 3: Optical and isokinetic soot volume fractions

Optical Experiments

Isokinetic Experiments

Soot Volume Fraction (ppm)

Equivalence Ratio