Investigation of Sooting in Microgravity Droplet Combustion: Fuel-Dependent Effects

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Introduction

Kumagai and coworkers first performed microgravity droplet combustion experiments [Kumagai, 1957]. The primary goal of these early experiments were to validate simple ‘d²-law’ models [Spalding, 1954, Godsave, 1954]. Inherent in the ‘d²-law’ formulation and in the scope of the experimental observation is the neglect of sooting behavior. In fact, the influence of sooting has not received much attention until more recent works [Choi et al., 1990; Jackson et al., 1991; Jackson and Avedisian, 1994; Choi and Lee, 1996; Jackson and Avedisian, 1996; Lee et al., 1998]. Choi and Lee measured soot volume fraction for microgravity droplet flames using full-field light extinction and subsequent tomographic inversion [Choi and Lee, 1996]. In this investigation, soot concentrations were measured for heptane droplets and it was reported that soot concentrations were considerably higher in microgravity compared to the normal gravity flame. It was reasoned that the absence of buoyancy and the effects of thermophoresis resulted in the higher soot concentrations. Lee et al. [1998] performed soot measurement experiments by varying the initial droplet diameter and found marked influence of sooting on the droplet burning behavior.

There is growing sentiment that sooting in droplet combustion must no longer be neglected and that “perhaps one of the most important outstanding contributions of microgravity droplet combustion is the observation that in the absence of asymmetrical forced and natural convection, a soot shell is formed between the droplet surface and the flame, exerting an influence on the droplet combustion response far greater than previously recognized.” [Law and Faeth, 1994]. One of the methods that we are exploring to control the degree of sooting in microgravity is to use different fuels. The effect of fuel structure on sooting propensity has been investigated for over-ventilated concentric coflowing buoyant diffusion flames [Glassman, 1996]. In these investigations, the fuel flowrate was increased until smoke was observed to escape from the “luminous visible flame” [Glassman, 1996]. A total of 29 fuels were used in order to characterize relative sooting propensity. The sooting propensity of a particular fuel was assessed by comparing the flowrates for soot emission from the tip of the flame. It was reported that the sooting tendency for diffusion flames increased for fuels with higher rates of pyrolysis.

Randolph and Law [1986 and not 1994] also examined the effect of fuel structure on droplet sooting behavior. In their experiments the droplets were separated from the bulk gas stream and quenched with nitrogen prior to gravimetric measurements. A variety of fuels were studied, namely aromatics, phenyl-alkanes and alkanes. The results were in qualitative agreement with the work of Glassman [1986]. Vander Wal et al. [1994] performed relative soot concentration measurements using laser-induced incandescence for heptane and decane fuel droplets burning under normal-gravity conditions. It was found that soot volume fractions for decane was more than a factor of two larger than that for heptane.

Although the normal-gravity investigations have provided some important insights regarding the influence of fuel structure on the sooting behavior of droplet flames, results cannot be easily extrapolated for microgravity studies since increased residence times and thermophoretic effects must be considered in greater detail. Several studies have compared sooting behavior of different fuel droplets burning under microgravity conditions [Card and Choi, 1990; Jackson et al., 1991; Jackson and Avedisian, 1994], however, detailed quantitative results were not provided. In all of these previous studies, the degree of sooting was only visually assessed from an incandescent backlighted image of the soot containing region. Such techniques can provide misleading results regarding sooting behavior [Choi, 1996].

Experimental Description

All experiments were performed in the 2.2 drop tower at NASA-Lewis Research Center. Figure 1 is a schematic of the experimental apparatus. Only a brief discussion will be given since details can be found elsewhere.

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669
[Lee et al., 1998]. On the top optical plate, light from a 635 nm variable intensity diode laser attached to a single fiber optic cable was expanded to a diameter of 50 mm. The beam was collimated and directed through the top optical port of the 12 liter stainless steel combustion chamber using a mirror positioned at 45 degrees. The beam was transmitted through the combustion chamber and focused using a 200 mm focal length plano-convex lens and redirected using a second mirror positioned at 45 degrees. The reflected beam was then imaged through a spatial filter to a high resolution CCD camera located on the bottom plate. The video output from the camera was recorded onto Betacam and S-VHS recorders at 30 frames per second via fiber optic link.

Discussion and Results

Experiments were performed for heptane and decane fuels at 0.75 atm. Reduction in pressure was necessary to circumvent the large signal attenuation that was observed for the 1 atm experiment. Pressure reduction is a proven method for reducing sooting in microgravity droplet combustion [Lee and Choi, 1997]. The reduction in flame temperature and the reduction in the residence time for fuel vapor transport within the fuel rich region are probable factors for reduced sooting at lower ambient pressures. Soot volume fraction was calculated by deconvoluting the radial distribution of the transmittance measurement using a 3-pt Abel technique [Choi and Lee, 1996]. Figure 2a displays the spatially-resolved soot volume fraction at various normalized time (time divided by square of initial droplet diameter) for a 1.47 mm decane droplet burning in air at 0.75 atm. This experiment is compared with a 1.55 mm heptane droplet experiment performed at the same condition (Figure 2b), and some remarkable differences are noted (analysis of similar sized droplet was essential since the initial droplet size is known to affect the residence time within the fuel-rich region and thus cause variations in the degree of sooting [Lee et al., 1998]. It is observed that the maximum soot volume fraction for decane is nearly 30% higher than for heptane. Comparisons based solely on soot volume fraction, while significant, may be misleading. It is possible for droplets to have similar magnitude of soot volume fraction but different magnitudes in sooting due to the extent of soot containing region and the location of the sootshell. For example, Figure 3 displays the sootshell standoff ratio and flame standoff ratio for the heptane and decane experiments. It is shown that while the flame standoff ratios are similar, the sootshell location for decane is much smaller. Therefore, the higher soot concentration (measured at the sootshell) for decane may have been an artifact of constraining the formed soot particles to reside in a smaller shell volume. In order to compare the bulk sooting behavior of the two fuels, the mass of soot was calculated by integrating the soot volume fraction distribution with respect to the volume of the soot containing region and multiplying by the density of soot.

\[
m_{\text{soot}} = \rho_{\text{soot}} \int_{r_i}^{r_f} f_s 4\pi r^2 dr
\]  
(Eqn. 1)

The limits r_i and r_f are the instantaneous droplet and flame diameters, respectively and the soot density used was 1.8 g/cm^3 [Choi et al. 1995]. The soot mass for the decane experiment was 1.35 \mu g and for the heptane experiment 1.16 \mu g. Although the soot volume fraction is different by 30 %, the total soot mass varies by only 16%.

A possible explanation for the difference in sooting tendency is related to the residence time for fuel vapor transport from the droplet surface to the flame front. The residence time for fuel vapor transport from the droplet surface to the flame front has been shown to be:

\[
t_r = \frac{d_o^2}{4(K\rho_l)} \rho_g^{-1} D
\]  
(Eqn. 2)

[Jackson and Avedisian, 1994] where d_o is the initial droplet diameter, \rho_l is the liquid phase density of the fuel, K is the burning rate and \rho_g is the gas phase density of fuel vapor and D is the mass diffusivity. The burning rate was calculated for the decane and heptane experiments by performing a linear regression fit to the data after the respective heat up period. The burning rate was 0.61 mm^2/s and 0.58 mm^2/s for the decane and heptane experiment, respectively. Since the thermophysical properties are similar [Beaton and Hewitt, 1989], this analysis reveals that the difference in sooting cannot be attributed to residence time.

The increase in sooting for decane compared to heptane is in qualitative agreement with the normal-gravity experiments of Kadota et al. [1977]. In their work, the effects of fuel structure (pentane, heptane, decane, dodecane, hexadecane) on droplets burning under normal-gravity was investigated under ambient pressures ranging from 0.1 to 3.0 MPa. The sooting propensity was characterized by gravimetrically measuring the soot mass emitted from the open-tipped flames. It was found that the mass of emitted soot increased with increased number of carbon atoms in
Fig. 1-Schematic of Experimental Rig

Fig. 2a-Soot volume fraction distribution for 1.47 mm n-decane droplet, 0.75 atm

Fig. 2b-Soot volume fraction distribution for 1.55mm n-heptane droplet, 0.75 atm

Fig. 3-FSR and SSR for n-heptane and n-decane experiments
the fuel. In the experiments of Vander Wal et al. [1994], the relative soot volume fraction for decane was 120% greater than the soot volume fraction for heptane. It is believed that the main factor for the increase is related to the higher rates of fuel pyrolysis [Glassman, 1988] and that perhaps heavier precursors are more readily formed for decane compared to heptane [Vander Wal et al., 1994].

As mentioned previously, the soot standoff ratio for the decane and heptane experiments are quite different. Soot-shell is formed at the location where thermophoretic and drag force (induced by Stefan flux) are in balance. The thermophoretic flux is proportional to the temperature gradient and independent of particle size (for particles that are much smaller than the mean free path) and is displayed in eqn. 3a.

\[
\rho V_r = \frac{-3\nu \frac{dT}{dr}}{4(1 + \frac{\alpha \pi}{8})T} \quad \text{(Eqn. 3a)}
\]

\[
\rho V_r = \frac{\rho K}{16r^2} d \quad \text{(Eqn. 3b)}
\]

where \(\alpha\) is the accommodation coefficient (0.9) and \(\nu\) is the kinematic viscosity. To estimate the magnitude of the temperature gradient that would exist, the adiabatic flame temperature was calculated for decane and heptane at 0.75 atm and found to be 2268K and 2266K, respectively. Considering that thermophysical properties and flame standoff ratio for decane and heptane are similar [Vargaftik 1983], it is believed that the thermophoretic flux distribution for the two cases are also similar. Since the measured burning rates for both fuels are nearly equal, it is expected that the Stefan flux induced drag (eqn. 3b) are also expected to correspond. Therefore, additional work is required to determine the factors causing the difference in the soot standoff ratios.

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**References**