SOME RECENT OBSERVATIONS ON THE BURNING OF ISOLATED N-HEPTANE AND ALCOHOL DROPLETS

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INTRODUCTION

In a joint program involving Prof. F.A. Williams of the University of California, San Diego and Dr. V. Nayagam of the National Center for Microgravity Research on Combustion and Fluid Dynamics, the combustion of liquid fuel droplets of n-heptane, n-decane, methanol, methanol-water, ethanol and ethanol-water having initial diameters between about 1 mm and 6 mm continues to be studied. The objectives of the work are to improve fundamental knowledge of droplet combustion dynamics for pure fuels and fuel-water mixtures through microgravity experiments and theoretical analyses. The Princeton contributions to the collaborative program supports the engineering design, data analysis, and data interpretation requirements for the study of initially single component, spherically symmetric, isolated droplet combustion studies through experiments and numerical modeling. The complementary UCSD contributions apply asymptotic theoretical analyses and are described in the published literature and in a companion communication in this conference. The combined program continues to focus on analyses of results obtained from Fiber Supported Droplet Combustion (FSDC) experiments (FSDC-2, STS-94) conducted with the above fuels in shuttle cabin air and Droplet Combustion Experiment (DCE) data obtained for unsupported and fiber supported droplets of n-heptane in Helium-Oxygen mixtures and cabin air (STS-83, STS-94). The program is preparing for a second DCE experimental mission using methanol/methanol-water as fuels and helium-oxygen-nitrogen environments. DCE-2 is to be conducted aboard the International Space Station.

Emphases of recent Princeton work are on the study of simple alcohols (methanol, ethanol) and alcohol/water mixtures as fuels, with time-dependent measurements of drop size, flame-stand-off, liquid-phase composition, and finally, extinction. Ground based experiments have included bench-scale studies at Princeton and collaborative experimental studies in the 2.2 second drop tower at NASA-Glenn Research Center.

METHANOL

Methanol, an alternative fuel used in internal combustion systems as well as a hydrogen carrier for fuel cells, belongs to a class of fuels for which some of their combustion intermediates and products are highly soluble in the fuel itself. Methanol is unique because, for over-all equivalence ratios and pressures of interest in combustion systems, its oxidation kinetics leads to negligible formation of soot precursor species. The absence of soot, the relative simplicity of the detailed oxidation kinetics, and the substantial characterization of physical and thermophysical properties makes isolated methanol droplet combustion an ideal subject for developing and validating robust detailed models which test coupling effects of fundamental physical and chemical submodels on burning rate, flame structure, extinction, non-luminous radiation, product dissolution, and liquid phase mixing. Based upon a computational approach initially developed by Cho et al. [1], a time-dependent numerical model for isolated droplet combustion has been developed that includes temperature-dependent physical properties, detailed gas phase kinetics, multi-component gas phase diffusion, gas phase spectral radiative transport, water dissolution-vaporization, and mass/energy transport within the liquid phase [2].

It is has been hypothesized that droplet formation processes, solutal and thermal Maragonni effects, and fiber tethers can generate liquid motions within isolated burning droplets, even when large relative gas/droplet motions are absent [3]. In order to maintain a tractable uni-dimensional

configuration, we estimate the effects of internal liquid phase motions as an increase in molecular transport properties in the liquid phase (increasing the "effective" liquid phase Peclet numbers for mass and energy transport). This approximation is reasonable for droplets at low gas/droplet relative velocity conditions because the Stefan flux velocity at the droplet surface is relatively large in comparison to the tangential surface velocity. As a result, while liquid phase motions have little influence on gas phase motions, the motions in the liquid phase strongly affect the liquid phase thermal and mass transport. The experimental observable with the most sensitivity to this effect is the integrated liquid phase water content as a function of time, and, in turn, its effect on droplet extinction. While this parameter has been measured in ground-based free fall droplet experiments [4], here, we report first data for drop tower experiments. The details of the experimental methodology will be described in the presentation. We have used experimental

Fig. 1. Comparison of computed accumulated liquid water mass fraction with experimental measurements. (Pe=0.5).

measurements to estimate the liquid phase mass Peclet number, i.e. the ratio of droplet surface regression velocity to liquid phase mass diffusive velocity, in various experiments including the new data, as well as those of Lee and Law [4]. Results for drop tower experiments, along with relevant calculations, are shown in Fig. 1.
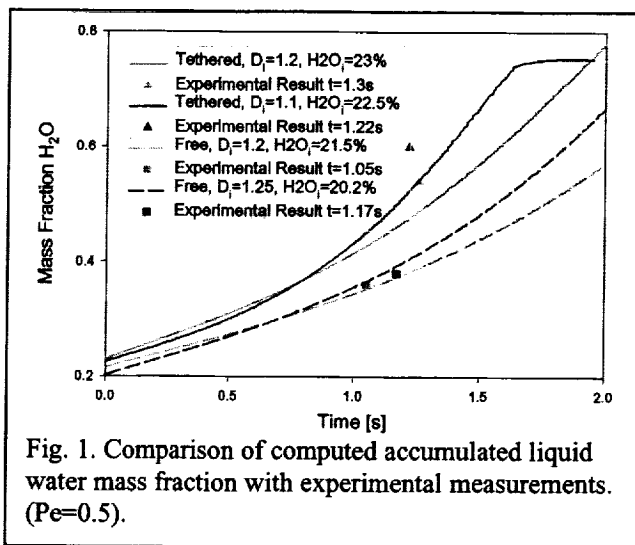
Well-mixed "distillation limit" behavior [5] is observed for a Peclet number less than 0.005, while the "diffusion limit" is reached for Peclet numbers greater than 5, with best comparison for drop tower results with a free droplet at a Peclet number of approximately 0.5. However, drop tower experiments with tethered droplets appear to be in the well-mixed, distillation limit, indicating that the presence of the fiber accelerates liquid phase motions within the burning droplet. The chemically reacting flow model has also been applied to the experimental conditions of Lee and Law [4] conducted using small, free falling droplets (150-250μm). Comparisons with the model suggest that the droplets in these experiments experienced a high degree of internal liquid phase motions.

ETHANOL

Ethanol can be manufactured with high efficiency from bio-mass and is under use or consideration as a motor fuel, as a fuel additive in reformulated and oxygenated gasoline, and as a hydrogen carrier for fuel cells. From the fundamental point of view, ethanol represents one of the simplest liquid fuels. Its gas-phase oxidation chemistry is relatively simple, it is readily soluble in water and, like methanol, ethanol absorbs water vapor from the gas phase, but it exhibits azeotropic behavior. Ethanol is known to be a non-sooting fuel at ambient pressures in air. However, at elevated pressures (starting from as low as 2 atm of air), ethanol droplet flames have been shown to form soot profusely [6,7]. Ethanol is therefore an ideal pure fuel for studying sooting phenomenon in general, and the effect of pressure on soot formation during droplet combustion in particular.

Microgravity experiments have been performed using ethanol and ethanol/water mixtures in freely falling ground based experiments [4] in drop towers [9,10], and as part of the FSDC-2 experiments noted above [11]. In the latter, each droplet combustion test occurred at pressures 0.996–1.107 bar, oxygen mole fraction 0.204–0.222, and relative humidity 39–46% of the

Spacelab environment. Droplets of pure ethanol and 4% water in ethanol (near the azeotrope) were studied; initial droplet diameter ranged from about 2.5 to 6 mm. Here we present the first numerical modeling results of the above experiments.

Detailed gas-phase kinetics was described with the reaction mechanism of [12] that consists of 142 reversible reactions of 33 species. The gas-phase diffusion and heat conduction terms [13], and transport data [13, 14] were taken from the literature or evaluated. The vapor pressure at the surface of ethanol/water mixtures was calculated using vapor pressure data [15], temperature-dependent latent heats were taken from the literature [16], and mixture liquid density was computed [16,17]. The value of the effective liquid diffusivity (D_l) was assigned by analogy with methanol results, and comparisons of the model predictions with the experimental measurements of [4] were used to validate the choice of D_l.

Exemplar model comparisons with droptower [9] and FSDC-2 experimental data are presented in Figs 2 and 3. The model shows a reasonable agreement with the experimental burning rate data for the entire range of the droplet sizes. With the exception of where droplet motions became excessive or tethered droplet sizes approached the size of the filament tethering-bead. Analysis of the model predictions indicated that the observed decrease in the average droplet burning rate is primarily a result of the influence of non-luminous radiative heat losses which become more pronounced at larger droplet sizes. The predictions of simplified steady-state models [18,19] that do not consider radiative losses
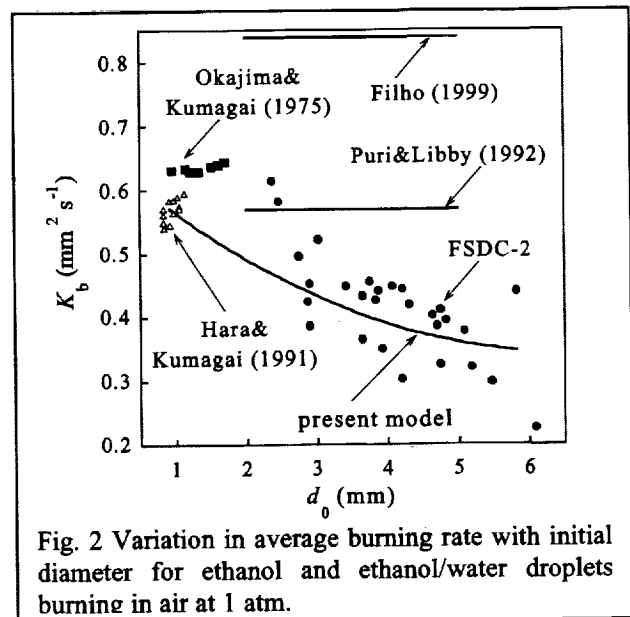

Fig. 2 Variation in average burning rate with initial diameter for ethanol and ethanol/water droplets burning in air at 1 atm.
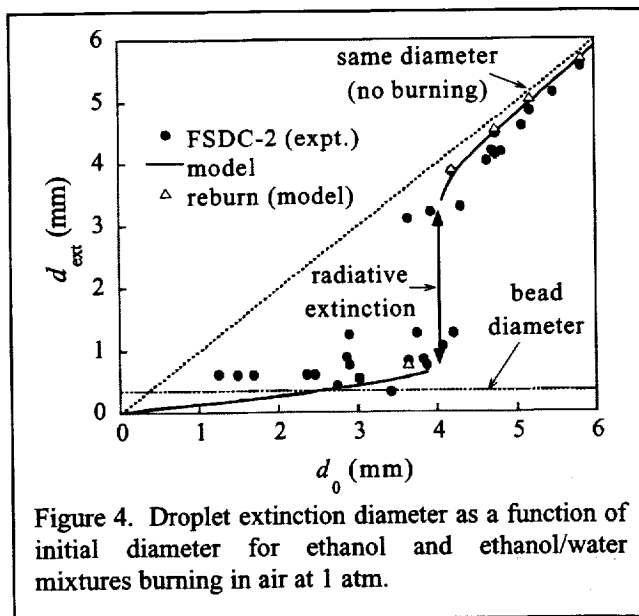

Figure 4. Droplet extinction diameter as a function of initial diameter for ethanol and ethanol/water mixtures burning in air at 1 atm.

(Fig. 2) are independent of initial drop size.

The model reproduced extinction diameter data obtained from FSDC-2 very well (Fig. 3). For droplets of less than 3 mm in diameter, extinction is mainly caused by water accumulation in the liquid phase, with experimental data compromised by the large tethering-bead. At $d_0 \sim$ 4 mm, both the model and experimental data show a sharp, nearly discontinuous increase in the extinction diameter, indicative of radiative extinction. At larger values of d_0, d_{ext} asymptotically approaches d_0, based upon transient burning after ignition, a factor dependent on ignition criteria. During the FSDC-2 experiments, some of the large droplets that burned and underwent extinction were subsequently re-ignited. This procedure was repeated until the droplet was fully consumed. Also shown in Fig. 3 is the sequence of re-ignition and extinction events calculated for the initially largest droplet in the data set (5.8 mm). The model closely follows the sequence of reburns reported in the experiments.

ONGOING WORK

We expect further refinement and testing of the methanol numerical model to benefited by a second DCE experiment devoted to the study of methanol and methanol/water combustion in helium/nitrogen/oxygen mixtures. The numerical model we have developed for n-heptane droplet burning is under modification to include an implicitly coupled description of processes related to droplet sooting. Results of DCE experiments will be compared with this model, initially, with further testing and validation of the model using data obtained in another program described at this work shop [20]. Finally n-heptane droplet ignition at elevated pressures and temperatures from 650 to 950 K [21,22,23] have exhibited "two-stage" ignition behavior. Others [24] have performed modeling of the higher temperature experiments (950 K) using a high temperature n-heptane semi-empirical kinetics [25]. We are presently conducting modeling studies of the lower temperature cases using a reduced mechanism that includes low temperature, cool flame, intermediate temperature, and high temperature kinetics.

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