ETHANOL DROPLET COMBUSTION AT ELEVATED PRESSURES AND ENHANCED OXYGEN CONCENTRATIONS

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In an effort to gain a better understanding of ethanol combustion, isolated droplet experiments were performed by varying initial droplet diameter, oxygen concentration, and ambient pressure. Experiments were performed at the NASA Glenn 2.2 sec. drop tower and the JAMIC 10 sec. dropshaft. These experiments revealed that while ethanol droplets burned in 1 atmosphere air without soot formation, luminous radiation from soot particles at higher pressures, with increased sooting at higher oxygen indices were observed. The measurement of the burning rate, soot standoff ratio and soot volume fraction are described. These experiments provide the first measurements of the soot volume fraction for ethanol droplets burning under microgravity conditions.

Introduction

Ethanol is a fuel that is regaining its popularity for use in practical applications. The use of ethanol as a motor fuel dates back to early 1900's. But, due to lower prices and wider availability, gasoline replaced ethanol as the primary motor fuel in those early years. Ethanol was reintroduced as a fuel additive in the 1970s with the advent of the oil crisis and stricter regulations on the amount of pollutants emitted from motor vehicles. Finally, the Clean Air Act Amendments of 1990[1] mandated the use of oxygenated fuels, such as ethanol and methanol, in regions of the country experiencing high levels of carbon monoxide.

There are many benefits of using ethanol as a motor fuel in terms of performance and pollutant mitigation. Recent investigations[2] on the emissions from engines operated using ethanol-containing fuels show a reduction in the carbon monoxide tail-pipe emissions in the entire operating range. Nag et al.[3] found that addition of ethanol produces a reduction of 92% in the ignition time, which confirms its usefulness as an octane number enhancer in gasoline. Blending of ethanol with diesel fuel also results in a reduction of soot mass concentrations[4]. Kitamura et al.[5] developed a chemical kinetic model (662 chemical species and 3005 elementary chemical reactions) used to analyze the suppression effects of oxygenated fuel blends (such as addition of ethanol to diesel fuel) on soot formation. In their study they represented diesel fuel with n-heptane which has a very similar cetane number. Their results show that addition of oxygenated fuels suppresses soot by reducing the amount of aromatic precursors (such as acetylene) that lead to drastic suppression of PAH / soot formation. They also found that adding oxygenates so that total oxygen content of the fuel is increased to 14 % by mass yields nearly soot-free combustion, which also agrees with published experimental work[4,6] .

In the present study, the burning and sooting behavior of isolated ethanol droplets in a spherically-symmetric condition was analyzed. The spherically-symmetric burning of an isolated droplet, produced under microgravity conditions, is a dynamic problem that involves the coupling of chemical reactions, multi-phase flow (liquid, gas, particulate) with phase change. To this end, microgravity droplet combustion serves as an ideal platform for advancing the understanding of the physics of diffusion flames for liquid hydrocarbon fuels and additives that are typically used in internal

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combustion engines and gas turbines. It is also evident from previous work of the authors[7], that a thorough interpretation of droplet burning behavior cannot be accomplished without examining and incorporating the influences of sooting and radiation on droplet combustion.

Ethanol droplet combustion has been extensively studied by researchers[8-14] using reduced-buoyancy techniques. In his classical investigation, Godsave[8] reported the burning of suspended ethanol droplet combustion. Kumagai and coworkers[9,10] studied ethanol combustion using droptower facilities to measure burning rates and flame diameters of ethanol for various initial droplet diameters and investigate the importance of relative velocity of moving droplets. Lee and Law[11] studied combustion of small, freely-falling methanol and ethanol droplets in which they reported droplet burning histories and time resolved bulk liquid-phase water mass fractions. Ethanol was also one of the primary fuels studied aboard the STS-94/MSL-1 Shuttle mission in the Fiber-Supported Droplet Combustion-2 (FSDC-2) program[12]. In those studies, the burning rate, flame diameter measurements, and extinction behavior for droplets ranging from 2.5 to 6 mm were investigated. In all of the studies mentioned above, experiments were performed for ethanol droplets burning in atmospheric pressure. Consequently, there was no observation of sooting in those experiments.

Soot formation in ethanol droplet combustion was first observed by Yap[13] in his experiments using freely-falling droplets in a high-pressure drop-tube. In 2001, Urban et al.[14] observed the formation of a sootshell for ethanol droplets burning in pressures of 2 atm in the NASA 2.2 sec. droptower. However, soot concentrations were not measured in these experiments due to a lack of appropriate diagnostic equipment.

In the present study, we describe new experiments on the burning characteristics of isolated ethanol droplets and the environmental conditions leading to soot formation and luminous radiation. Experiments were performed by varying the pressure from 1 to 2.2 atm, the oxygen concentrations from 21% to 50%, and the initial diameters from 1mm to 2.5 mm.

Experimental Descriptions

The central component of the experimental apparatus (fig. 1) is the 12 liter stainless steel combustion chamber which contains the fuel delivery system, droplet generator, and the ignition assembly. During the experiment in microgravity, the fuel droplets are generated using two opposed hypodermic needles of 0.25 mm diameter that are separated by 0.5 mm. Fuel is pumped through the needles by a 1.0 mL solenoid-activated syringe attached to each needle. Each hypodermic needle is attached to a separate rotating galvanometric device. The dispensed fuel forms a liquid bridge and the rapid rotation of the needles in opposite direction deposits the droplet onto a long 15 μm diameter SiC fiber. The fiber is used to fix the location of the droplet and prevent the droplet from moving out of the field of view. The liquid fuel droplet is ignited using two horizontally opposed hot-wire igniters. The laser backlit images are obtained using an expanded and collimated 635 nm, variable-intensity diode laser. The diode laser is attached to a single-mode fiber optic cable and is expanded to 50 mm diameter. The expanded and collimated beam is directed through the top optical port of the combustion chamber using a front reflecting 75 mm diameter mirror positioned at 45^o. The optical port is fitted with a 50 mm diameter quartz window treated with a broad-band anti-reflection coating. The beam is transmitted through the combustion chamber and then focused using a second 75 mm mirror positioned at 45^o. The reflected beam is then imaged through a spatial filter to a high-resolution CCD camera located on the bottom optical plate. A 105mm f/1.8 camera lens is used to obtain the magnification required to spatially resolve the droplet and the soot containing region. An image quality interference filter of wavelength 635 nm with a full-width, half-max bandwidth of 10 nm and an absorption neutral density filter of optical density of 3.0 are placed directly in front of the camera lens to eliminate flame emission.

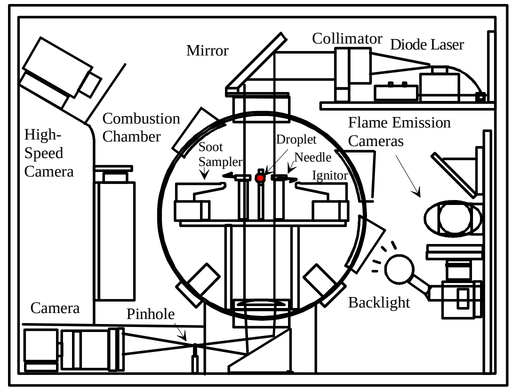

Figure 1 Schematic of the experimental apparatus

Experiments were performed at both the NASA Glenn 2.2 sec. drop tower and the JAMIC 10 sec. dropshaft. Detailed description of the two facilities can be found elsewhere[15,16].

Soot Volume Fraction Measurement

The soot volume fraction within the region bounded by the droplet surface and the flame were measured using a full-field light extinction technique and subsequent tomographic inversion using 3-pt Abel transforms[17]. The intensity distributions were measured by digitizing the backlit images with a high-resolution frame acquisition and processing board and a custom image processing algorithm. The images were filtered using a linear, 3- by 3 pixel mean filter. The intensity ratio distributions were calculated by dividing the gray-level values for the sooting image along the line of analysis by corresponding intensities measured for the background image (which was captured prior to droplet ignition and therefore unattenuated by soot). The intensity ratios were then averaged using a moving 5-pt operator. The projected light-extinction ratio distributions obtained as a function of time were used to determine the soot volume fraction, f_v, using a 3-pt Abel deconvolution technique[17], with soot optical property determined using the light-extinction/gravimetric calibration technique[18].

Droplet Burning Rate

Using the digitized images of the laser back-lit droplet, a custom software was used to determine a graylevel threshold to distinguish the droplet from the background. The burning rates were obtained from a linear fit to the evolution of the square of the droplet diameter with time after the transient heat-up period.

Effect of Pressure:

Figure 2 displays the measured burning rate for ethanol droplets of approximately 1.9 mm at pressures ranging from 1.0 to 2.2 atm in 21 % oxygen concentration in nitrogen. The results indicate that for the pressure range investigated, the droplet burning rate is independent of pressure. It is important to note that all of these experiments exhibited soot-free burning. In previous experiments[19] using n-heptane droplets burning in atmospheric and sub-atmospheric pressures, a significant variation in the droplet burning rate was observed. However, the variation in the burning rate in the n-heptane experiments were attributed to the reduction in the sooting propensity at the lower pressures.

Effect of Oxygen Concentration:

Flame temperature has a very strong dependence on the ambient oxygen concentration. The simple d^2 law for droplet burning rate, K, predicts that the increase in the oxygen concentration will produce an increase in the flame temperature, effective thermo-physical properties of the gas-phase bounded by the droplet and the flame front, and the transfer number, B:

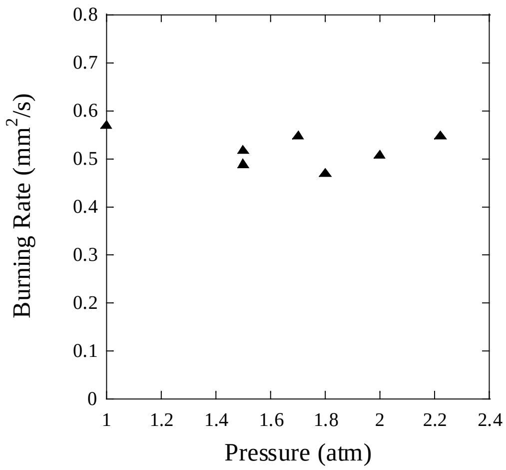

Figure 2 Droplet burning rate vs. ambient pressure

$$K = 8\frac{D_g\rho_g}{\rho_l}\ln(1+B) \qquad \ldots(1)$$

$$B = \frac{C_p(T_\infty - T_s) + \Delta h_c/\upsilon}{h_{fg}} \qquad \ldots(2)$$

$D_g = \alpha = \lambda_g/C_{pg}\rho_g$ based on the unity Lewis number assumption where α is thermal diffusivity, λ is the thermal conductivity, ρ is density, C_p is specific heat at constant pressure, υ is kinematic viscosity, Δh_c is heat of combustion and h_{fg} is latent heat of vaporization. Subscript (s,g) denotes surface conditions at the gas-phase side. The increase in oxygen concentration will also cause the flame to reside closer to the droplet surface, thereby effectively enhancing the rate of heat conduction and hence, higher burning rates. This behavior can be clearly observed in figure 3 that displays the burning rate for droplets of approximately 1.8 mm in diameter burning in 1 atm air.

Effect of Initial Diameter:

Figure 4 displays the measured burning rate as a function of initial droplet diameter ranging from 1 mm to 3 mm burning in air at 1 atm pressure. These results indicate that the burning rate decreases with increases in the initial droplet diameter. These experiments also displayed soot-free burning. Therefore, the reduction in the burning rate is likely caused by the increased radiative heat losses which scales as the cube of the droplet diameter[20].

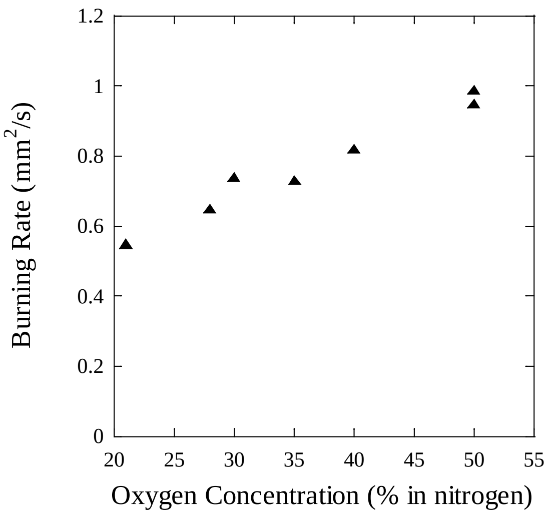

Fig. 3 Burning rate of ethanol droplets as a function of oxygen concentration

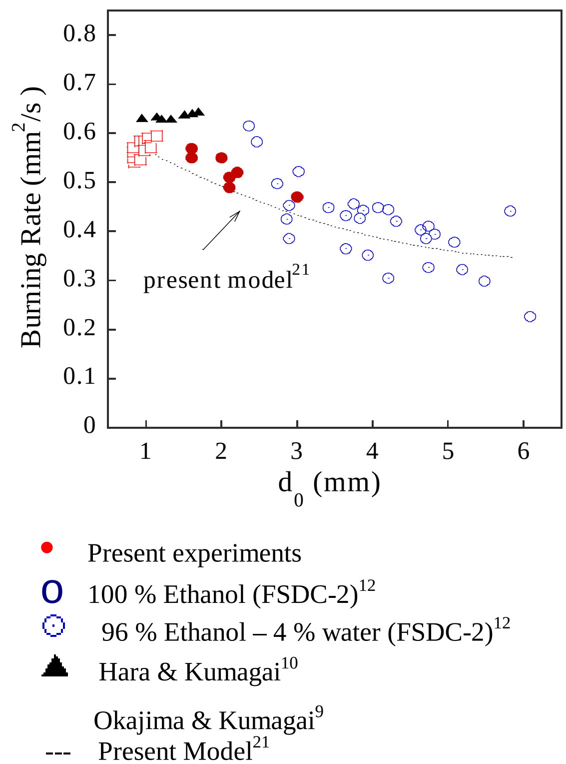

Fig. 4 Burning rate of ethanol droplets as a function of initial droplet diameter

Although classical theories of droplet combustion neglect the influence of radiative heat losses, it can influence the burning behavior for larger droplets. Due to the small droplet sizes with respect to the radiating volume of gases (resulting in small view factors), radiative heat produced from the high-temperature gases reduce the flame temperature without appreciable absorption of the radiation by the liquid droplet. Radiant emission measured using a broadband radiometer indicates there is a measurable increase in the radiative heat loss as the initial diameter is increased from 1.0 mm to 3 mm.

Similar experimental observations were made for ethanol experiments using a larger range of initial droplet sizes. Marchese et al.[20] performed experiments with large ethanol droplet as part of the Fiber-Supported Droplet Combustion-2 experiment[12]. In these experiments, droplets ranging in size from 1.5 mm to 6 mm were studied. Results shown in figure 4 display a significant reduction in burning rate as initial diameter was increased. This behavior was believed to be caused by the influence of non-luminous radiative heat losses that become more pronounced at larger droplet sizes. For larger droplets between 5 mm and 6 mm burning rate drops to values as low as 0.2 mm^2/s.

The data from the present experiments fill an important void for the comparison and validation of the numerical model. The experimental data are in good agreement with the trend predicted by the numerical model of Kazakov et al.[21] that incorporated radiative heat losses associated with gas-phase components.

Observations of Sooting

For non-premixed flames, increases in oxygen concentration will also result in greater degrees of soot formation owing to the sensitivity of the endothermic pyrolysis reactions[22]. Figure 5 displays the backlit view image obtained for 1.9 mm droplets burning in 1 atm at elevated oxygen concentrations. These experiments indicate that while the rate of burning is increased due to the higher temperatures, conditions leading to sooting behavior was not attained.

In earlier studies, increases in the ambient pressure resulted in the observation of higher luminosity (freely-falling experiments of Yap[13]) and the formation of a sootshell (microgravity experiments of Urban et al.[14]) for ethanol droplets. Figure 6 displays the backlit view of ethanol droplets of 1.7 mm diameter burning in air at pressures ranging from 1.0 to 2.2 atm. The lack of the presence of a sootshell is clearly observed in the 1.0 and 1.5 atm experiments with slight attenuation caused by small concentrations of soot in the 2.2 atm experiment. The present interpretation[14,23] is that increased pressures lead to increased decomposition of ethanol to ethylene and water, one of the two decomposition channels of ethanol[24,25] ,

$C_2H_5OH \rightarrow C_2H_4 + H_2O$ Reaction 1

Abstraction reactions produce vinyl radicals from the ethylene that are then converted to acetylene via:

$C_2H_3 \rightarrow C_2H_2 + H$ Reaction 2

Reaction 2 is very sensitive to ambient pressure at conditions relevant to the present experiments. Acetylene is generally accepted as a key species contributing to soot formation processes. Increased levels of acetylene promote aromatic hydrocarbon formation either through the C_4 mechanism of Frenklach and co-workers[26] or through subsequent formation of C_3 species followed by the C_3 ring formation mechanism of Miller and Melius[27].

In an effort to increase the likelihood of forming soot in ethanol experiments, ambient pressure and the oxygen concentration were varied in conjunction. Figure 7 displays the backlit laser-backlit view of ethanol droplets burning in various oxygen concentrations in nitrogen at 2.2 atm. At 21 % and 25 % O_2 in N_2, there is no visible luminosity exhibited in the flame view and the attenuation of the laser beam in the backlit view was lacking. As the oxygen concentration is increased to 30% O_2 in N_2, the formation of a distinct sootshell and a luminous flame are observed. Another interesting behavior was noted in which the sooting propensity appears to decrease at 40 % O_2 in N_2 case compared to the 30% O_2 in N_2 case. Additional experiments and analysis are required to investigate this interesting behavior. From the experiments shown in figure 7, the maximum soot volume fraction, $f_{v,max}$, was measured using the tomographic inversion technique. These measurements clearly bear out the interpretation from the visual observation – at 21% O_2 in N_2, there is no measurable soot concentration, while at 30% O_2 in N_2, the maximum soot volume fraction is approximately 13 ppm. The soot volume fraction was not measured for the 40% O_2 in N_2 case since the distribution of soot was not uniform. These measurements represent the first soot volume fraction data obtained for ethanol droplet combustion in microgravity environment. These experiments clearly demonstrate the strong dependence of sooting behavior of ethanol droplets on ambient pressure and oxygen concentration.

Concluding Remarks

This study provided the first detailed measurement of the spherically-symmetric burning and sooting behavior of isolated ethanol droplets burning in enhanced oxygen and high pressure conditions. The burning rate measurements are strongly influenced by ambient oxygen concentrations (21% to 50% O_2 in N_2) but are independent of pressure in the range studied (1.0 to 2.2 atm in air). Use of enhanced oxygen concentration combined with higher pressures resulted in distinct sootshell formation. Measurement of soot volume fraction indicates that the sooting propensity increases non-monotonically with oxygen concentration. The effective control of the sooting behavior of ethanol from a soot-free flame to a highly sooting flame by using pressure and oxygen concentration is important for its use as one of the primary fuels to investigate the influence of sooting and radiation influence on droplet combustion.

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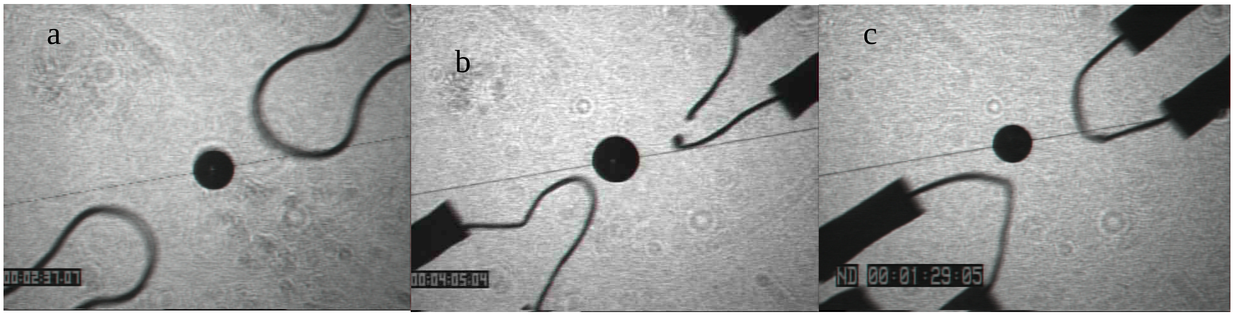

Fig. 5 Ethanol droplets burning at 1 atm and enhanced oxygen concentration a) 21 % b)35 % c) 50%

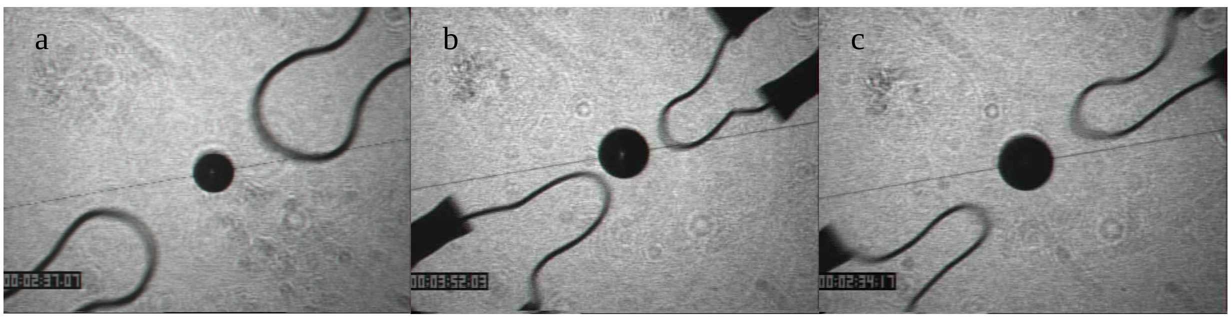

Fig. 6 Ethanol droplets burning at 21 % oxygen in nitrogen and elevated pressure a) 1 atm b)1.5 atm c)2.2 atm

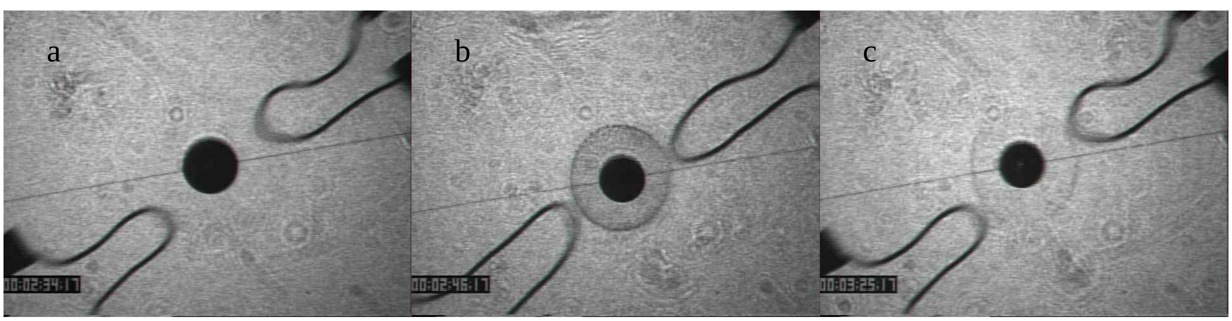

Fig 7 Ethanol droplets burning at elevated pressure and enhanced oxygen concentration a) 2.2 atm 21 % O_2 b)2.2 atm 30 % O_2 c) 2.2 atm 40 % O_2

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