Bradley D. Urban[1], Kenneth Kroenlein[1], Andrei Kazakov[1] and Frederick L. Dryer[1], Ahmet Yozgatligil[2] and Mun Young Choi[2], Samuel L. Manzello[3], Kyeong Ook Lee[4], Ritsu Dobashi[5]

Sooting Behavior of Ethanol Droplet Combustion at Elevated Pressures under Microgravity Conditions

Liquid ethanol is widely used in practical fuels as a means to extend petroleum-derived resources or as a fuel additive to reduce emissions of carbon monoxide from spark ignition engines. Recent research has also suggested that ethanol and other oxygenates could be added to diesel fuel to reduce particulate emissions. In this cursory study, the combustion of small ethanol droplets in microgravity environments was observed to investigate diffusion flame characteristics at higher ambient pressures and at various oxygen indices, all with nitrogen as the diluent species. At the NASA Glenn Research Center 2.2-second drop tower, free ethanol droplets were ignited in the Droplet Combustion Experiment (DCE) apparatus, and backlit and flame view data were collected to evaluate flame position and burning rate. Profuse sooting was noted above 3 atm ambient pressure. In experiments performed at the Japan Microgravity Center 10-second (JAMIC) drop shaft with Sooting Effects in Droplet Combustion (SEDC) apparatus, the first data that displayed a spherical sootshell for ethanol droplet combustion was obtained. Because of the strong sensitivity of soot formation to small changes in an easily accessible range of pressures, ethanol appears to be a simple liquid fuel suitable for fundamental studies of soot formation effects on spherical diffusion flames. The results impact discussions regarding the mechanism of particulate reduction by ethanol addition to fuels in high-pressure practical combustors.

[1] Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544

[2] Department of Mechanical and Engineering Mechanics, Drexel University, Philadelphia, PA 19104

[3] Building and Fire Research Laboratory, NIST, Gaithersburg, MD 20899

[4] Center for Transportation Research, Argonne National Laboratory, Argonne, IL 60439

[5] Department of Chemical Engineering, University of Tokyo, Tokyo, Japan

Mail address:
Mr. Kenneth Kroenlein, Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544, USA, Phone: (609) 258-0316, Email: kennethk@Princeton.EDU

Paper submitted: 09.04.2002
Submission of final revised version: 08.01.2004
Paper accepted: 07.07.2004

Introduction

Understanding ethanol combustion processes has significant practical as well as fundamental scientific implications. Ethanol has been used as a motor fuel since the development of automobiles because it can be derived from biomass. Currently, ethanol contributes approximately 1% of the total highway fuel market in the United States [1]. As a supplemental source to fossil-derived fuels, biomass-derived ethanol substitution has the potential to reduce net CO_2 release to the atmosphere.

In recent years, the use of ethanol as a fuel additive has also been stimulated by the Clean Air Act Amendments of 1990 [2, 3] that require utilization of reformulated and oxygenated gasoline to reduce carbon monoxide and volatile organic compound emissions from spark ignition engines. Although there is considerable controversy as to whether oxygenate addition actually produces the claimed reduction in gaseous emissions in more modern engine designs, its use is relatively benign in terms of air emissions and groundwater contamination. On the other hand, the most common oxygenate additive to gasoline, methyl tertiary butyl ether (MTBE), is highly soluble in groundwater and has caused notable groundwater contamination. Health concerns have been raised by both the ground water contamination problem, and by other, less documented air pollutant-related health effects. Increased use of ethanol may occur as MTBE use declines. Ethanol addition to spark ignition engine fuels can also be used to modify fuel road octane number.

Recent research has also suggested that oxygenates, used either as additives or as a substitutes for petroleum-derived distillates [4, 5], can reduce particulate emissions from that found with diesel engines operating on petroleum-derived distillate resources alone. A proposed alternative, to produce alkylated distillate fuels for diesel applications, also appears to reduce particulate emissions, but the required generation of hydrogen results in additional refining and the same combustion production of CO_2 (in comparison to utilizing oxygenates generated from biomass). The increased hydrogen production required for alkylation also impacts petroleum refining costs. Ethanol has been among several oxygenates considered in this research.

In addition to conventional internal combustion engine applications, there are possibilities for using ethanol as a relatively benign hydrogen source through reformation, or as a direct fuel for use in fuel cell power generation [1]. Finally, NASA has also raised consideration of ethanol as a low polluting liquid propellant for rocket propulsion applications [6].

This paper is principally interested in the sooting characteristics of ethanol combustion. Several fundamental works on the sooting characteristics of premixed ethanol/oxidizer systems exist. For example, Street and Thomas [7] have observed soot production in pre-mixed ethanol/air flames at atmospheric pressure, but at much larger fuel-air ratios than those typically needed to initiate sooting with paraffin hydrocarbons. Frenklach and Yuan [8] report that both methanol and ethanol suppress shock-initiated soot formation of premixed mixtures. In their shock tube experiments, Alexiou and Williams [9] report that addition of ethanol to toluene at low concentrations actually causes an initial increase in soot yield. However, further increase in ethanol concentration eventually resulted in a decrease in observed soot yield until sufficient ethanol was added to suppress the measured soot volume fraction entirely.

Most practical combustion applications involve diffusive/mixing limited combustion situations. Based upon visual observations in atmospheric pressure air, the commonly held assumption is that ethanol diffusion flames do not produce noticeable amounts of soot. These notions are further amplified by fundamental isolated droplet combustion studies that presently appear in the literature [10-16]. Even recent observations in microgravity studies of isolated ethanol and ethanol/4% water droplet combustion (in air at 1 atm) conducted as part of the Fiber Supported Droplet Combustion - 2 (FSDC-2) experiments aboard the STS-94 shuttle mission in 1997 suggest that ethanol diffusion flames produce only non-luminous radiation. In other ground based studies, consistent with the notion established by the above, a decrease in soot production due to addition of methanol, ethanol, and other oxygenates to benzene, toluene and other fuels is well-documented [8, 9].

However, in these situations, sooting tendency alteration can be caused by several factors, e.g., changes in oxygen level in fuel-rich regions, dilution of sooting components in the raw fuel composition, reduction in peak combustion temperatures, and production of key species that affect soot formation chemistry. A complete understanding of these results remains to be achieved [17]. A majority of fundamental droplet combustion studies on ethanol have had as their principal interest the potential condensation and dissolution of water from the combustion into the remaining fuel droplet, and/or the effects of azeotropic properties on droplet combustion. However, from the work discussed below, and seemingly contrary to the tendency for ethanol to reduce soot production in diesel engine combustion [18], it appears that ethanol itself is prone to producing soot in droplet diffusion flames at higher ambient pressures and/or increased oxygen indices. The apparent first observations of this fact are cursory isolated, freely falling droplet combustion experiments (in post combustion gases of pre-mixed flames) reported by Yap [19]. Yap noted that as ambient pressure is increased from one to three atmospheres, isolated droplets of ethanol burning in air become highly luminous. Sooting, the source of the increased flame luminosity was noted to be so profuse at 3 atmospheres that the droplet surface could no longer be observed under backlit conditions. Only visual observations were reported, with no other supporting experimental data. No fundamental explanations for these observations were speculated and these results had not been pursued in the literature until these initial observations were corroborated by a recent study conducted by these authors [20]. Not only do these observations contribute to the discussion of the mechanisms through which ethanol addition to fuels reduces soot emissions, but in fundamental terms, this work suggests that ethanol is a favorable candidate as a fuel to study the effects of sooting on the combustion of isolated droplets in microgravity combustion.

The spherically symmetric burning of an isolated droplet in microgravity is a dynamic problem that involves the coupling of chemical reactions, multi-phase flow (liquid, gas, and 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 combustion engines and gas turbines. Based upon other recent work involving the authors, a thorough interpretation of droplet burning behavior cannot be accomplished without examining and incorporating the influences of sooting and radiation on droplet burning properties. Concurrently, isolated droplet combustion studies offer an opportunity to investigate sooting phenomena on the dynamics of diffusion flames, and over parameter ranges not available in quasi-steady experiments such as annular jet diffusion flames.

Today, advances in transient, detailed numerical modeling of isolated droplet burning, combined with the sphero-symmetric experiments conducted under microgravity conditions, make droplet experiments a one-dimensional diffusion flame research tool of similar importance to the commonly used laminar premixed flame problem. The relative simplicity of the numerical modeling task (in comparison to multi-dimensional transient systems) affords an ability to include richly detailed sub-models that are developed and tested against fundamental experiments and parametrically studied numerically. These abilities lead not only to improved opportunities to better investigate the sensitivities of various levels of sub-models proposed, but to an environment in which detailed and reduced model performance can be compared. The modern paradigm connecting these fundamental research studies to applications is through the production of accurate reduced sub-model components that are reasonable for use in demanding computational problems and associated multi-dimensional design tools. These design tools significantly impact the experimental development and optimization of practical energy conversion devices that maximize efficiency under the constraints of minimal pollutant emissions.

Based upon the cursory observations of Yap [19] and the authors' past investigation [20], we have come to conclude that ethanol offers an ideal liquid fuel candidate for investigating the effects of sooting in isolated droplet combustion configurations. Ethanol represents one of the simplest liquid fuels with properties similar to those of practical liquid fuels. Its gas-phase oxidation chemistry is relatively simple and reasonably established [21,22]. Recent detailed, transient numerical modeling of sphero-symmetric combustion of ethanol under non-sooting conditions has been very successful; results of this work are discussed and compared against existing experimental data elsewhere [16]. The extension of the detailed model to include sooting effects is presently in progress. Thus, we have set about the task of confirming and expanding the observations of Yap.

Here we report new experimental results concerning soot production characteristics of isolated ethanol droplets in microgravity in the 2.2-second drop tower at the NASA Glenn Research Center in Cleveland, Ohio and in the 10-second drop shaft at Japan Microgravity Center (JAMIC) tower in Hokkaido, Japan. The qualitative measurements reported below firmly establish the strong pressure and oxygen sensitivity of sooting in the case of pure ethanol isolated droplet burning, a fact not previously documented in the literature, provide an initial set of parameters for the more quantitative work, and are of immediate significance to other researchers working on droplet combustion phenomena and sooting of oxygenated fuels. More detailed measurements by the authors to generate quantitative data that can be used in model development and validation for sooting effects on droplet burning based upon ethanol combustion properties are reported elsewhere [23,24].

Experiments and Observations

2.2-second drop tower studies:

In the 2.2-second Tower at NASA Glenn, free, isolated droplet experiments were conducted in the DCE combustion apparatus that is described in detail elsewhere [25]. After microgravity conditions are obtained, an untethered droplet was formed and deployed by opposed hypodermic needles attached to stepper motors. Kanthal hot wire igniters provided ignition energy to opposite sides of the droplet and are retracted after ignition. Untethered droplets of approximately 1.5 mm initial diameter were investigated to assure that sooting characteristics were not perturbed by fiber support [26]. Both the backlit and direct luminosity flame images were used to characterize the droplet flames and qualitatively assess extent of soot production. In Figs. 1 through 4, the images labeled 'a' are backlit images. The combination of backlight and camera aperture used in this experiment was difficult to optimize, as the sooting tendency of the

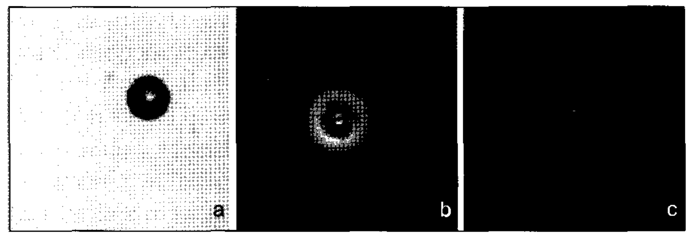

Fig. 1: 2.2 second drop tower ethanol droplet backlit view and flame view just after ignition and at steady state in 1 atm air.

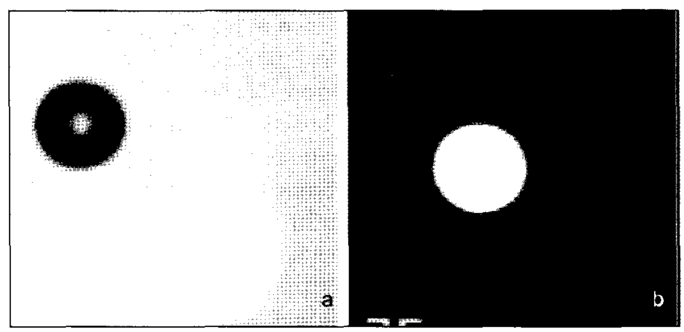

Fig. 2: 2.2 second drop tower backlit and flame images of an ethanol droplet in 1.5 atm air.

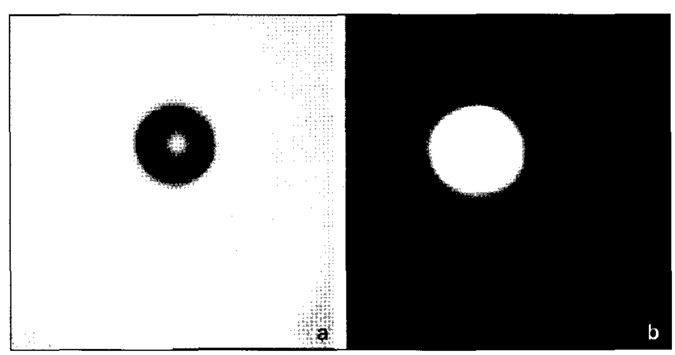

Fig. 3: 2.2 second drop tower backlit and flame images of an ethanol droplet in 2 atm air.

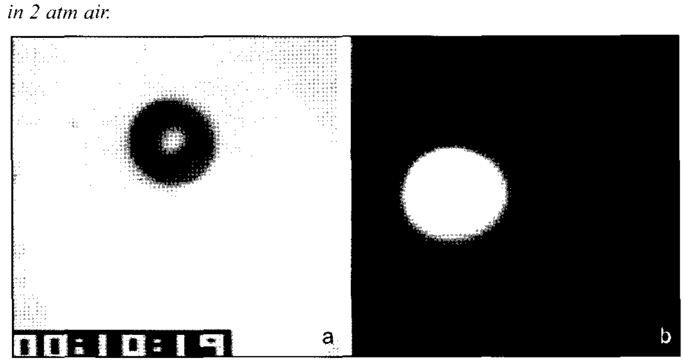

Fig. 4: 2.2 second drop tower backlit and flame images of an ethanol droplet in 3 atm air.

observations to be recorded were not known *a priori*. The field of view of the reported images was approximately 7 mm. Images labeled 'b' or 'c' show direct images of flame luminescence, with minimal artificial illumination. The luminescence results from a combination of chemi-luminescence and continuum soot radiation. The fields of view for these images are much larger than backlit images, on the order of 25 mm.

The observations made in the 2.2-second tower correspond to the initial transient period of burning of the free-floating isolated droplet, before quasi-steady state burning can be fully achieved. The total observation times for droplet burning in the present experiments was only about 1.5 seconds, compared to estimated droplet burn times for complete combustion of up to 20 seconds. As the droplet vaporizes during growth and deployment, a region of stratified fuel vapor and air surrounding the droplet is formed. Although some soot particulates are created in this rich stratified region during ignition, this residual soot fraction may be burned away during the subsequent flame development and transition to diffusive burning. For example, Fig. 1a is a backlit view of a 1.5 mm ethanol droplet burned in 1 atm air and while Fig. 1b is a flame view image, both taken just after ignition. Fig. 1b displays a bright luminous radiation. However, after approximately one second of burn time, evidence of sooting is no longer present in either the flame or droplet views (Figs. 1a and 1c).

By increasing the pressure to 1.5 atmospheres (Fig. 2), a lasting presence of soot after the transition to diffusive burning is observed, indicating that the diffusion flame itself was producing soot. Nearly 1 second after ignition, soot particulate matter was just barely visible around the backlit droplet and the flame burns with bright yellow luminescence. Fig. 3 is for a droplet ignited in 2 atm air. In this experiment, the flame luminescence was much more intense than for the 1.5 atm pressure case. For a droplet burned in air at 3 atm, shown in Fig.4, the flame luminescence was strong enough to obscure the backlit

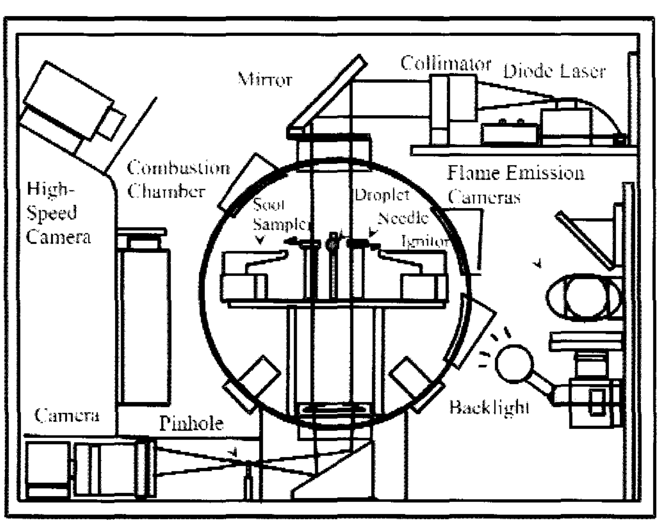

Fig. 5: Schematic of microgravity droplet combustion experimental apparatus.

image of the droplet and produce over exposure of the flame image. Over the range of pressures studied, an obvious gradual increase in soot production with pressure is evident.

JAMIC 10-second drop shaft studies:

The early experimental packages used in the NASA 2.2-second facility were not designed to quantitatively measure sooting characteristics of isolated burning droplets [13,27,28]. In 1996, Choi and Lee [29] implemented a full-field light extinction technique to measure soot concentration distributions surrounding isolated droplets burning in microgravity conditions. The experimental apparatus used in the JAMIC facility was a modified version of the rig used earlier [29, 30]. Figure 5 is a schematic of the experimental rig. The central component of the experimental apparatus is the 12-liter stainless steel combustion chamber that contains the fuel delivery system, droplet generator, and the ignition assembly. The fuel droplets were generated using two opposed hypodermic needles of 0.25 mm diameter that are separated by 0.5 mm prior to the initiation of the experiment. Fuel was pumped through the needles by a 1.0 ml solenoid-activated syringe attached to each needle. Each hypodermic needle was attached to a separate rotating galvanometric device. The dispensed fuel formed a liquid bridge and the rapid rotation of the needles in opposite direction deposited the droplet onto a 15-μm SiC fiber. The fiber was used to fix the location of the droplet and prevent the droplet from moving out of the field of view. The small dimension of the fiber was such that modification to the burning and sooting behavior was not present. The liquid fuel droplet was ignited using two horizontally opposed hot-wire igniters.

Laser backlit images were obtained using a 635 nm variable-intensity diode laser. The diode laser was attached to a single-mode fiber optic cable and was expanded to 50 mm diameter. The expanded and collimated beam was directed through the top optical port of the combustion chamber using a front reflecting 75 mm diameter mirror positioned at 45°. The optical port was fitted with a 50 mm diameter quartz window treated with a broad anti-reflection coating. The beam was transmitted through the combustion chamber and then focused using a second 75 mm mirror positioned at 45°. The reflected beam was then imaged through a spatial filter to a high-resolution CCD camera located on the bottom optical plate. A 105 mm f/1.8 camera lens was used to obtain the magnification required to spatially resolve the droplet and the region containing soot. An image quality interference filter of wavelength 635 nm with a half-bandwidth (full width at half maximum of transmission peak, FWHM) of 10 nm and an absorption neutral density filter of optical density of 3.0 were placed directly in front of the camera lens to eliminate flame emission.

Figure 6a displays the backlit view of a 3 mm ethanol droplet burning in air at a pressure of 1.8 atm. Figure 7a is the flame emission view obtained for the same experiment at the same time. Notice that there is no attenuation of the laser-intensity (in Figure 6a) or continuum luminosity from the flame (Figure 7a), and therefore no evidence of sooting. Only faint non-luminous radiation is observed in Figure 7a. In Figures 6b and 7b, the pressure was increased to 2 atm and the oxygen concentration in nitrogen was increased to 0.25 mole fraction of oxygen in nitrogen for a 2.8 mm droplet. The presence of the sootshell is clearly evident. The corresponding luminosity of the flame (Figure 7b) was significantly higher than that observed for the non-sooting case (Figure 7a). These experiments represent the first observations of sooting resulting in the formation of a spherical sootshell for ethanol droplets burning in microgravity conditions. In Figures 6c and 7c, the oxygen concentration was increased to 0.31 mole fraction of oxygen in nitrogen while the pressure was maintained at 2 atm for a 2.9 mm diameter droplet. The sootshell appears more opaque (corresponding to higher soot concentration) and the luminosity of the flame is markedly brighter. The increase in sooting is due to the expected higher flame temperatures as the oxygen concentration is increased. Measurements of the flame luminosity and soot volume fractions can be found elsewhere [23,24].

Discussion and Conclusions

The above data are the first to clearly document that isolated burning droplets of ethanol can be made to soot profusely by small increases in ambient pressure. The sooting behavior is a function not only of pressure, but also of oxygen index and the inert in the mixture. These properties can be used as controls in ethanol droplet experiments to achieve sooting and non-sooting conditions. These parameter variations are within a range that is easily achieved in microgravity experiments.

Two types of soot production can be distinguished in the isolated droplet combustion experiments reported here. Soot is formed during the ignition process as the reaction propagates through the stratified fuel-oxidizer mixture initially surrounding the droplet and transition to diffusive flame structure occurs. The addition of energy from the igniter source to the rich regions of this initial gas-phase stratified fuel/oxidizer mixture leads to rapid fuel pyrolysis followed by the observed soot formation. This is particularly evident in the 2.2-second tower experiments at 1 atm pressure, where yellow flame luminescence is observed for a short period immediately after the ignition. Shortly after the initial appearance of continuous-spectrum radiation, the flame color becomes blue and luminosity is reduced, indicating the complete burnout of the initially formed soot. Similar soot formation properties after ignition have been observed for *n*-heptane droplets at atmospheric pressure. On the other hand, this behavior was not observed for methanol where the flame appeared blue during the entire ignition and subsequent burning periods. These results are consistent with the early observations of Street and Thomas [7] who reported soot

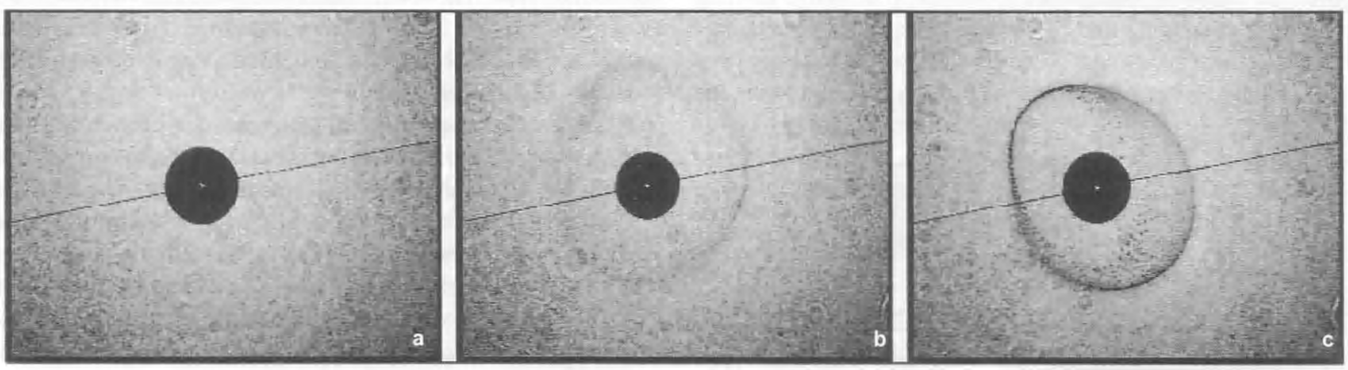

Fig. 6: Backlit view of 2.6-2.9mm initial diameter ethanol droplets in a) 1.8 atm, 21% oxygen, b) 2 atm, 25% oxygen, c) 2 atm, 30% oxygen.

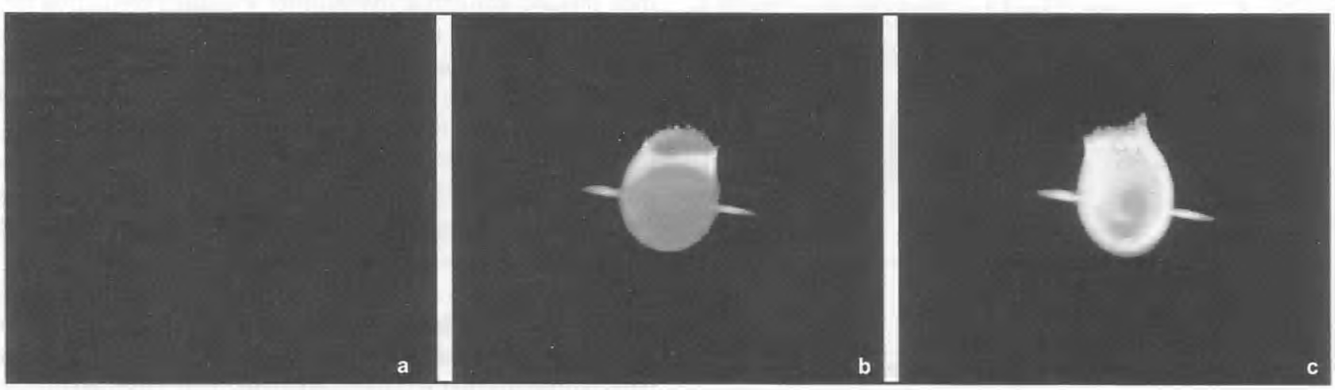

Fig. 7: Flame luminosity images for the cases in Fig. 6.el

formation upon ignition in the premixed ethanol/air system starting from the equivalence ratio of about 1.5, but that methanol "... gives no yellow color, even in diffusion flames".

In some cases, soot is also formed in the sustained diffusion flame structure. For instance, *n*-heptane droplets usually exhibit diffusion flame soot production that results in the formation of dense soot shells inside the flame structure. This soot production mechanism can be eliminated by reducing the ambient pressure to below 0.25 atm; however, the increased vaporization of *n*-heptane droplets prior to ignition at this pressure results in significantly enhanced soot formation from stratified burning before the development of the diffusion flame. For ethanol droplet combustion, droplets ignited at 1 atm will burn without sooting. However, an increase in ambient pressure above 1 atmosphere or increased oxygen index leads to more substantial production of soot precursors, consequently promoting soot particle inception rates to the level necessary for the sooting characteristics to be observed.

While the process requires much more extensive investigation to determine the reason(s) for these variations of sooting with combustion parameters, the present experiments identify that ethanol is an excellent fuel candidate to facilitate the study of these issues. Small changes in pressure and oxygen index that are easily accessible experimentally lead to a full range of soot characteristics including non-sooting and heavily sooting cases in which soot shells are observed. In addition, the fact that ethanol has considerable potential to soot in diffusive/mixing limited combustion environments at high pressure impacts consideration of the mechanisms through which addition of ethanol to hydrocarbon fuels might result in reduced particulate emissions from practical energy conversion systems.

Acknowledgements

The authors to wish to thank Joe Carrion, John Easton, Vedha Nayagam, and Paul Ferkul at NASA Glenn Research Center for helpful discussions and their support of experimental operations of the 2.2-second facilities. Support from JSUP (Mr. T. Sakuraya) and the assistance from the JAMIC staff (Dr. M. Yamaoka, Director) is gratefully acknowledged. This work was supported by NASA through Grant NCC3-822 (MYC) and a subcontract of this grant to Princeton University (FLD).

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