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COMBUSTION BEHAVIOR OF FREE BORON SLURRY DROPLETS

by

F. Takahashi, F.L. Dryer, and F.A. Williams

Department of Mechanical and Aerospace Engineering Report 1702 AFOSR TR No. (Applied For)

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This paper reports first observations of the combustion properties of isolated boron/JP-10 slurry droplets in high temerature, atmospheric pressure oxidizing streams under low Reynolds number conditions. Slurry droplets of initial diameter between 400 and 500 micrometers and initial solid mass fraction of 0.3 were studied using single-lens reflex and high speed cine photography under both self illuminated and backlighted conditions. During the experiments, the mole fraction of oxygen in the surrounding high temperature environment (ca. 1900K) was varied between 0.07 and 0.39.

Boron slurry droplets burned for short periods of time with an envelope diffusion flame structure, but then experienced violent disruption for all cases studied. The intensity of the disruption process was found to be strongly influenced by the temperature of the envelope diffusion flame. The ignition of the boron particles emitted from the initial fuel droplet was also affected by this flame temperature. As this flame temperature was increased by increasing the environmental oxygen content. the disruption occurred at earlier times in the vapor phase burning period. Ignition of the boron particles at high flame tempera-tures (> 2500K) was accompanied by a (popping) sound and a bright greenish flash of luminosity. The secondary atomization of the primary slurry droplet yielded much smaller solid agglomerates (< 50 microns) which were consumed very rapidly. The spontaneous atomization phenomena may be related to the presence of solids in this slurry as well as to the liquid phase composition. The paper discusses several possible effects which may precipitate these phenomena.

Abstract

Introduction

The potential advantages of boron slurry as a high energy density fuel for air breathing propulsion systems have stimulated numerous research and development efforts to understand and improve slurry combustion properties (Burdette, 1978; Faeth, 1984:. To realize these potential advantages will require very efficient atomization of the slurry, accompanied by rapid ignition and burnout of the boron with full condensation of the combustion products, (Faeth, 1984). A number of earlier works have considered the ignition and combustion properties of boron itself (Talley, 1959; Macek and Semple, 1969, 1971; Macek, 1973; King, 1972, 1974, 1982; Mohan and Williams, 1972, Gremyachkin et al., 1980: Safaneev et al., 1981; Gaponenko et al., 1981; Glassman et al., 1982), primarily as single boron particles or clouds of particles in a surrounding oxygen rich environment. Although many guestions on the fundamental elements of boron particle combustion remain, the formulation of boron particle slurries as a mechanism to introduce boron into the combustion chamber adds new complications to the problem. Studies on both coal-oil (eg., Miyasaka and Law, 1980) and carbon black slurries (Szekely and Faeth, 1982a, 1982b,: Szekely et al., 1984) have demonstrated that as slurry droplets burn, a large agglomerate containing nearly all of the solid particles within the initial droplet is formed. This large agglomerate, which requires considerably more time to burnout than single particles of the initial suspension size, creates serious combustion inefficiencies in compact sized combustors and may also result in enhanced erosion problems. The ignition of boron agglomerates has been studied earlier by Chevchuk et al. (1975), while more recently, the ignition, combustion and burnout of boron agglomerates has been studied by Turns et al. (1984). In both cases these agglomerates were essentially collections of dry particles. It is clear that if the agglomeration process could be reduced by some process which led to spontaneous atomization of the primary slurry droplet during the vapor phase combustion of the liquid phase hydrocarbon, combustion characteristics might be significantly improved. Yet little data on even the general overall combustion properties of the slurries themselves are available.

The principal purpose of the current ongoing research is to investigate the fundamental combustion properties of isolated boron slurry droplets to yield information useful for fuel and combustor developmental efforts. This paper reports the first preliminary observations made on the combustion properties of boron/JP-10 isolated slurry droplets.

Experimental Technique

The experimental apparatus (Fig. 1) consists of a droplet generation system and an inverted flat flame combustion system. The droplet generation system utilizes an "aerodynamic" technique first proposed by Bolt and Saad (1957) and later modified by Green et al. (1983) to produce a stream of monodisperse slurry droplets. The technique was specifically developed (Green et al. 1983) for the generation of droplets of very viscous fuels, and has been further modified such that a primary stream of monodisperse boron slurry droplets containing up to 50 % solids (by weight) ranging in initial size 300 to 1000 microns and separated by more than 300 diameters can be produced.

The droplet generator is composed of a hypodermic needle (.838 mm Id.), which has a fine wire (0.076 mm dia. by 3 mm length) soldered at the apex of a chamfered tip. The needle is placed concentrically inside a glass converging nozzle (1.2 mm throat diameter) such that the wire tip resides at the throat position. Nitrogen is flowed through the concentric passage between the needle and the glass tube walls.

The fuel slurry is volumetrically pumped at low flow rates (less than 8 ml/hr.) through the hypodermic needle and forms a small droplet on the tip wire which is removed by aerodynamic drag forces. This procedure forms a very regularly spaced stream of monodisperse slurry droplets in the surrounding nitrogen flow.

Broplet diameter and droplet velocity, can be varied by varying the volumetric fuel flow rate and the nitrogen flow rate. Typical values for the droplet studies reported here were 30wt% solids, 435 microns initial diameter, 2.2 meters per second initial droplet velocity and 60 diameter droplet spacing. The nitrogen stream containing the monodisperse droplets is injected coaxially through the center of an inverted premixed flat flame burner.

The flat flame burner consists of a rectangular $(70 \times 70 \text{ mm})$ water cooled sintered bronze flame holder followed by a rectangular $(84 \times 84 \text{ mm})$ combustion duct with quartz windows on all sides. The burner is inverted, and therefore, both injected droplets and post combustion gases flow downward at nearly equal velocities. In the current work, the burner was operated at atmospheric pressure on a fuel lean mixture of methane, oxygen, and nitrogen as shown in Table I. The equivalence ratio of the mixture was varied for different experiments between 0.3 and 0.7 The adiabatic flame temperature (Tr) and the mole fractions of major combustion products (X_i) for the various conditions were calculated using the NASA CEC Program of Gordon and McBride To achieve these various equivalence ratios, the methane (1971). flow rate was held constant and the oxygen/nitrogen ratio was varied. Through this approach, it was possible to widely vary the oxygen mole fraction in the post combustion gases from 0.07 to 0.39 without significant variation in either the flame tempera ture or the post flame gas velocity.

Figure 2 shows the experimental axial temperature distribu-

tion along the trajectory of the droplets. These measurements were obtained using a silica coated Pt-6% Rh/ Pt-30% Rh thermocouple (wire diameter, 0.05 mm, bead diameter, 0.15 mm). Radiation corrections were performed using the techniques described by Friedman (1953) and the emissivity of the coated thermocouple (0.22) recommended by Kaskan (1957). It should be noted that the temperature histories along the droplet trajectory are nearly the same for all results reported in this paper.

Figure 3 shows typical cross-sectional temperature distributions at several axial locations. These temperature profiles show cross-sectional uniformity for all regions except those very near the burner surface.

Figure 4 shows the axial variations in gas velocity which result from the axial temperature variation. These velocities were calculated from the known mass flow rate of gases provided to the burner and the burner axial temperature distribution.

Both single-lens reflex and cinematographic observations of the isolated droplets were made using either the luminescence from the combustion region or a backlighting source. A laserscattering-triggered stroboscopic backlighting technique (Dryer and Kennedy, 1984; Marrone et al., 1984) was used to obtain well focused 35 mm single lens reflex photomicrographs. High speed (5000 frames per second) 16 mm cinematography strips were obtained to analyze the disruptive droplet burning behavior (described later).

Laser scattering from two beams placed in the droplet trajectory (1.9 mm between beams) was used to determine the velocity of the isolated droplets at various locations. Using the measured velocities, the calculated gas velocities, and the gas properties, the local droplet Reynolds numbers were found to never exceed 4.

The boron JP-10 slurry fuels used in these experiments were obtained from several manufacturers (Sun Tech, Inc., UOP, Inc., Atlantic Research Corp.) and the exact chemical constituents were proprietary, particularly those associated with the type of boron and the additives used to stabilize the slurries. The slurries, as delivered, contained different weights of solids. All slurries were diluted with pure JP-10 (exo-tetrahydrodi cyclopentadiene) to a boron mass fraction of 0.3. Unless otherwise noted, the results reported here were generated using the diluted product obtained from Sun Tech, Inc.

Results

Direct time exposure (1/30th second) photographs of isolated (separation distance = 2.70 centimeters), burning slurry droplets are shown in Figs. 5(a)-5(c). Slurry droplet combustion is seen to take place in two distinct stages: a vapor phase quiescent burning phase which is terminated by droplet disruption, followed by a particle/agglomerate burnout phase. Ignition of the injected slurry droplet occurs in about 7 to 11 milliseconds of exposure to the post flame combustion gases. A quiescent burning stage follows during which part of the liquid phase of the slurry is preferentially vaporized. A yellow luminosity is associated with the diffusive combustion of the vaporized JP-10, probably as a result of formation of soot. This luminosity is recorded as a streak along the droplet trajectory, and it is seen in Figs. 5(a)-5(c) that the period of quiescent burning is a function of the amount of oxygen in the post flame gas mixture. By counting timing marks on the high-speed movie film, the quiescent burning times for mixtures 1, 3, and 5 in Table I were found to be approximately 40, 80 and 120 milliseconds, respectively.

The quiescent burning is terminated by a violent disruption before all of the liquid phase has vaporized. Some of the remaining droplet mass (which contains all of the particles in the initial droplet) is dispersed into a larger volume of oxidizing media as small fragments containing both liquid and boron particles, while a portion of the remaining droplet continues along the original droplet trajectory.

Figs 6(a)-6(c) are direct time exposure photographs of the region of droplet disruption. In addition to a "popping" sound, a greenish-white bright flash results, particularly for higher oxygen concentration (gas mixtures 1-3, Table I). The green color of the emission is associated with the presence of excited boric oxide (Johns, 1961), and the accompanying noise may be indicative of rapid heat release from boron combustion. The intensity of the disruption in terms of both audible noise and light emission, decreases as the post flame oxygen concentration decreases, and is no longer present for conditions of gas mixture 5. In the case of gas mixture 4, the appearance of the greenish light emission is sporadic and very weak in intensity.

For high oxygen concentrations, there are three distinct characteristics associated with time exposure photographs of the disruption phenomena: the region of bright greenish white emission, a number of yellow streaks emanating from this region, and a major yellow streak continuing in the general direction of the original droplet trajectory. Figure 7 shows sequential frames of direct lighted high speed cinematography taken during the disruptive phenomena for a slurry droplet exposed to gas mixture 1. Consecutive frames represent an observation time change of approximately 200 microseconds for the first six frames (t=0-1ms)or 1 millisecond for the last twelve frames (t=1-13ms), with the time (t) noted in the figure corresponding to the time elapsed after droplet disruption for the first frame.

In the first frame of the sequence (t=0), the luminosity from the quiescent burning condition is seen. In later frames of the sequence (t=0.2-0.6 mg), the luminous region expands nearly symmetrically at a velocity of approximately 5-14 meters/sec. The brightness of the luminous region, appeared in subsequent frames (t>0.8 ms), prohibits observation of any detailed structure in this region. A number of luminous regions, which generate the fine yellow streaks in the time exposure photographs presented earlier (Fig. 6) are ejected from the highly luminous core. Other luminous regions suddenly appear at remote locations separated from the bright core. This observation indicates that fragments ejected from the primary slurry droplet can be extinguished by the disruption and later re-ignite (see Fig. 6(a)).

At 4 to 5 milliseconds after disruption, an asymmetric, spinning, luminous region comparable in size to that for the initial burning slurry droplet proceeds out of the highly luminous core with a trajectory similar to that of the initial slurry droplet.

Figure 8 shows a stroboscopic backlighted still photograph of a burning boron slurry droplet for which disruption has just begun. Approximately 50 micron fragments have been ejected from the droplet surface. The eruption appears to be from internal vaporization of some of the JP-10 liquid. It is likely that fuel vapor is projected away from the primary droplet along with the droplet fragment (see Fig 8, t=0-0.4 msec.). Very small boron particles and agglomerates (less than 10 microns) are ignited in the diffusion flame region and result in the bright greenish luminosity.

Slurry droplets with low boron loading (mass fraction less than 0.1) produce very weak disruptive behavior while pure JP-10 burn quiescently, except for a flash extinction which occurs at the termination of combustion. The disruptive behavior found for more heavily loaded boron slurry droplets (from Sun Tech) was also noted for heavily loaded slurries from Atlantic Research and UOP. However, the disruptions occurred at different exposure times than those noted for the Sun Tech product. Thus the disruptive behavior appears to be a characteristic of slurries themselves rather than of a particular manufacturer.

Discussion

The observed disruption appears to result from entrapment the liquid components inside the droplet with subsequent of superheating prior to vaporization. Since a large rotating droplet usually emerges from the region of disruption, it appears that the site of the disruption is near the surface of the primary slurry droplet rather than deep in the interior. A number of different effects may contribute to this behavior. One possibility is that the presence of boron does not play an es-Less volatile additives with high Lewis numbers sential role. inside the liquid fuel may form a thin shell with an elevated surface temperature during vaporization, leading to homogeneous nucleation of the fuel inside: the nucleation site will be near the surface if the shell is thin and if the liquid temperature is The boron seems likely to modify this appreciably nonuniform. well-known mechanism of binary liquid mixtures by providing internal heterogeneous nucleation sites on slurry particles, thereby inducing earlier and less intense disruption. An alternative role of less volatile additives is that they may be heated to an extent at the droplet surface such that chemical interactions cause physical binding and agglomeration of boron particles at the surface. A relatively nonporous boron shell may thus develop, inhibiting vaporization, bringing the flame closer to the surface, and conducting heat into the interior, thereby promoting pressure buildup through internal vaporization that results in disruption. It is possible that this resistive boron shell would develop even for the burning of a sufficiently highly loaded slurry of a neat fuel without additives. Further study is needed to ascertain the relative importance of these various possibilities.

Figure 9 shows a typical axial variation in the droplet diameter (d), the droplet velocity (Ud), the gas velocity (Us), and the droplet Reynolds number,

$$Red = : Ud - Ug : d / v_s .$$

The kinematic viscosity of the gas mixture (v_g) was calculated using Wilke's equation (Wilke, 1950). It is seen that there is no significant variation in any of these parameters over the entire quiescent burning period. The constancy of d is consistent with an assumption introduced in the vaporization and combustion theories of Antaki (1985). The value of Red is small enough for nearly spherical symmetry to be expected during vaporization and quiesent combustion. Therefore the analysis of Antaki (1985) may be considered in addressing disruption mechanism.

The stoichiometric adiabatic flame temperature (T_f) of JP-10 fuel for the various gas mixture parameters shown in Table 1 were calculated using the NASA CEC Program (Gordan and McBride, 1971) and are shown in Figure 10. The composition of the gas mixtures at the measured gas temperature were also calculated, and the minor species were neglected by including them in the calculated mole fraction for N₂. The location where the disruption occurs along each droplet trajectory is shown for each of the gas mixtures in Figure 10. It is clearly seen that the disruption occurs at earlier times as the droplet diffusion-flame temperature increases. It is suspected that this is a result of increased heat flux to the droplet surface as the flame temperature increases).

The results of Antaki (1985) give formulas for the heatup time (t_h) of a slurry droplet prior to achieving the vaporization temperature at the surface and the subsequent burning time (try needed for complete consumption of the liquid fuel. The value of th may be compared with the experimental ignition time (t_i) , and (t_h+t_b) may be compared with the experimental time to disruption (td), to see whether disruption occurs at a constant fraction of the total theoretical lifetime. Although uncertainties in gas and droplet properties introduce appreciable uncertainities in the theoretical times th and t_b , the predicted variations in these values for different gas mixtures are relatively insensitive to the uncertainties. The predicted values of t_h vary little for the different gas mixtures, and the experimental values of t_i also vary little; the calculated th is within about a factor of 2 of the obser-The theoretical values of to also vary little, ved t₁. being about 15% smaller for gas mixture 1 of Table I than for gas This arises mainly because of the relatively weak mixture 5. (logarithmic) dependence of the burning rate on the stoichiometric adiabatic flame temperature. The previously cited variation in to is much larger, roughly a factor of 5 different for gas mixtures 1 and 5. Thus the disruption clearly does not occur at a constant fraction of the burning time. An increase in Tf substantially decreases the percent of liquid-fuel combustion prior to disruption. This result suggests that heat conduction through agglomerated boron plays a role in the disruption mechanism.

Previous work has shown that a boron particle covered by a shell of oxide coating several tens of angstroms thick typically ignites at temperature near 1900 K, and that the minimum ignition temperature is relatively independent of the ambient oxygen concentration (Macek, 1969; Faeth, 1984). In the present experiment, the post-flame combustion product gas temperatures for all gas mixtures studied was less than 1900 K everywhere. It is only because boron particles ejected from the primary slurry droplet are exposed to the higher temperatures produced by the JP-10diffusion-flame that boron particle ignition occurs. Therefore. the diffusion flame temperature at disruption is important to the ignition of the boron particles. In the cases of gas mixtures 1-3 of Table I, the JP-10 diffusion flame temperatures at disruption exceed both the boiling point of B_2O_3 (2316 K) and the melting point of boron (2450 K). For gas mixture 5, the flame temperature is much lower than these values, and in the case of mixture 4, the flame temperature is nearly equal to these tempe-Thus the observation of no green emission in the case ratures. mixture 4 is consistent with suggesting that the boron oxide of coating be rapidly removed from the boron particle for ignition particle to occur. The extinguishment temperatures of of the burning boron particles predicted by King (1982) are 1770 K (7.5% O_2 , d= 4 microns), 1680K (15% O_2 , d=4 microns), 1300 K (15%02, d=10 microns). These values are comparable with or lower than the post flame product temperatures for mixtures 1-3. Moreover, the burning times of single boron particles with these sizes are between 3 and 8 milliseconds (King, 1982). These times and behavior are consistent with the period of time that the bright flash occurs at disruption.

Concluding Remarks

Direct lighted and backlighted photographic observations oľ burning isolated boron slurry droplets have revealed that combustion occurs in two stages; a quiescent burning stage, followed by a disruptive burning stage. The temperature of the JP-10 diffusion flame surrounding the droplets during the quiescent burning stage plays a significant role in when the disruption occurs and whether the boron particles emitted from the parent droplet during disruption are ignited. As this flame temperature is increased (by increasing the ambient oxygen concentration), the droplet burning time previous to disruption decreases, apparently due to an increased heat flux to the droplet surface. 11 the flame temperature at the point of disruption is higher than the melting point of boron (or the boiling point of B2O3), ignition of boron particles takes place. The disruption of the primary boron slurry droplet appears to result in three types of fragments; individual boron particles, and/or very small agglomerates, a number of larger fragments, and a residual slurry droplet somewhat smaller in diameter than the initial slurry Apparently substantial liquid remains in the slurry droplet. droplet even after the disruption occurs, since fragment can undergo secondary disruptions.

These observations are in contrast to the speculations of

several previous investigators of boron slurry combustion who have suggested that the liquid more or less vaporizes entirely leaving behind a dry boron agglomerate containing nearly all of the boron particles in the initial slurry droplet. The secondary atomization of the boron slurry droplets and the liquid contained in the secondary fragments can substantially enhance the overall burning rate of the primary slurry droplet. More importantly, the disruptive burning phenomena appear to be characteristic of boron slurry fuels rather than a property of any one specific formulation. Additional research is underway to further clarify these issues.

Acknowledgments

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Gas Mix.	Flow Rate (l/min)				T _f	<u></u>			
No.	Ø	сн ₄	0 ₂	N ₂	(K)	x ₀₂	X _{N2}	х _{с02}	X_{H20}
1	0.3	10	66.7	43.3	2054	0.39	0.36	0.08	0.17
2	0.4	10	50.0	60.0	2065	0.25	0.50	0.08	0.17
3	0.5	10	40.0	70.0	2073 .	0.17	0.58	0.08	0.17
4	0.6	10	33.3	76.7	2078	0.11	0.64	0.08	0.17
5	0.7	10	28.6	81.4	2086	0.07	0.68	0.08	0.17

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TABLE 1 BURNER OPERATING CONDITIONS AND ESTIMATED

GAS MIXTURE COMPOSITIONS

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Figure Captions

Figure 1 Schematic of the Experimental Apparatus.

- Figure 2 Axial Variations in the Measured Gas Temperature Along Burner Centerline.
- Figure 3 Crossectional Variations in the Measured Gas Temperature (Gas Mixture No. 1, Table 1).
- Figure 4 Axial Variations in the Estimated Gas Velocity Along Burner Centerline.
- Figure 5 Direct Photographs of Free Burning Boron Slurry Droplets. $Y_B = 0.3$. Gas Mixture Conditions for Photographs: (a) 1, (b) 3, (c) 5 (Table I). Exposure time, 1/30 second.
- Figure 6 Disruption of Free Burning Boron Slurry Droplets. $Y_F = 0.3$. Gas Mixture Conditions for Photographs: (a) 1, (b) 3, (c) 5 (Table 1). Exposure time, 1/15 second.
- Figure 7 High Speed Direct Cinematographs of Disruption of Free Burning Boron Slurry Droplet. Y_B = 0.3. Gas Mixture Condition for Photographs is 1 (Table 1).
- Figure 8 A Stroboscopic Backlighted Photograph of a Disrupting Free Burning Boron Slurry Droplet. Gas Mixture Condition for Photograph, 1 (Table I)
- Figure 9 Axial Variations in the Droplet Velocity, Diameter, Reynolds Number and the Gas Velocity.
- Figure 10 Axial Variations in the Adiabatic Flame Temperature of JP-10 with the Ambient Gas Condition.









Figure 4





TAKAHASHI et c'. Figure 7



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Figure 1C



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