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Oxidizer coarse-to-fine ratio effect on microscale flame structure in a bimodal composite propellant

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ABSTRACT

The microscopic flame structure of a composite propellant is expected to change significantly as the particle size distribution is varied. In this paper we report observations of the diffusion flame structures in burning ammonium perchlorate (AP) composite propellants with varying ratios of coarse to fine AP. The coarse-to-fine ratio (C/F) was varied between 1:16 (mostly fine AP) to all coarse AP. Five kHz OH planar laser-induced fluorescence (PLIF) was used for *in situ* imaging of the highly transient and microscale flames above single and groups of AP particles at atmospheric and elevated pressures. Jet-like diffusion flames are observed for all propellants above coarse AP crystals at 1 atm. At elevated pressures, both jet-like and lifted arched inverted overventilated diffusion flames (IOF) were observed. Jet-like flames were seen more frequently for propellants with low C/F ratios, and were rarely seen for propellants with the highest C/F ratios. On the other hand, lifted IOF were seldom seen for the low C/F ratio propellants but were more frequently observed for higher C/F ratio propellants. Differences in the flame structures are postulated to be due, in part, to the dissimilar local burning rates and flame temperatures between the fine AP/binder matrix and coarse particles. For the first time, the diffusion flames from multiple coarse particles were observed to merge (group combustion), especially for higher C/F ratio propellants. As a consequence, flame height is affected by this group combustion of clustered coarse AP particles that behave similarly to a larger single particle. The ignition delay and lifetime for single AP particles in a composite propellant were measured at 1 atmosphere (atm). The particle lifetime and ignition delay for coarse AP particles vary as a function of C/F, indicating that nearby coarse particles can significantly affect coarse particle ignition and combustion. The data obtained is useful for comparison to numerical simulations of AP composite propellant combustion.

1. Introduction

Composite ammonium perchlorate (AP) propellants typically contain multimodal AP particle size distributions. The combination of coarse (100 s of microns) and fine (10 s of microns) particle sizes permits propellant burning rate tailorability, higher solid loadings, and improved propellant rheological properties during grain casting [1,2]. Near-surface flame structure is expected to be responsible for global propellant burning rate differences as the ratio of coarse to fine particles is varied. Recently, high-speed OH planar laser-induced fluorescence (PLIF) has been successfully used to image the final diffusion flame structures above individual AP particles in propellants with a 1:1 coarse-to-fine ratio (C/F) [3–7]. The 1:1 C/F was chosen in part to yield mostly isolated coarse particles. One could expect more interaction between coarse particle flames at higher C/F, and since many fielded propellants have higher C/F ratios, improved understanding of these interactions could lead to improved propellant modeling. Modelers have already incorporated the qualities of pseudo-premixed fine AP/binder matrix flames into computer models [8–12]; however, the formation of large final diffusion flames above adjoining particles (group combustion) has not been specifically investigated and has never been directly observed previously. Although group combustion is evident in some modern simulations [13], there is no flame structure experimental data as a function of C/F available for comparison.

Propellant global (strand) burning rates change with C/F, especially with increasing pressure [14–19]. A decreasing C/F (*i.e.* a larger percentage of fine AP) has been found to cause burning rate increases in experimental [15] and computational [17] studies. Though the ratio of coarse to fine particles is important, it does not solely control burning rate; the fine AP particle size is a significant contributor as well [18–20]. The fine AP/binder matrix can control burning rate even under conditions where the fine AP/binder matrix cannot sustain combustion on its own (that is, if the coarse AP crystals were removed). In

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these cases the coarse AP particles contribute to combustion by providing heat flux from primary diffusion flames (also called the leading edge flames (LEF)) and from the final diffusion flame [21].

The distance between the coarse particles, and the fine AP/binder matrix decomposition product temperature and composition affects the burning rate of the coarse particles. For example, it is theorized that as the C/F is decreased there is a greater distance between coarse particles on average. Also, the LEF over one coarse particle becomes increasingly isolated from other particles, leading to decreased interaction between coarse crystal flames. It has been postulated that if there are enough coarse particles on the surface at a high enough pressure the LEF will close over the fine AP/binder matrix and blanket the surface, resulting in a more plateaued (less sensitive to pressure) burning rate [19].

Examination of quenched propellant surfaces showed that coarse AP particles protrude or recess differently on the propellant surface depending on the pressure [22]. At low pressures the AP particles protrude above the surrounding binder and at higher pressures the particles are recessed in surface depressions [23,24]. The surface profile is primarily influenced by the relative burning rates of the AP and binder (which typically contains fine AP) at different temperatures and pressures. Surface features are shaped in part by the flame interactions between the pseudo-premixed flame over the fine AP/binder matrix, the LEF between the coarse AP particles and the binder, and the strongly pressure-dependent AP monopropellant flame.

Propellant diffusion flames have been investigated previously in an abstracted manner. Parr and Hanson-Parr have used 20 Hz OH, NH, and CN PLIF on sandwich configurations to study propellant oxidizer-fuel flames [25]. Flame structure above oxidizers such as AP, ammonium dinitramide (ADN), and cyclotrimethylene-trinitramine (RDX) were probed for quantitative species measurements [26,27]. Much can be learned from sandwich configurations, but they do not reflect the full dynamics of actual composite propellants. Recently, high-speed OH PLIF was successfully applied to flame structures above composite propellants over a range of pressures and for various binders and burning rate catalysts, but only one C/F was considered [3–6].

The objective of this study was to better understand how C/F affects the *in situ* flame structure by using high-speed OH PLIF to image the final diffusion flames in AP composite propellants.

2. Experimental methods

2.1. Propellant formulation

Propellants used in this study contained nominally 400 µm coarse AP (Firefox Enterprises) and 20 µm fine AP (Alliant Techsystems). The binder used was an R45-M prepolymer (Firefox Enterprises) with Tepanol HX-878 (3 M Corporation) as a bonding agent, icodecyl pelargonate (RCS RMC) as a plasticizer, and Desmodur E744 (Bayer Corporation) as a curative. The solids loading was held at 80% for all propellants. Ingredients and their percentages can be found in Table 1. The propellants are described by the percentage of coarse AP (% cAP) present in the mix. An increase in % cAP is equivalent to an increasing coarse-to-fine ratio.

All propellants but the 6% cAP propellant were mixed by hand. The 6% cAP propellant was mixed on a LabRam resonant mixer (Resodyn Acoustic Mixers, Inc.) to more adequately disperse the large amount of fine AP in the formulation [24]. The mixed propellants were cast into 6.35 mm diameter plastic molds 80 mm in length. After curing at room temperature for approximately one week the propellants were cut into strands 10 mm long for the experiments. Further details and particle size distributions can be found in Ref. [3]. In some cases, such as for the 35% cAP propellant, an additional propellant mix was used to check data trends. In these cases no statistical differences were found for flame heights, particle lifetimes, and ignition delays between propellant batches.

Table 1
Propellant formulation.

Propellant % cAP	Coarse-to-fine ratio	400 µm AP (wt%)	20 µm AP (wt%)
6	1:16	4.71	75.29
20	1:4	16	64
35	7:13	28	52
43	17:23	34	46
50	1:1	40	40
63	5:3	50	30
75	3:1	60	20
94	16:1	75.29	4.71
100	Monomodal	80	0

Note: For all C/F ratios, the binder formulation is:
R-45M: 14.58%.
Desmodur E744: 2.30%.
Isodecyl Pelargonate: 2.92%.
Tepanol HX-878: 0.20%.

2.2. Planar laser-induced fluorescence

High-speed OH planar laser-induced fluorescence (PLIF) was used to image flame structures and determine ignition delay, burning time, and lifetime for individual AP particles. A Sirah Credo dye laser was pumped by an Edgewave Nd:YAG (IS200-2-L) solid state laser. The Nd:YAG laser operated at 532 nm and pulsed at 5 kHz. The dye laser was tuned to output at the OH $Q_1(7)$ line at 283.2 nm. The pulse energy was measured to be approximately 0.55 mJ in the UV. Various lenses as described in Ref. [3], with the addition of a positive spherical lens, were used to focus the UV laser beam to a diagnostic sheet. The sheet created was 5.1 cm tall with a 50 µm waist thickness.

To capture the OH fluorescence emitted by the flame at 310 nm, a Nikon UV-grade lens (Nikkor 105 mm F/4.5) was mounted to a series of Semrock interference filters (FF01-300/80-25 and FF01-315/15-25) designed to block out interference from around the fluorescence wavelength and soot flame luminosity above about 700 nm. The filters were in turn mounted to a Video Scope International high speed image intensifier (VS4-1845HS). The intensifier is capable of operating at up to 100 kHz with a gain of up to 80,000. The intensifier was coupled to a Vision Research Phantom 7.3 camera, which can operate up to 6686 fps at full resolution of 800 × 600 with 14 bit image depth. A bellows was incorporated in the optical train, allowing for greater magnification and depth of field.

The propellant was burned either on a pedestal in the open atmosphere or inside a pressure vessel for burns at 0.5 MPa. Further details of the system can be found in Ref. [3].

Note that the OH PLIF results are qualitative. We are looking at the microscale flame structure above the propellant surface and not quantitative OH concentration with the techniques used.

2.3. Strand burns

All propellants were tested to determine global burning rates at pressures between 1 atm and 12.4 MPa. At 1 atm the propellants were burned in a fume hood, and at all other pressures the propellants were burned in a Crawford-type strand burner. The strand burner was pressurized with nitrogen and propellants were ignited with Nichrome wire. The Vision Research Phantom 7.3 was used to gather burning rate data at frame rates between 500 and 3000 fps. The location of the burning propellant surface was plotted against time, and the propellant burning rate was determined to be the slope of this line. At least two burns were performed for each propellant at each pressure.

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