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Near-surface flame structure characterization of simplified ammonium perchlorate/hydroxyl-terminated polybutadiene compositions

Sarah Isert[a,*], Terry L. Connell[b], Grant A. Risha[c], Trevor D. Hedman[d], Robert P. Lucht[a], Richard A. Yetter[c], Steven F. Son[a]

[a] Maurice J. Zucrow Laboratories, Purdue University, West Lafayette, IN 47906, USA
[b] Department of Mechanical and Nuclear Engineering, The Pennsylvania State University, University Park, PA 16870, USA
[c] Division of Business and Engineering, The Pennsylvania State University, Altoona College, Altoona, PA 16601, USA
[d] Naval Air Warfare Center Weapons Division, China Lake, CA 93555, USA

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ABSTRACT

Simplified model propellant configurations, such as monomodal propellants, can be valuable in the development and validation of predictive numerical tools. These idealized experiments also yield insight into the effect of diffusion length scales on combustion, but comprehensive data covering a large range of diffusional length scales do not currently exist. Here, monomodal propellants with ammonium perchlorate (AP) particle sizes under 800 μm and AP pellets ported and filled with hydroxyl-terminated polybutadiene (HTPB) were used to systematically study the effect of diffusion length scales, or AP equivalent particle sizes) of up to 4.1 mm on flame structure. In general, burning rates increased with pressure and decreasing particle size, as expected. Burning rates for samples with particle sizes greater than 400 μm converged with AP monopropellant burning rate data above approximately 2 MPa, the AP low-pressure deflagration limit (LPDL). For a given pressure above the LPDL, burning rates eventually became constant for both increasing and decreasing particle sizes. Conversely, for a given pressure below the LPDL burning rate was shown to be a function of particle diameter. Flame structures above the composite propellants were observed using 5 kHz OH planar laser-induced fluorescence (PLIF). The transient flames were underventilated (jet-like) over the AP particles at 1 atm while lifted, inverted, and overventilated at 5 atm. Distinct diffusion flame structures were observed visually above the ported samples at 1 atm. Very luminous flames were observed at the interface between the AP and binder. The effect of strain rate on sample combustion was examined using an opposed flow burner; at 1 atm, sample burning rate was not affected by strain rate. At the largest strain rate, the sample self-extinguished after igniter shutoff, indicating that secondary diffusion flames are important in the opposed flow configuration.

1. Introduction

There is a critical need for detailed measurements of ammonium perchlorate (AP) composites in simplified configurations for model development and validation. Models of composite propellants have made great strides in recent years; however, there is currently a lack of experimental data to fully support high fidelity models [1–3]. For example, due to constraints in computational power the most detailed AP composite models do not resolve the finest AP particles and therefore the fine AP and binder mixture is modeled as a homogeneous mixture, essentially as a subgrid model. However, the particle size at which the fine AP can actually be considered homogeneous with the binder – that is, at which it does not produce its own diffusion flame – varies with pressure. Consequently, monomodal data are needed to constrain these unresolved homogenized models, and simplified propellant experiments can help determine the parameter space where homogenization may not be safely assumed.

Other important areas that require experimental validation are individual AP particle burning rates, studying flame structures above the individual AP particles, investigating how AP particles burn together in group combustion, and learning how the flames above the individual AP particles may interact with each other. The application of 5 kHz OH planar laser-induced fluorescence (PLIF) allows flame structures to be directly observed in a propellant environment, providing new insight to the burning environment in a propellant.

Some previous work has examined simplified configurations. For example, “sandwiches”, or adjacent layers of pressed AP particles and fuel, have been considered under varied pressure environments

* Corresponding author. Fax: +1 765 494 0530.
E-mail address: sarah.isert@gmail.com (S. Isert).

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and have proven to be useful to model development [4–10]. Some monomodal AP composites have been studied to examine the overall effect of particle size on the burning rate [7,11,12]. However, these latter studies have not characterized the resulting flame structure or looked at a very wide range of AP particle sizes. In addition, combustion studies utilizing sandwich configurations also neglect the AP particle dynamics and other surface behavior phenomena. A more comprehensive study of the effect of particle size (or diffusion length scale) is needed for flame structure characterization.

Composite propellants containing a single AP nominal size distribution have been shown to exhibit a clear relationship between flame type, particle size, and global propellant burning rate [13–16]. For a specific pressure, monomodal propellant burning rates increase as AP particle sizes decrease, although this effect diminishes as particles become very small [11,12,14–16]. At the smaller particle sizes, the fine AP/binder matrix burns with a pseudo-premixed flame due to the intimate mixing of the fuel and oxidizer [2]. Burning rates are high as the flame is in close proximity to the propellant surface.

The assumption that the fine AP/binder matrix burns with a pseudo-premixed flame is used extensively in numerical propellant simulations as a way to enable meaningful burning rate predictions. It is not currently feasible to numerically resolve both the finest AP particles typically used in a composite propellant and the entire regressing surface. Resolution of the smallest particles requires multiple nodes over a distance of approximately 10 μm. Even in two dimensional simulations, resolution of the finest AP particles sizes becomes too computationally expensive to be a valuable predictive tool.

To overcome this computational expense, a certain AP diameter in current research is designated the “premixed limit” [3]. Particles below this diameter are assumed to be homogenized with the binder, and the fine AP and binder burn together with a pseudo-premixed flame. The premixed limit varies with pressure, accumulation of AP and binder on the surface, and solids loading [17]. Monomodal propellants can be used to model the fine AP/binder matrix (the finer AP and binder between coarse particles) and can be used to explore some of these issues.

Diffusion effects become more significant with increasing particle size and pressure. As particle size increases, the reaction zone occurs further above the propellant surface, resulting in diminished heat feedback to the surface and a decrease in burning rate. Detailed simulation is needed to know accurately where the premixed-to-diffusion transition occurs. Very large AP particles are considered to burn in the monopropellant flame limit, where the burning rate is dominated by the monopropellant flame instead of the diffusion flame. The monopropellant flame products and volatilized fuel mix and burn well above the propellant surface for very large AP particles.

The lower heat feedback from the diffusion flame and the presence of the relatively cooler monopropellant flame results in a slower burning rate that exhibits a relative insensitivity to particle size. Similar results are seen in sandwich burning experiments [6]. Global burning rates in general have been found to increase with pressure regardless of particle size [18], and it has been suggested that burning rates for propellants with large AP particle diameters are low at pressures below the low-pressure deflagration limit (LPDL) of AP as there is no strong AP monopropellant flame to contribute to the combustion [10].

The burning rates of pressed AP pellets from atmospheric [19–21] to pressures well above the AP LPDL [22] have been examined using a pressurized counterflow burner where the AP pellets were combusted with gaseous methane and ethylene. In these experiments the flame is strained, meaning an axial velocity gradient parallel to the diffusion flame is introduced by changing the gas flow velocities over a fixed separation distance. Under atmospheric pressure conditions, straining the diffusion flame (formed between the AP and opposing fuel flow) increased the pellet burning rate, while at higher pressure conditions the burning rate was unaffected by the strain rate [22]. The change in burning rate dependence with pressure was attributed to the pressure-dependent chemical kinetics of the AP combustion process moving the monopropellant flame toward the AP surface. When the monopropellant flame dominates the combustion, heat release occurs closer to the propellant surface, reducing the effect of the diffusion flame.

Accurate numerical prediction of global burning rate and flame-to-surface heat transfer requires knowledge of the microscale flame structures immediately above the propellant surface. The structure of an AP/fuel flame can be quite complex. For example, a solid AP pellet burning in an acetylene/ethylene/nitrogen opposed flow configuration results in a flame structure with several distinct regions: a monopropellant flame, the first formation of OH radicals, the primary diffusion flame, and a soot flame [22]. The luminous sooting flames observed above propellant sandwiches are described as occurring over the binder or where there are relatively wide areas of fuel and oxidizer [10,23–25]. This is expected, as the luminosity is caused largely by radiation from carbon formed from the burning fuel and oxidizer. Propellants with oxidizer to fuel (O/F) ratios closer to stoichiometric or where the oxidizer and binder are well-mixed exhibit fewer luminous flames due to the lowered soot levels.

The flame structure above individual AP particles in a propellant environment has also been examined using OH PLIF in sandwich, counterflow, hole, and propellant configurations [25–30]. At 1 atm this flame structure resembles that proposed by the BDP model. Jet-like underventilated diffusion flames, with very thick reaction zones, were seen centered over individual AP particles [26–30]. Underventilated flames occur when there is an excess of fuel present for combustion. In this case, the oxidizer jet from the coarse AP particle is issuing into a locally fuel-rich environment (caused by the fuel-rich combustion of the fine AP/binder matrix), resulting in an inverted underventilated diffusion flame.

The jet-like flame structure had previously been predicted to occur at elevated pressures for large particle sizes [13,31]; however, at elevated pressures a large volumetric oxidizer flux causes the diffusion flame to lift off the propellant surface and form a thin flame sheet [26,28,30]. The lifted flame sheets had not been simulated or predicted prior to being experimentally observed and were found to be aspects of arched flame structures with thin reaction sheets corresponding to the temporally and locally overventilated conditions [28]. At elevated pressures, the ratio of the individual coarse AP crystal and fine AP/binder matrix burning rates is high enough that the excess of oxidizer from the rapidly burning coarse AP crystals produces a locally overventilated condition [28].

In this study we quantify the propellant global burning rate as a function of AP particle size and pressures between 1 and 40 atm (0.1 and 4 MPa) to investigate how the premixed limit changes with pressure. We investigate how flame structure changes as a function of particle size and pressure to provide insight into global burning rate trends. In addition, we examine coarse AP particle lifetimes and burning rates to see how particle size affects flame structure and global burning rate. Finally, we study how global burning rate changes as a function of flame strain rate for ported AP pellets. It is hoped that the results from this study will provide validation for accurate predictive propellant combustion models.

2. Experimental systems and methods

Two methods were used to obtain the required diffusional length scales for this study. Monomodal composite AP propellants were used for particle sizes up to about 800 μm. Larger length scales were simulated using ported AP pellets, bridging the gap between commercially obtainable AP particle sizes and the infinite particle diameter limit. Burning rates and flame structures were examined for both the composite propellant and the ported pellet samples described in the sections that follow.

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