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Study of aluminum particle combustion in solid propellant plumes using digital in-line holography and imaging pyrometry

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ABSTRACT

The combustion of molten metals is an important area of study with applications ranging from solid aluminized rocket propellants to fireworks displays. This work uses digital in-line holography (DIH) to experimentally quantify the three-dimensional position, size, and velocity of aluminum particles during combustion of ammonium perchlorate (AP) based solid-rocket propellants. In addition, spatially resolved particle temperatures are simultaneously measured using two-color imaging pyrometry. To allow for fast characterization of the properties of tens of thousands of particles, automated data processing routines are proposed. Using these methods, statistics from aluminum particles with diameters ranging from 15 to 900 µm are collected at an ambient pressure of 83 kPa. In the first set of DIH experiments, increasing initial propellant temperature is shown to enhance the agglomeration of nascent aluminum at the burning surface, resulting in ejection of large molten aluminum particles into the exhaust plume. The resulting particle number and volume distributions are quantified. In the second set of simultaneous DIH and pyrometry experiments, particle size and velocity relationships as well as temperature statistics are explored. The average measured temperatures are found to be 2640 ± 282 K, which compares well with previous estimates of the range of particle and gas-phase temperatures. The novel methods proposed here represent new capabilities for simultaneous quantification of the joint size, velocity, and temperature statistics during the combustion of molten metal particles. The proposed techniques are expected to be useful for detailed performance assessment of metalized solid-rocket propellants.

1. Introduction

Solid propellants often contain aluminum particulates in order to increase specific impulse and improve combustion stability. When one surface of a propellant strand is ignited, a combustion front forms and the nanometer to micrometer-sized metal particulates begin to melt due to the heat released from the reaction of the oxidizer and fuel binder [1]. Individual molten particles tend to agglomerate together and begin to combust via an aluminum oxidation reaction forming larger spherical molten particles with oxide caps and flame zones as shown in Fig. 1. Drag forces produced by combustion at the surface eject these agglomerates into the surrounding gases. The size, velocity and temperature of these metal particulates are important quantities needed to inform combustion modeling [2] and to evaluate new propellant formulations [3,4]. Additionally, in accident scenarios, such as rocket motor launchpad failures where propellants burn near atmospheric pressure [5], knowledge of the solid particle sizes, velocities and temperatures is needed to understand potential hazards.

Aluminized propellants display complex interactions between the oxidizers, the metal particles, and the surrounding gas [6]. Some groups have attempted to isolate and study each of these constituents separately in controlled conditions [7,8]. A variety of optical techniques have been applied to measure the particle behavior including videography [1], shadowgraphy [9], Schlieren [10], phase Doppler anemometry [11], and laser diffraction [12]. As these previous works show, imaging techniques are particularly useful due to their ability to reveal spatial and temporal combustion dynamics.

Prior attempts to image and size the particles during a propellant burn have been limited by camera focal depth [1]. Traditional holography with photographic plates can overcome this problem [13] but requires tedious experiments and manual data processing. To reduce these complexities, we recently demonstrated digital in-line holography (DIH) for application to propellant combustion

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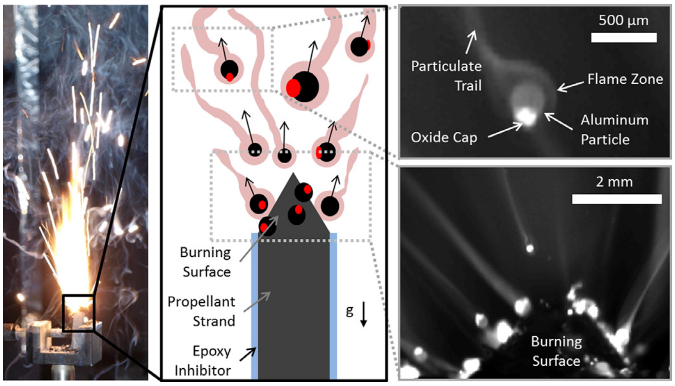

Fig. 1. Aluminum particle combustion and agglomeration are illustrated near the burning surface of a propellant strand. From left to right, the figure shows an experimental image of the propellant strand combustion, a schematic of the components, and two black-and-white images of the emission at 700 nm.

[14,15]. In this technique, hologram images, which contain diffraction patterns from particles along the line-of-sight, are recorded with digital sensors. Afterwards, the hologram images are numerically refocused along the optical depth in order to reveal in-focus images of particles at their original three-dimensional (3D) locations. By expanding the measurement volume across a large effective depth, this technique is particularly advantageous for rapid quantification of particle size statistics [14]. In addition, this technique allows for accurate sizing of out-of-focus particles, which increases the number of particles counted in each frame and provides the out-of-plane position for each particle [16,17]. Furthermore, when combined with double-framing or high-speed imaging, it is possible to measure the three-component velocities [15]. This work extends upon the initial demonstrations of propellant DIH provided in [14,15], with focus on the quantitative understanding of combustion behavior.

In addition, measurement of the thermodynamic state, via the particle temperature, is vital for accurate definition of combustion models [2,18]. Prior to this work, the temperature of the metal particulates [7,19,20] and the gas near solid propellants have been studied using single point spectroscopy [6,21,22], single-point pyrometry [23,24], and coherent anti-Stokes Raman spectroscopy (CARS) [25,26]. In the literature, there has been little focus on spatially resolved temperature measurements of aluminum particles in propellant flames [8]. For the current investigation, imaging pyrometry is a particularly attractive technique due to its ability to provide quantitative two-dimensional temperature maps, which are complementary to DIH. Pyrometry utilizes images taken at two or more wavelengths to determine the temperature of an object [27,28]. The object can be black body, gray body, or have a known emissivity as a function of wavelength. This technique is especially useful for measuring the temperature of multiple materials with different gray body emissivities and can be used to measure the temperature of metalized propellants by avoiding any narrow band emissions [29].

In this paper, we combine the capabilities of DIH with pyrometry in order to simultaneously measure the temperature, size, velocity and three-dimensional position of particles in a propellant sample burn. Unique analysis programs are developed to automatically process thousands of frames and quantify tens of thousands of particles. The work begins with a brief review of aluminized propellant combustion. This discussion helps to bound the expected conditions and motivates quantities of interest. Next, the experimental configuration for simultaneous DIH and pyrometry is illustrated. Calibration methods and characterization of instrument uncertainties are discussed. A first set of experiments show the capabilities of DIH for measuring particle size and for studying particle aggolmeration statistics as a function of initial propellant stick temperature. A second set of experiments creates a unique dataset exploring simultaneous measurement of position, size, velocity and temperature of aluminum agglomerates. When available, comparison with results from the literature, models, and previous measurements help to validate the proposed measurement techniques.

2. Aluminized propellant combustion

The aluminized propellants used in these experiments are composed of approximately 70 wt.% ammonium perchlorate (AP), 20 wt.% micrometer-size aluminum particulate, and 10 wt.% hydroxyl terminated polybutadiene binder (HTPB). When the propellant sample is ignited, the combustion occurs at a burn front that propagates through the material [30]. Ammonium perchlorate melts at 830 K and serves as the oxidizer, reacting in the condensed phase at the burn surface as well as in a gas-phase diffusion flame close to the surface [30]. The nominal adiabatic flame temperature of AP is $\sim$ 1200 K; however, the size and distribution of AP grains, the presence of burn rate modifiers [31] and processing methods can have a large effect on surface temperatures. For example, an increase in the AP grain size tends to cause larger diffusion flame standoff distances, lower surface temperatures, and lower burn rates [31–33].

The addition of aluminum particles (Al, melting near 930 K) and their aluminum oxide shells (Al_2O_3, melting near 2345 K) raise the flame temperature by 1100 K or more. In [25], we report CARS measurements of the gas-phase near the burning surface of one of the aluminized propellants investigated here. Gas-phase temperatures from 1000 K to over 3000 K are observed. Other groups have predicted maximum temperatures near 3800 K in regions with the highest Al_2O_3 concentration [8]. Due to this wide variation in gas-phase temperatures, along with expected complex spatial and temporal effects, particle temperatures are also expected to vary widely. Therefore, the proposed pyrometry system has been designed for temperature measurement between 1800 and 4000 K.

2.1. Agglomeration effects

Agglomeration and combustion of the Al particles provide further complexities. Individual Al particles, which have typical initial sizes between 5 and 40 µm [34], may melt together and combine on the burning surface to form large agglomerates. For combustion near atmospheric pressure, these agglomerates can be hundreds of micrometers in diameter and display a broad size distribution [35]. However, not all initial particles agglomerate, and the result is a complex bi-modal size distribution of particles ejected from the burning surface [30]. In addition, some particles may sinter together, without completely melting, giving rise to large coral-like structures. Finally, larger agglomerates may oxidize producing Al_2O_3 caps (vaporizing near 3240 K) and may themselves eject smaller particles ($\sim 1\mu m$) of Al_2O_3 as smoke [9,30].

Previous investigations of these phenomena indicate that a wide range of factors, such as propellant thermal layer temperature, AP grain size ratio, and aluminum particle distribution can affect the amount of agglomeration and the particle size-distribution [36,37]. Some relatively simple models predict the amount of agglomeration based on particle residence time in the AP flame zone [31], while more elaborate models use 3D particle packing to predict the size-distribution of agglomerates, which may form in pockets around large AP crystals [37]. Section 4.1 investigates these effects and provides additional experimental data.