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Comparison of Conventional and Resonant Acoustic Mixing of AP/HTPB Propellants

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Abstract

Composite solid propellants can be mixed using a variety of techniques. Recent R&D efforts have considered implementation of resonant acoustic mixing (RAM) strategies for improved homogeneity, reduced complexity, and faster mixing capabilities. In the current study, an 80% monomodal AP/HTPB baseline and a standard high-performance propellant formulation (85% trimodal AP, 1.3% Fe_2O_3, 1.0% Al_2O_3) were mixed by standard laboratory techniques (i.e., hand mixed) and RAM to allow for direct comparison of these methods. Propellant samples were burned in a constant-volume, optically accessible strand burner at pressures between 3.45 and 20.7 MPa (500–3,000 psia). Propellant microstructures were evaluated with standard scanning electron microscopy (SEM) techniques. Implementation of RAM yielded no effect for the 80% monomodal baseline and a significant increase in the propellant burning rate (~25%) in comparison to the standard hand-mixing strategy for the high-performance formulation. The observed performance improvement in the RAM formulation was attributed to improved catalyst/oxidizer contact, yielding improved catalysis during propellant combustion.

1. Introduction

Composite propellant mixing strategies are focused on homogeneity and ease of manufacturing (e.g., decreased material amount and cost, faster mixing time, reduced complexity, etc.). Resonant acoustic mixing (RAM) has the potential to improve propellant mixing in all these areas. Lab-scale experiments investigating vessel geometries and their effect on mix time and homogeneity of the final product have been reported in the literature [1-5]. Vessel geometry has been reported to have an effect on mixedness. More explicitly, the role of the vessel geometry decreases as the mix duration increases. Curved-bottom vessels eliminate the occurrence of dead mixing zones. Finally, mixing in 'end use' vessels has been shown to decrease material consumption and complexity. These studies indicate that RAM can be utilized to decrease manufacturing cost, time, and complexity.

Comparisons between accepted mixing methods and RAM have also been presented in the literature. Cross et al. [6] investigated Ammonium Perchlorate (AP)/Hydroxy-Terminated Caprolactone Ether (HTCE) propellants manufactured using a twin blade planetary mixer and RAM. Material and safety properties between batches were similar, with some temperature-dependent deviations. A modest increase in the burning rate of 7.9% for the RAM batch was reported at 10.34 MPa (1,500 psia). Zebregs et al. [7] investigated Ammonium Nitrate (AN)/Hydroxyl Terminated Polybutadiene (HTPB) propellants manufactured by mechanical mixing and RAM over a pressure range of 2-10 MPa (300-1,450 psia). The burning rates reported for the mechanically mixed propellants were very similar (within scatter) to those reported for the formulation mixed by RAM. Lastly, Smith et al. [8] conducted an experiment comparing planetary mixing and RAM to manufacture AP/HTPB propellants with aluminum and iron oxide (Fe_2O_3). The burning rates reported at 6.89 MPa (1,000 psia) for the planetary mixer and RAM formulations were 10.4 and 9.5 mm/s (0.41 and 0.37 in/s), respectively. The three studies discussed display disparate results in whether or not RAM mixing has an effect on global propellant ballistic properties (i.e., burning rates). Furthermore, the phenomena driving the observed changes in propellant properties were not elucidated in these studies.

The comparative studies above shown the need for more complex formulations to be investigated with RAM. In the current study, an 80% monomodal baseline and a high-performance (85% trimodal AP, 1.3% Fe_2O_3, 1.0% Al_2O_3) were mixed conventionally and also using RAM, and their ballistic properties were compared. The following section details the experimental methods used to mix the propellant formulations (conventionally and with RAM); conduct ballistic

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testing; and prepare microscopy imaging samples. The experimental methods are followed by a discussion of the ballistic results and imaging. Finally, a summary and key findings are detailed in the conclusion section.

2. Experimental Procedures

The two formulations evaluated herein are detailed in Table 1. These formulations include a baseline, plain AP formulation (80% monomodal AP) and a high-performance formulation containing 85% AP and metal-oxide catalysts (1.3% Fe_2O_3 and 1.0% Al_2O_3). The HTPB, AP, metal oxides (Fe_2O_3 and Al_2O_3), and IPDI used in the current study were acquired from FireFox Enterprises, American Pacific (AMPAC), FireFox Enterprises/Alfa Aesar, and Millipore Sigma, respectively. The smallest-size AP (2 μm) was manufactured by an in-house, wet-milling procedure previously developed by the authors [9]. A description of the AP milling process is discussed briefly for completion. Ninety μm AP, milling media, and hexane were placed into a jacketed temperature vessel that is compatible with a LabRAM II and chiller (POLYSTAT 3C15++). The AP was milled for 6.5 hours at various forces. After milling, large particles were sieved out using the LabRAM, and smaller particles were collected and used herein.

Table 1. Matrix of the propellant formulations investigated in the current study.

Formulation	1	2	3	4
Mixing Method	Conventional	RAM	Conventional	RAM
Solids Loading	80%		87.3%	
AP size (μm) (w/%)	200 (80%)		400 (35%) 90 (20%) 2 (30%)	
Catalyst (w/%)	None		Fe_2O_3 (1.3%) Al_2O_3 (1.0%)	
HTPB (w/%)	18.25%		9.31%	
Plasticizer (w/%)	None		DOA (2.5%)	
IPDI (w/%)	1.75%		0.89%	

2.1 Conventional Mixing

Conventional mixing methods have been optimized by the authors through several decades of manufacturing propellants and are well documented in the literature [10-19]. This process is detailed briefly herein for completion. Liquid ingredients (i.e., HTPB and/or DOA) were added to a beaker and mixed for 10-15 mins. Solid ingredients (i.e., AP, metals, and/or metal oxides) were then incrementally added in increasing particle size order (e.g, smallest to largest). The solid ingredients were each mixed in for 10-15 mins to form a propellant paste. The propellant paste was vacuumed between the addition of each solid ingredient. Lastly, the curative (isophorone diisocyanate, IPDI) was added to the propellant paste to cure the HTPB. The propellants were extruded into ¼" Teflon tubing and cured at 63 °C for 1 week. The mass and length of each propellant sample were taken prior to ballistic testing.

2.2 Resonant Acoustic Mixing

Propellant ingredients were placed in a glass vial, starting with the largest to smallest solid ingredients, followed by the liquid ingredients. The glass vial was mixed in a LabRAM II at 100 g's for 30 mins. The curative (IPDI) was added and initially mixed in with a stirring rod, followed by another 15-minute cycle in the LabRAM II. The formulation was heated and vacuumed after mixing for two cycles of 5 and 15 mins, respectively, and extruded into ¼" Teflon tubing. Propellant samples were cured and characterized the same way as the conventionally mixed samples.

2.3 Ballistic Testing

Strand burner experiments were conducted using a constant-volume pressure vessel at pressures of 3.45-20.68 MPa (500-3,000 psia). The four optical ports of the strand burner are equipped with several diagnostics and an alternative ignition method (CO_2 laser). The three side optical ports allow for high-speed video (Photron FASTCAM SA3 120K),

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light emission diode (New Focus 2031), and visible/near infrared spectroscopy (Ocean Optics USB2000) diagnostics. The transient pressure of the system during combustion is tracked using in-line pressure transducers (OmegaDyne PX02C1-7.5KG). Details regarding the operation and design of the strand burner are expanded upon by Carro et al. [20-21].
The burning rate (r) of propellant samples are computed by:

$$r = \frac{L}{t_b} \tag{1}$$

L is the sample length and t_b is the burn time. The sample length is measured using scientific calipers and the burn time is determined using the transient pressure data, light emission diode, or high-speed video. A representative pressure trace with an example burn time analysis is shown in Fig. 1.

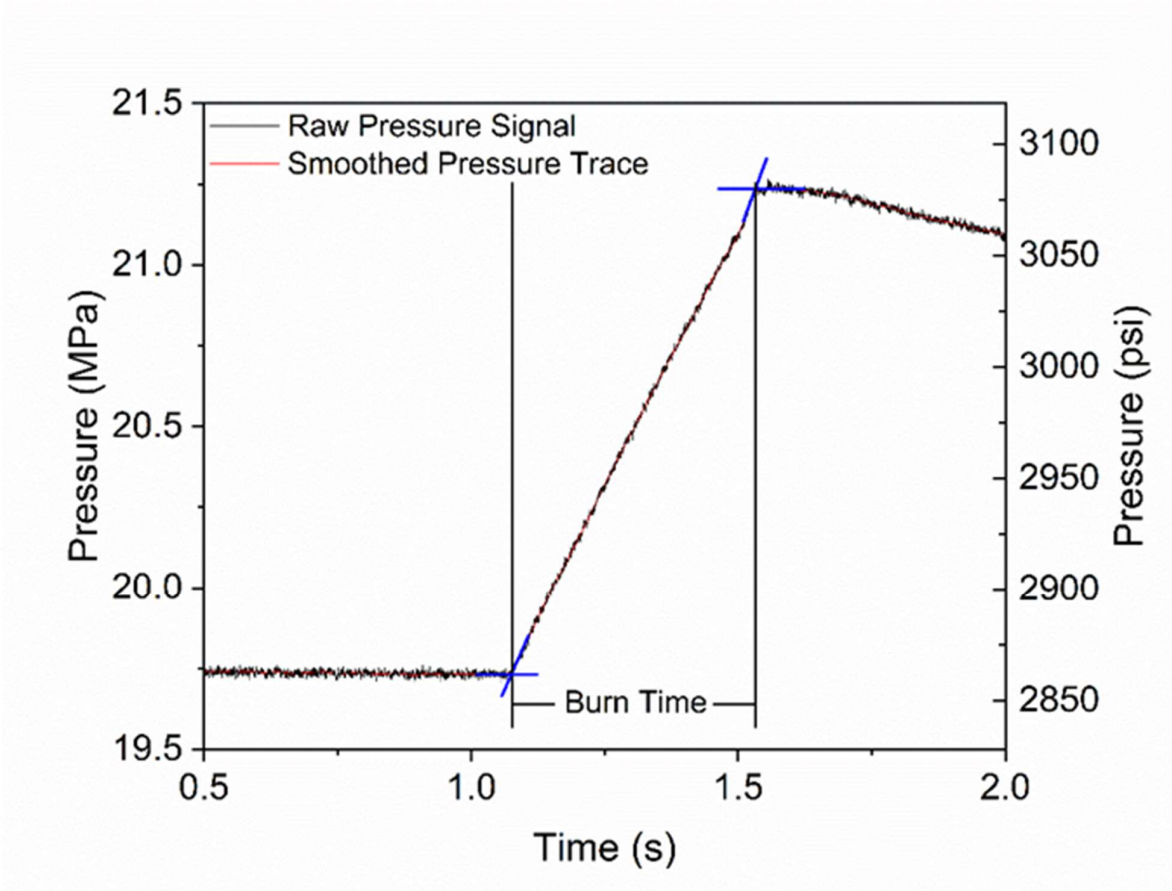

Figure 1. Representative pressure trace from a composite propellant ballistic experiment with the corresponding burn time measurement.

2.3 SEM Sample Preparation

Composite propellant cross sections from the high-performance propellant formulations were imaged on a Tescan VEGA3 SEM at the Texas A&M University Microscopy and Imaging Center (MIC) [22]. SEM was utilized herein to achieve a better understanding of the material distributions within the composite propellant matrix. Propellant cross sections were prepared by cutting propellant strands using a razor blade into ~0.1" sections. The propellant sections were affixed atop of aluminum pedestals using double-sided carbon tape. Samples were sputter coated with gold using a Cressington 108 sputter coater prior to imaging. Sputter coating prevents the accumulation of electrons that can cause charging and decrease the quality of the images.

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3. Results and Discussion

The burning rate results for all formulations investigated in the current study are shown in Fig. 2. Open and closed symbols correspond to conventional mixing and RAM, respectively. Black, red, and blue symbols correspond to the 80% monomodal baselines, high-performance (conventional), and high-performance (RAM), respectively. There is no difference in burning rate between the two mixing methods for the 80% monomodal formulations. However, there is an approximately 25% increase in burning rate for the high-performance formulation when RAM is used instead of conventional mixing. The high-performance RAM formulation exceeds 25.4 mm/s (1 in/s) above ~4.59 MPa (665 psia). It is worth noting that the high-performance RAM formulation was tested twice for demonstration of repeatability of the observed effect (half closed symbols in Fig. 2).

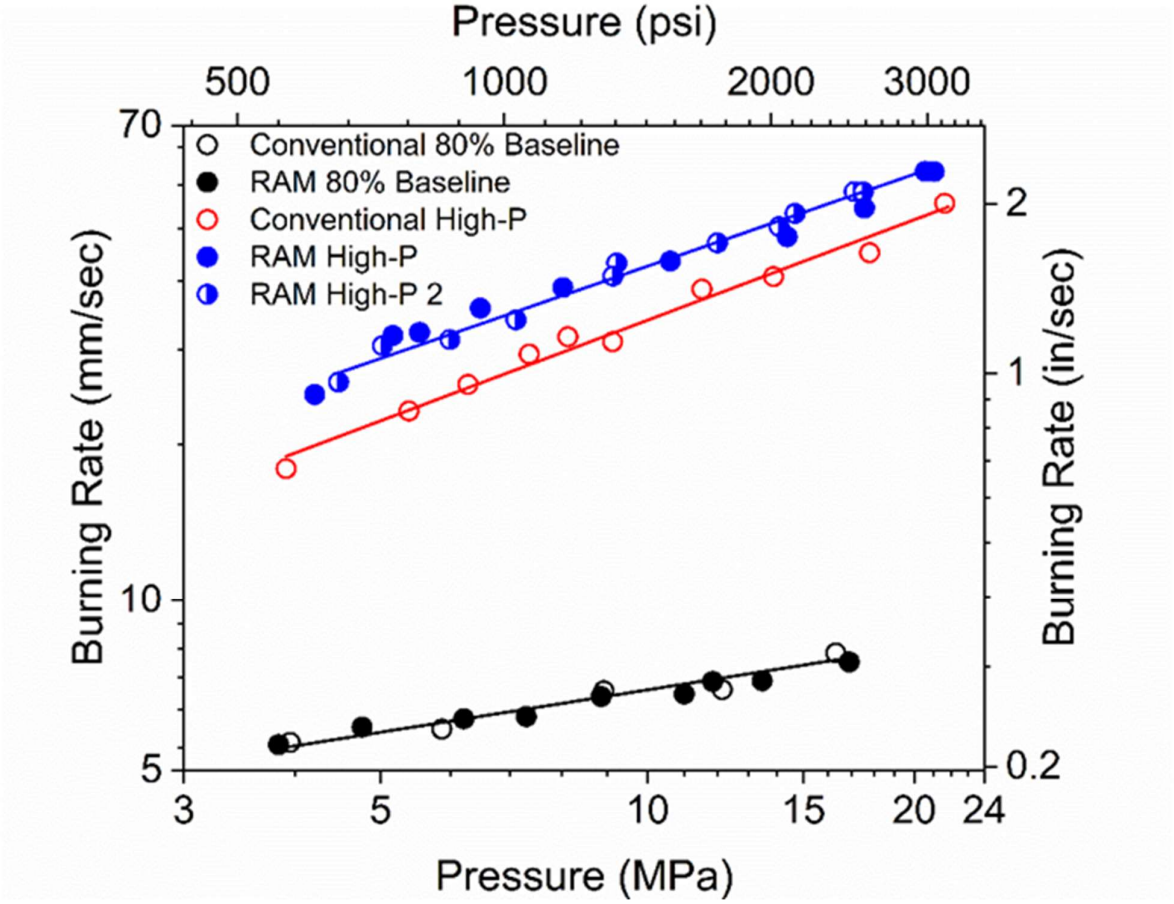

Figure 2. Burning rate data for baseline and high-performance propellant formulations mixed by conventional and RAM methods.

Representative SEM images of the conventionally mixed and RAM high-performance propellant formulations are shown in the top and bottom rows of Fig. 3, respectively. Metal atoms were illuminated using backscattered electron SEM imaging to gain a better understanding of the catalyst dispersion. More explicitly, the Fe_2O_3 particles in the propellant correlate to the bright white spots in the images of Fig. 3. There is a distinct difference in particle agglomeration between conventionally mixed and RAM samples. Better dispersion of catalyst additives has been directly correlated to increases in burning rate [22]. Accordingly, the ~25% increase in burning rate observed across the investigated pressure range for the RAM formulation can be attributed to the superior dispersion of the catalytic additives achieved with RAM.

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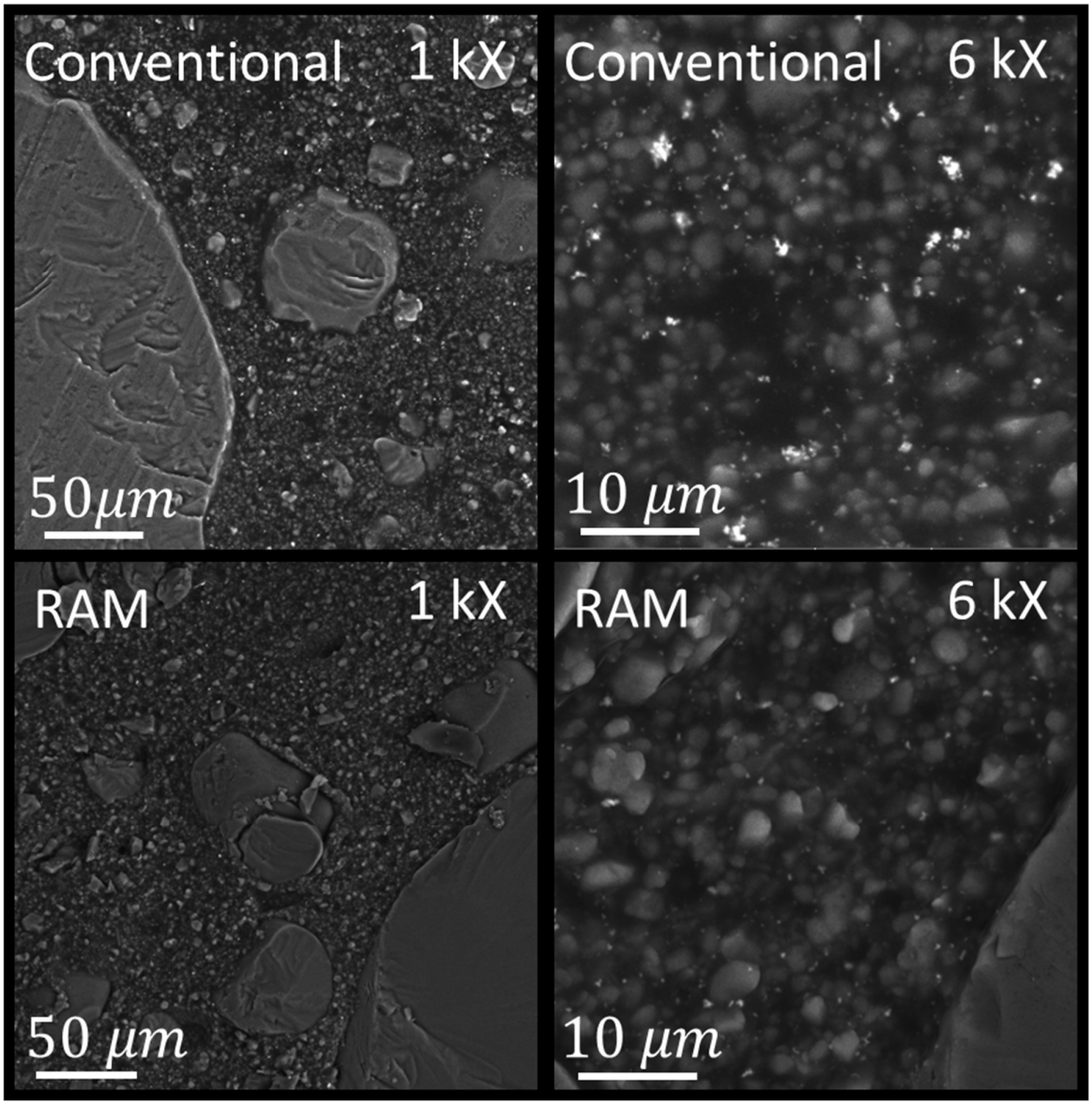

Figure 3. SEM imaging of (top) conventionally mixed and (bottom) RAM propellant samples at (left) 1 kX and (right) 6 kX.

4. Conclusion

Conventional mixing and RAM were successfully compared by manufacturing and burning of 80% monomodal and standard high-performance propellant formulations. The burning rates for both mixing methods agree well for the 80% monomodal formulation. However, a ~25% increase in burning rate was observed for the high-performance formulation when RAM was used to manufacture the propellant. The increase in burning rate is attributed to better dispersion of the catalytic additives as corroborated by backscattered electron SEM images. This work is the first direct observation of the underlying phenomena responsible for alternation of ballistic properties accompanying RAM strategies.

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