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Abstract. We provide measurements of the chemical reaction propagation rate (RPR) as a function of pressure using diamond anvil cell (DAC) and strand burner technologies. Materials investigated include HMX and RDX crystalline powders, LX-04 (85% HMX and 15% Viton A), and Composition B (63% RDX, 36% TNT, 1% wax). The anomalous correspondence between crystal structure, including in some instances isostructural phase transitions, on pressure dependent RPRs of HMX and RDX are correlated to confocal micro-Raman spectroscopic results. The contrast between DAC GPa and strand burner MPa regime measurements yield insight into explosive material burn phenomena. Here we highlight pressure dependent physicochemical mechanisms that appear to affect the deflagration rate of precompressed energetic materials.

Keywords: HMX, Composition B, LX-04, RDX, deflagration rate, Raman spectroscopy
PACS: 62.5.-p

INTRODUCTION

Measured chemical reaction rates of energetic materials at high pressures and temperatures are lacking in the published literature. Knowledge of high-pressure chemistry is fundamental toward understanding combustion and detonation processes. When combined with < 1 GPa strand burner measurements, DAC RPR data enable one to more confidently model the pressure dependent deflagration velocity of solid explosives. These results are important to understanding processes such as deflagration-to-detonation-transition (DDT), thermal explosion (and its associated violence), sympathetic detonation, and buildup of reaction following impact ignition, and generally assist in predicting and preventing accident scenarios.

We find that deflagration rates of precompressed explosives and polymer blended explosives never exhibit purely monotonic pressure dependence. Here we consider weather anomalous discontinuities observed in pressure dependent vibrational spectroscopic results may *a priori* signal nonlinear HE deflagration phenomena.

EXPERIMENTAL PROCEDURE

High pressure strand burner (HPSB) measurements consist of a stacked HE pellet

assembly that is initially pressurized to MPa pressure, prior to burn initiation, using argon gas [1]. Transient pressure is determined in situ using piezoelectric transducers while 1-D linear burn propagation is monitored by the extinction of uniformly separated signal wires. Deflagration rate is determined by the separation distance between consecutive wires divided by their corresponding extinction time.

Deflagration rates were also determined from materials precompressed to >1 GPa pressure within a diamond anvil cell. Sample thickness ranged from 40 to 80 μm, and sample diameters were between 140 and 300 μm. Pressure and deflagration measurements were performed at the same axial position within the sample chamber [2]. Strontium tetraborate served as the optical manometer, which was also used to determine pressure gradient within the chamber [3]. Samples were illuminated using a 9 Watt CW argon ion laser beam. The transmitted light creates a speckle pattern that is magnified by 10X, passed through a 100-300 μm vertical slit, and then magnified by 9.5X to create a line image that is focused onto the entrance slit of a EG&G-L-CA-20 streak camera. A 7 ns, 532 nm pulse from a Nd:YAG laser with 4-6 mJ energy served to photo initiate deflagration reactions. The spot size of the ignition pulse was ca. 4-5 microns. As a deflagration wave propagates radially away from the ignition center the reaction products typically extinguish the speckle pattern. The streaked image is light intensified and then exposed across a Polaroid (ISO 3000) film. Fixed streak rates were set to a pre selected value of between 0.5-5 μs. The spatial calibration of the film was accomplished using the image of an illuminated 44 micron pinhole. A streaked image typically consists of extinguished speckle lines, see Fig 1. The slope of the speckle extinction reveals the propagation velocity in μm/μs. We measured the reaction propagation speed of HMX consisting of two different particle size distributions. Both size distributions are from a lot B-881, where RDX volume is <7.4%. The B-881 lot has a trimodal grain-size distribution that is consistent with our

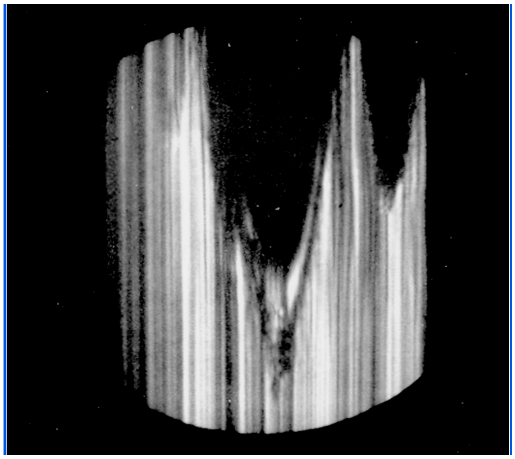

FIGURE 1. DAC RPR streak image from LX-04 initiated at 10.8 GPa. A sympathetic burn follows several hundred ns after the primary burn. The rates of each reaction front (30 m/s) are nearly identical.

LX-04 sample, which has 65% of its volume larger than 10μm. We studied this large grain HMX in the pressure range 0.7 to 7.7 GPa. We ground lot B-881 to produce a batch of what we term small-grain HMX where 90 % of the volume is smaller than 10 μm. We previously studied small grain HMX from 0.7 to 35 GPa where RPR's were determined by a disturbance to speckle lines [2]. Here we repeated that study in the range of 7.5 – 30.5 GPa where RPR's are determined by a complete extinction of speckle lines. We also performed tests on RDX (22 μm uniform grain size) in the pressure range from 2 to 23 GPa. A powder sample of ground composition B (5 μm uniform grain size) was studied in the range of 2 - 9 GPa. Non-ground composition B was studied for comparison where there is a wide grain size profile of centered at approximately 55 microns.

We conducted confocal μ-Raman spectroscopy on non-hydrostatically compressed HMX. The HMX was first heated within a DAC sample chamber to 110°C in a dry nitrogen environment. After several hours the sample chamber was sealed, brought back to ambient temperature, and pressurized. The probe consisted of a CW 632.8 nm laser

excitation beam attenuated to <3 mW and focused to a 4-5 μm spot. Backscattered Raman shifted light was focused onto an 80 μm slit and into a 0.3 m spectrograph, dispersed using an 1800 l/mm grating, and imaged onto a thermoelectrically cooled CCD. The signal collection time was 5 – 10 minutes per spectral window. A neon lamp served to calibrate grating dispersion. The instrument response function was determined by collecting parasitically scattered from the Re gasket. This data and CCD dark current data were subsequently used to remove background noise from HMX Raman data.

RESULTS/DISSCUSSION

In Fig. 2 we show HPSB and DAC RPR data from LX-04. The pressure dependent deflagration trend is essentially monatomic across two different measurement regimes spanning from ambient to 34 GPa. Our conclusion is that a consistent laminar burn process occurs with a pressure exponent value of 0.98 ± 0.01 (weighted average). In Fig. 3 we illustrate a pressure induced transition in burn phenomena occurring in Comp B. At ambient temperature in the HPSB, Comp-B burns rapidly, erratically, and has a pressure exponent of 2. These results suggest that these burns are convective rather than laminar. When Comp B is preheated to 100C the HPSB and DAC results are more consistent and less erratic. We conclude that preheating Comp B enabled molten TNT to fill the pours, voids, and defects found within the open structural matrix. As a result the burn process became laminar and more like the DAC result where GPa pressures have the effect of collapsing void structures.

DAC RPR studies of HMX and LX-04 are summarized in Fig. 4. Regardless of grain size, there are deflagration rate discontinuities observed at ca. 7, 19, and 26 GPa. We collected HMX μ–Raman spectra from ambient pressure to 40 GPa over a spectral range of 100 cm^{-1} to 3250 cm^{-1}. Figure 5 illustrates anomalous changes in the vibrational structure of HMX that may be correlated with RPR results. For example, CNC wag, NNO and NNC bend, and

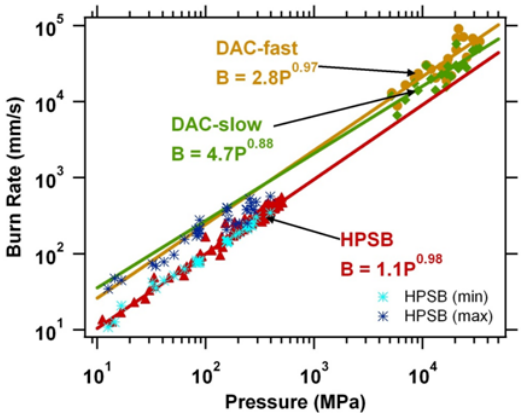

FIGURE 2. HPSB and DAC RPR's from LX-04.

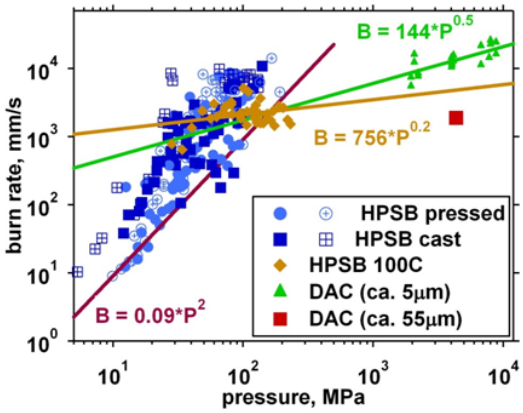

FIGURE 3. HPSB and DAC RPR's from Comp B.

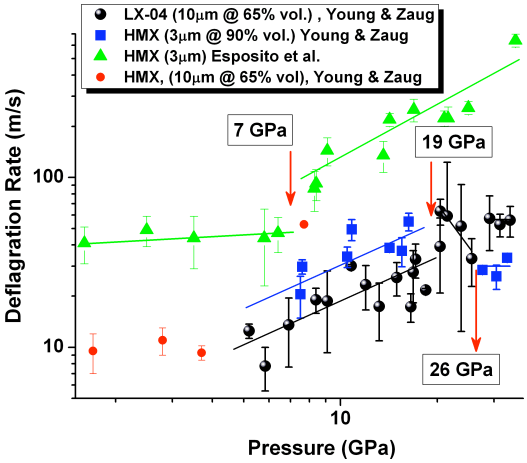

FIGURE 4. RPR results from HMX and LX-04

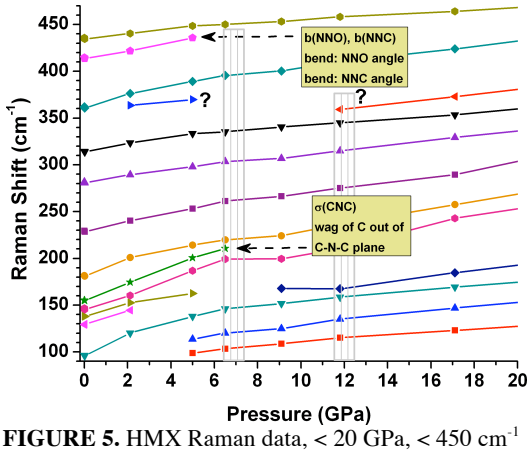

FIGURE 5. HMX Raman data, < 20 GPa, < 450 cm^{-1}

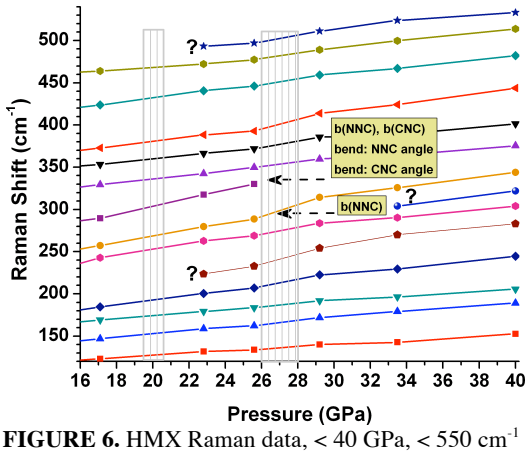

FIGURE 6. HMX Raman data, < 40 GPa, < 550 cm^{-1}

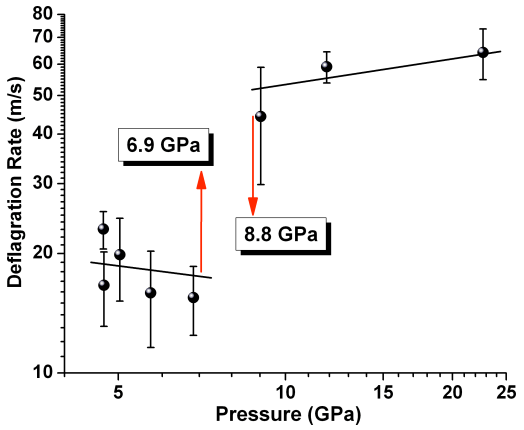

FIGURE 7. RPR results from RDX.

unassigned mode populations crash in the region where the RPR rate increases by 40-50%. A different set of NNC and CNC bend modes either terminate or show hardening at 26 GPa where the RPR rate doubles. Additional anomalous correlations are observed over the remainder of the intramolecular region including quite remarkable changes in slope for the CH2 stretch modes (not shown here).

One can see in the RPR data shown in figure 7 that a RDX deflagration discontinuity exists at 7-9 GPa. We also note that it was not possible to initiate a reaction in RDX when precompression pressure was below 4 GPa. The recently published Raman spectra from RDX by Dreger et al [4] reveal anomalous correlations to the GPa pressure deflagration inflections.

SUMMARY

All published DAC RPR studies of energetic material reveal pressure dependent discontinuities of the deflagration rate. These discontinuities are typically attributed to changes in crystal structure. A detailed analysis of pressure dependent inter- and intra-molecular vibrational modes and/or volume compression data from a series of CHNO explosives may reveal pivotal correlations to RPR's that will illuminate our understanding of pressure dependent deflagration chemistry.

ACKNOWLEDGEMENTS

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