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Quasi-Isentropic Compression of Vapor-Deposited Hexanitroazobenzene (HNAB): Experiments and Analysis

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Abstract. Vapor-deposited hexanitroazobenzene (HNAB) is an explosive with unique physical characteristics resulting from the deposition process that make it desirable for the study of microstructure effects. A relatively understudied high explosive, few data are available on the equation of state (EOS) of HNAB reactants or products. HNAB samples exhibiting high density and sub-micron porosity and grain size were prepared using physical vapor deposition onto polymethyl methacrylate (PMMA) and lithium fluoride (LiF) substrates. The samples were ramp compressed quasi-isentropically using VELOCE, a compact pulsed power generator. Evidence of a low pressure phase transition was observed in HNAB. Interferometric measurements of reference and sample interface velocities enabled inference of the unreacted EOS for HNAB using DAKOTA, an optimization toolkit. Initial simulations of the HNAB critical thickness experiment have been carried out using the parameterized EOS, and a products EOS from thermal equilibrium calculations.

INTRODUCTION

Experimental performance characterization of vapor deposited HNAB films has shown that HNAB has the unique property of sustained detonation at extremely small thicknesses in 2D geometries, with detonation velocities being recorded in samples as thin as 67 μm [1]. HNAB also maintains stability at temperatures above its melting point [2]. HNAB samples manufactured through vapor deposition start out with a metastable amorphous material structure which then crystallizes to HNAB-II and an unknown phase over time. The film density, final volume fraction of crystalline polymorphs, and crystallization process are all sensitive to the imposed crystallization temperature.

Being a less-studied material, significant characterization data are not available for HNAB. To further investigate detonation behavior in HNAB, equation of state (EOS) and constitutive properties are necessary. Because vapor deposited samples have practical limitations on thickness and mass, VELOCE [3], a compact pulsed power generator, was used to obtain characterization data. Quasi-isentropic methods have been used previously to obtain similar data for PBX materials [4, 5]. In this study, VELOCE is used on crystallized HNAB samples deposited directly onto the viewing windows to obtain the continuously varying HNAB/window interface velocity.

EXPERIMENTAL PROCEDURE

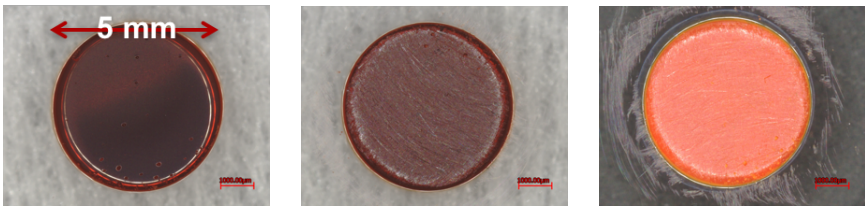

FIGURE 1. Optical micrographs of HNAB after deposition (left),after roughening (middle), and after crystallization.

Details of the HNAB vapor deposition process can be found in previous work [6]. A circular mask was used to grow films on two different window materials. The density of vapor deposited HNAB-II films is estimated to be 1.735 g/cm^3, or 99.5% of theoretical maximum, from ion-polished cross-section data [7, 8]. The final thicknesses for the two samples were 169 μm and 189 μm for films deposited on LiF and PMMA, respectively. To aid in uniform crystallization to the HNAB-II polymorph, deposited samples were roughened gently with 15 μm polishing paper. Crystallization was then carried out at room temperature, until the samples fully converted from amorphous to HNAB-II (see Fig. 1).

VELOCE is a compact pulsed power machine capable of delivering 3 MA of current which allows samples to be ramp compressed to pressures from 5 to 20 GPa [3]. As shown in Fig. 2, the sample and references panels are secured together with a shorting contact. The closed current loop generates a magnetic field, and the resulting Lorentz force compresses the sample in a quasi-isentropic manner. The continuous pressure ramp induces material motion which eventually reaches the window interface, where material motion is observed with both VISAR (velocity interferometer system for any reflector) and PDV (photonic Doppler velocimetry).

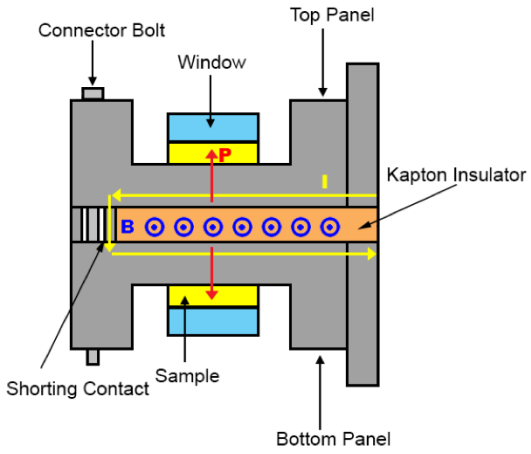

FIGURE 2. Diagram of VELOCE sample test blocks.

The VELOCE target used Al-6061 reference and sample panels with a 1.5 mm floor thickness. The reference panel had an LiF window bonded to it with EPO-TEK® 301 epoxy, and served as a reference measurement to accurately determine the driving pressure in the experiment, since both the panel and window equations of state are well known. On the sample panel, the vapor deposited HNAB sample and substrate, which served as the window, were bonded to the panel using Sylgard® 527 silicone. The VELOCE machine was charged to 52 kV and discharged through an Ar atmosphere, achieving a peak current of roughly 2 MA and a rise time of approximately 500 ns.

EXPERIMENTAL RESULTS

Window/sample interface velocities for the two HNAB samples are shown in Fig. 3. Velocity records from both experiments indicated the presence of a low pressure phase transition identified by the arrows in Fig. 3. Typically the velocity record is postprocessed using the Inverse Lagrangian analysis technique [9], where the interface record is mapped to *in situ* material velocities which can be used to determine the EOS similar to direct Lagrangian methods. The presence of a low pressure phase change complicates this analysis, as the method assumes isentropic flow. Analytical methods are currently being sought to obtain the EOS despite the phase change.

In the absence of analytical tools to determine the EOS and constitutive behavior, backward iterative methods first used by Hayes [4, 10] and Baer [5] were employed to calibrate the material model for use in simulations. These methods make use of the reference panel that consists of 6061-Al and a LiF window, whose properties are well known to determine the drive pressure. The backward integration technique uses a fixed time grid where the VISAR record is an initial condition. The equations of motion are marched backwards in space to the initial drive surface. In this manner the final material step results in the stress-time history at the drive surface. Once the drive conditions are determined, a standard forward calculation is run using the drive stress as a boundary condition for verification. In

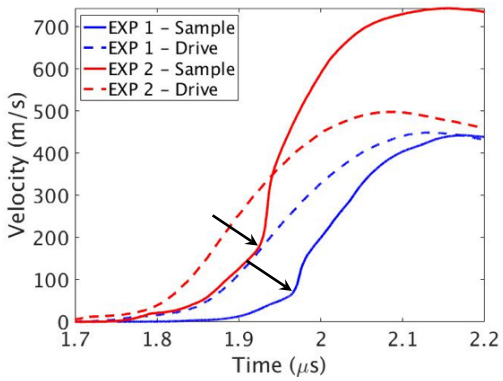

FIGURE 3. VELOCE drive and sample velocities. EXP1 used a LiF window, while EXP2 used a PMMA window.

Fig. 4, the reference interface velocity is shown along with the calculated drive surface velocity from the backward integration technique. Also, a CTH forward calculation on the reference geometry is shown as a verification of the calculated drive.

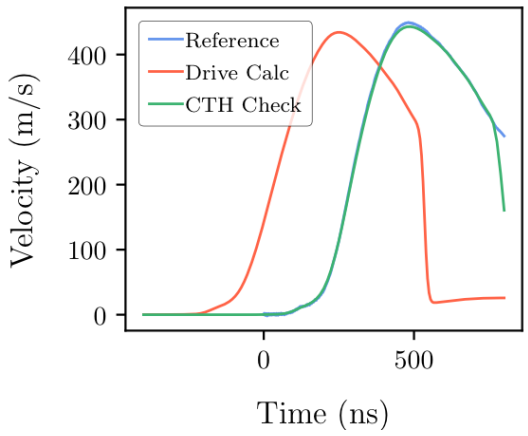

FIGURE 4. VISAR record (blue), calculated drive velocity (red), and forward calculated VISAR velocity (green) used for verification.

At this point in the analysis, the HNAB EOS and constitutive model are still unknown, and are determined through optimization using the known VISAR trace as an objective function. DAKOTA [11], an optimization toolkit developed by SNL, was used to calibrate a Mie-Gruneisen EOS and elastic perfectly plastic Von-Mises stress model to the data from the first experiment using the LiF window. In this study, a suitable model for phase change could not be found, so the determined EOS and strength model represent best fits to the data without explicitly modeling phase change. After the material models were determined, they were verified in a simulation of the second experiment, and shown to produce a satisfactory match to the data (see Fig. 5).

CRITICAL THICKNESS EXPERIMENT

The Critical Thickness Experiment at Sandia National Laboratories (SNL) is an experiment for characterizing the dependence of detonation velocity on thickness and the critical thickness in two-dimensional vapor deposited explosive samples. The experiment has been used previously to characterize PETN [12, 13], HNS [14], and HNAB [15]. Details on this experiment and data analysis techniques can be found in previously published work [13]. The “infinite” sample width was determined by testing several widths and comparing curves of detonation velocity vs. thickness. A sample

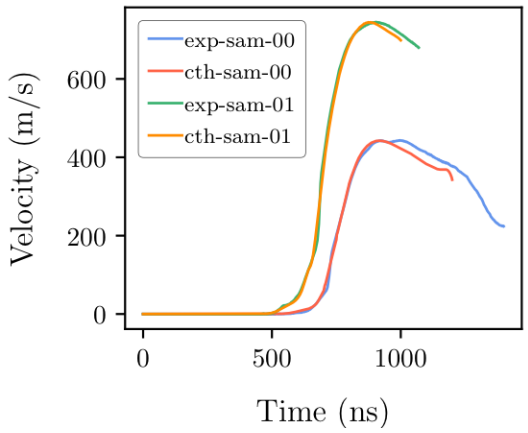

FIGURE 5. HNAB/LiF interface velocity from optimized material model and data (yellow/green) and predicted HNAB/PMMA interface velocity using the optimized model and data (red/blue).

width of 1.6 mm was determined to be sufficient to negate the boundary effects in the experimental measurement.

Using "infinite width" simplifies the numerical simulations by justifying the use of a two dimensional computational grid. Library EOS and material models were used for window and panel materials, and the VELOCE determined HNAB material data was also used. The PETN driver model employs two programmed burn PETN material definitions at two different densities to account for a known density gradient in the PETN initiation increment. The thickness of each section was determined by comparison to experimental data of the PETN detonation velocity [13]. Arrhenius reactive burn (ARB) was used to model the rate of conversion of the explosive, and the determination of the HNAB reactive burn model parameters was carried out in a manner similar to the HNAB EOS and strength model. A second order power law function was fit to the experimental thickness vs. detonation velocity data for room temperature crystallized HNAB. The objective function used for DAKOTA optimization of the burn model parameters was the mean square error between the power law fit and the simulation predicted velocities at 5 different thicknesses.

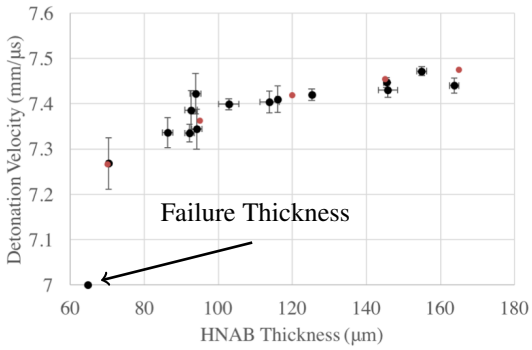

FIGURE 6. Experimental (black) and simulated (red) HNAB detonation velocity vs. thickness.

Figure 6 shows the predicted velocities at five points based on the optimized model, together with the experimental values. The ARB model is able to capture well the drop off in detonation velocity at reduced thicknesses, and also captures the asymptotic value of 7.42 mm/μs at very large thicknesses (1 mm). Also shown on the plot is the thickness of 65 μm where sustainable detonation is not achieved, and which is not predicted by the simulations. There are several likely reasons for this that are currently being tested. Due to the instability of detonation propagation near failure, the simulation is very sensitive to the PETN driver model. Slight changes in the driver model, e.g., changes in density or thickness in the two material model, result in large changes in prediction of the failure thickness with virtually no change in predictions of detonation velocity for other thicknesses. Alternative experiments are being considered that can decouple the measurement and prediction of critical thickness from the initiation train.

FIGURE 7. Optical micrograph of dent track from detonation of an HNAB film.

Recovered substrate samples from experiments conducted on HNAB tracks 400 μm wide and near the failure thickness resulted in a cellular detonation structure (see Fig. 7 and [1]). Also evident in these substrate records is that edge effects, although not observed to influence steady detonation behavior at larger thicknesses, appear to play a much larger role near the failure thickness. This would suggest that while the two-dimensional model is adequate for capturing the velocity deficit with thickness, a three-dimensional model will likely be required to accurately capture the failure at the critical thickness.

CONCLUSIONS

Samples of vapor deposited HNAB were crystallized at room temperature onto LiF and PMMA windows and attached to VELOCE sample blocks. The samples were subjected to quasi-isentropic ramp compression. Using a backward integration technique, the drive conditions were inferred from the recorded drive panel/window interface. DAKOTA was then used to calibrate a Mie-Gruneisen EOS and EPPVM strength model that matches the velocity data. Critical thickness data of detonation velocity vs. HNAB thickness was captured accurately using these material models and an ARB model. It was found that a more accurate PETN driver model, and a full three-dimensional simulation will be required to capture the observed failure thickness numerically.

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