ISENTROPIC COMPRESSION OF HIGH EXPLOSIVES WITH THE Z ACCELERATOR

David B. Reisman, Jerry W. Forbes, Craig M. Tarver, and Frank Garcia
Lawrence Livermore National Laboratory
Livermore, CA 94551

Dennis B. Hayes
Tijeras, NM

Michael D. Furnish
Sandia National Laboratories
Albuquerque, NM

Jerry J. Dick
Los Alamos National Laboratory
Los Alamos, NM 87545

Isentropic compression experiments (ICE) were performed on a variety of high explosives. The samples were dynamically loaded by Sandia National Laboratory's Z-accelerator with a ramp compression wave of 300 ns rise time and peak stress of 100-350 kbar. Sample/window interface velocities were recorded with VISAR. Experiments were performed on LX-04 to obtain the stress-strain relation using a backward integration technique. Experiments were also performed on LX-17, and the results compared to hydrodynamics calculations using the Ignition and growth reactive flow model. Recent experiments were also conducted on single crystals of HMX with the goal of detecting the phase transition at approximately 270 kbar.

INTRODUCTION

A technique[1,2] have been developed for doing quasi-isentropic compression experiments (ICE) on materials using pressure from a strong magnetic pulse generated by the Z-accelerator. This technique allows a compression isentrope of a material to be measured in one experiment over the range of zero to peak pressure in the ramp waves. It has been demonstrated[3] that explosives can be studied using this technique. Temperature in the explosives will be lower for isentropic ramp wave compression than for shock compression for the same peak stress level. Therefore, isentropic data can be obtained at higher stresses without reaction occurring. This isentropic data can be used to calculate what the unreacted Hugoniot would be at high stress levels, which is useful in testing predictions of detonation wave structure.

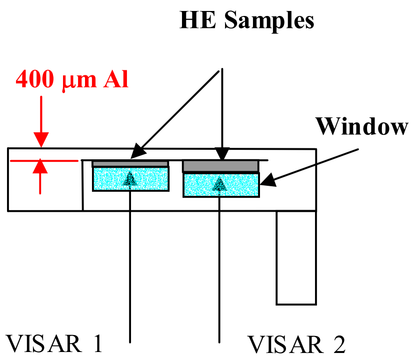

Figure 1. Arrangement of HE samples on ICE. The "ramp" pressure load is applied from the top.

The ICE wave profiles develop structure in them due to material properties such as phase transitions or changes in compressibility[1-3]. Therefore, it is a technique that can measure material properties of unreacted explosives. Studies of the kinetics of reacting energetic materials can also be done using this experimental technique.

EXPERIMENTAL PROCEDURES

Isentropic compression experiments were performed on high explosives using the "square short" assembly shown in Figure 1. In this configuration pairs of samples consisting of nominally 6 mm diameter disks of thickness 250-

600 μm were placed on driver panels of aluminum of base thickness 400 μm. Windows of LiF or PMMA were bonded to the back of the HE samples to minimize wave reflections from the measurement surface which simplifies data analysis. A pressure ramp of approximately 300 ns duration and peak pressure ranging from approximately 100 to 400 kbar, depending on the experiment, was applied to the aluminum surface. The resulting HE/window interface velocity history was recorded using single-point VISAR.[4]

The pressure profile chosen was tailored to delay shock up of the sample. This was accomplished by firing 9 lines of Z's 36 lines with 200 ns pulses. The pressure profile, based on B-dot measurements is shown in Figure 2.

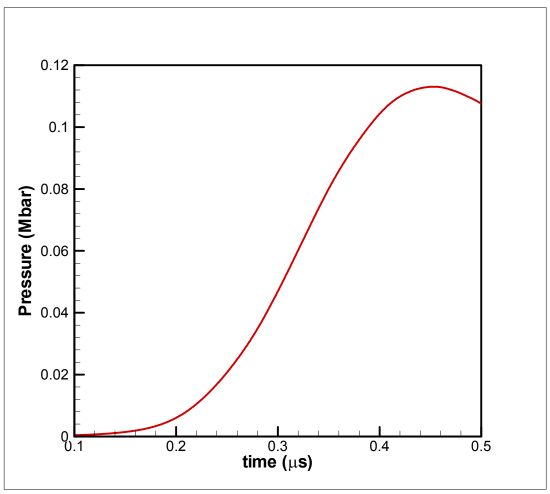

Figure 2. Generic pressure drive for ICE. All pressure waveforms in this work had this basic "tailored" shape.

LX-04 EXPERIMENTS

Experiments were performed on LX-04 to obtain the isentrope. Sample thickness pairs were chosen to be 300/500 μm and 300/600 μm. The samples were isentropically loaded up to 100 kbar. Figure 3 gives the velocity waveforms for this experiment.

Analysis of the samples was performed using a backward integration technique[5]. This is an iterative technique that involves backward integrating the equations of motion to a common point in space using the velocity waveforms. In this processes two or more VISAR velocity histories, the "initial conditions", are integrated to a common spatial point (in this case the front surface of the sample x=0) while two equation of state (EOS) parameters are varied. When the pressure drives of each sample agree with each other, the EOS parameters are determined. The EOS form was taken to be the Mie-Grüneisan form. Two EOS parameters were varied until the pressure histories at x=0 for the two experiments agreed. These were C_0 and S defined by $U_s=C_o + S\ u_p$ which represents the Hugoniot[6] for the unreacted LX-04 with u_p being the particle velocity and U_s the shock velocity. S is slope of line in U_s,u_p space, and C_o is value of shock velocity at u_p=0.

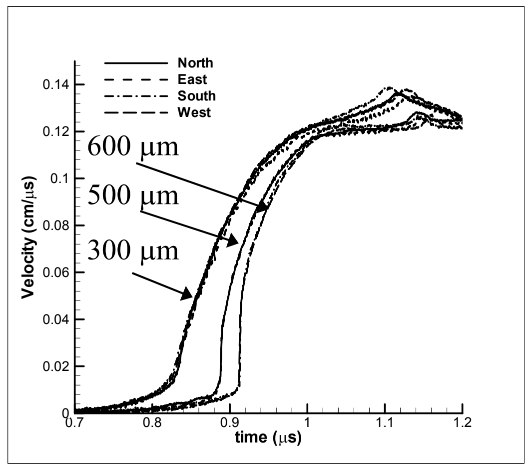

Figure 3. Velocity waveforms for LX-04 experiments measured with VISAR.

This analysis was performed on two of the 300/500 μm sample pairs. The sample pairs with the 600 μm samples were not used as the waveforms displayed a shock or steady wave which violates the reversibility assumption of the backward integration technique. A Grüneisan gamma of 0.8, determined from previous experiments, was assumed. The EOS parameters were found to be C_0= 0.244 cm/μs and S=2.261. The measured Hugoniot from shock wave experiments[7] resulted in values of C_0= 0.236 cm/μs and S=2.43. Hugoniots calculated from both sets of C_o, S are shown in Figure 4 with agreement of 2% which is within experimental error.

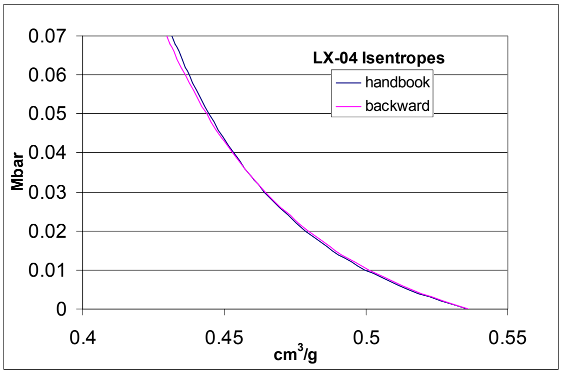

Figure 4. P-V isentrope of LX-04 from this work compared with that determined from shock waves.

LX-17 EXPERIMENTS

We also performed similar experiments on LX-17. Sample thickness pairs were 200/350 mm and the velocities were observed through a LiF window. The results are shown in Figure 5 along with a hydrodynamic simulation performed with the Ignition and Growth reactive flow model. We chose to concentrate on the velocity waveforms from a 300 and 350 μm samples. Although samples of thickness ranging from 200 to 400 μm were fielded only the intermediate thicknesses could be used. The thin samples encountered a wave reverberation from release that the sample/window interface that altered the loading history. The thick samples encountered a shock that precludes their use in isentropic compression analysis.

Using the velocity waveforms from the 300 and 350 μm samples we backward integrated to x=0. Again a Mie-Grüneisan model was used and we assumed the LLNL HE Handbook[7] values of C_0=0.233 cm/μs and S=2.32. The resulting pressure waveforms at x=0 are shown in Figure 6. The good convergence of both calculations for the pressure drive from the two measured wave profiles suggests our experimental isentrope is within experimental error of the theoretical one based on the values of C_0 and S determined from shock wave experiments.

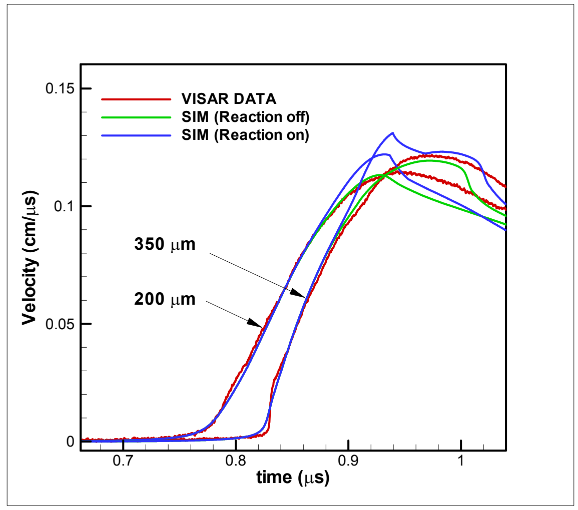

Figure 5. Experimental velocity waveforms of LX-17 (VISAR DATA) compared to HD calculations (SIM) performed with Ignition and Growth model. Best agreement is obtained when reactions are turned off in model.

REACTIVE FLOW MODELING OF LX-17

The Ignition and Growth reactive flow model of shock initiation and detonation of solid explosives has been used to solve many 1D, 2D, and 3D explosive and propellant safety and performance problems.[8-14] The model uses two Jones-Wilkins-Lee (JWL) equations of state, one for the unreacted explosive and one for its reaction products, in the temperature dependent form:

$$p = A\, e^{-R_1 V} + B\, e^{-R_2 V} + \omega C_V T/V \qquad (1)$$

where p is pressure in Megabars, V is relative volume, T is temperature, ω is the Gruneisen coefficient, C_V is the average heat capacity, and A, B, R_1, and R_2 are constants. The reaction rate law for the conversion of explosive to products is:

$$dF/dt = \underset{(0<F<F_{igmax})}{I(1-F)^b(\rho/\rho_o-1-a)^x} + \underset{(0<F<F_{G1max})}{G_1(1-F)^c F^d p^y}$$

$$+ G_2(1-F)^e F^g p^z \qquad (2)$$
$$(F_{G2min} < F < 1)$$

where F is the fraction reacted, t is time, ρ is the current density, ρ_o is the initial density, and I, G_1, G_2, a, b, c, d, e, g, x, y, and z are constants. The mixture equations assume pressure and temperature equilibration between the unreacted explosive and its reaction products.

This three-term rate law describes the three stages of reaction generally observed in shock initiation and detonation of heterogeneous solid explosives. For shock initiation, the first term represents the ignition of the explosive as it is compressed by the leading shock wave creating heated areas (hot spots) as the voids in the material collapse. The fraction of explosive ignited is approximately equal to the original void volume.[6] The second reaction models the relatively slow growth of isolated hot spots as they transfer heat to and cause decomposition in the neighboring explosive particles. The third term is used to describe the relatively fast completion of reaction as the hot spots coalesce and the transition to detonation occurs. The first term in Equation (2) uses the compression rate of the unreacted explosives to start the ignition process. This was originally done to allow the model to calculate slower and multiple compression processes as well as single shock compression.

The original experimental data upon which this ignition criteria was tested is the ramp wave initiation of PBX 9404, an HMX-based explosive, through various thicknesses of pyroceram by Setchell.[13] The good agreement reported previously by Reisman et al.[3] between ICE experiments and Ignition and Growth modeling for LX-04, a less sensitive HMX-based explosive than PBX 9404, showed that the ignition term works well for other HMX explosives. However, we find that for LX-17 the best agreement is obtained when the reactions are turned off in the model (Figure 5). The Ignition and Growth model for LX-17 was normalized to shock initiation data and thus represents an upper limit to the amount of reaction occurring during any compaction process. Even during shock compression, LX-17 requires a pressure of over 70 kbar to begin reacting. Since ICE is a much slower compaction process, less reaction is expected. The close agreement between the measured ICE velocity histories and the calculations assuming no reaction in Figure 5 implies that the LX-17 undergoes little or no reaction during the experiment.

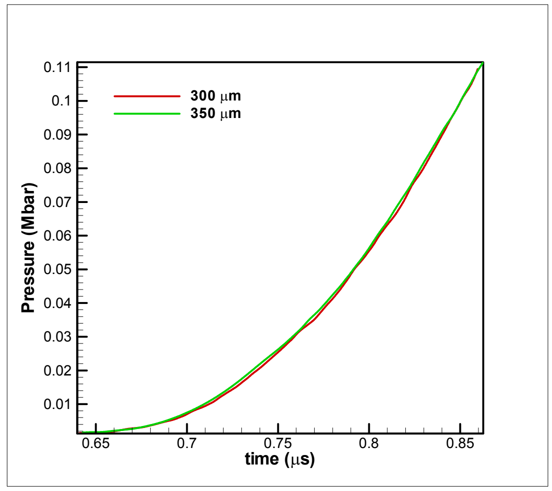

Figure 6. Backwards integration to x=0 of 300 and 350 mm LX-17 samples assuming handbook values of C_0 = 0.233 cm/μs and s = 2.32.

SINGLE CRYSTALS OF HMX

The purpose of this experiment was to dynamically observe the 270 kbar phase transition in HMX that was first reported under static conditions in the diamond anvil cell (DAC) work of Yoo and Cynn.[16]

Experiments were performed on (010) oriented single crystal HMX. The experimental arrangement was similar to that shown in Figure 1 but slightly modified to achieve higher current densities and therefore higher pressures. Sample thickness pairs were chosen to be 400/600 μm and 500/600 μm with both LiF and PMMA windows. The samples were again loaded with a 300 ns tailored pressure pulse. The maximum pressure achieved in the HMX was estimate to be approximately 400 kbar based on impedance matching to the LiF windows.

The characteristic two-wave structure of a phase transition with negative volume change was not observed. However, inflections in the velocity histories indicate an anomalous change in bulk modulus with pressure. This could be due to slow kinetics of a phase transition or else an unknown artifact of the experiment.

To guide our analysis we performed hydrodynamic flow (HD) calculations using a modified EOS. The equation of state QEOS was used.[17] This is global equation of state based on Thomas-Fermi theory. A modified version of this code allows the fitting of the EOS to experimental data by adding adjustable terms to the zero-Kelvin energy isotherm.[18] This was done using the DAC data of Yoo and Cynn.[16] The fit to the DAC data compared to a Mie-Grüneisen EOS based on a linear Us-Up fit to Hugoniot data is shown in Figure 8.

Calculations of VISAR window velocities were performed by assuming a pressure drive obtained from experimental current measurements and scaling to the maximum pressure reached in this particular experiment. These calculations clearly showed the classic two-wave structure whereas the VISAR data did not. However, the inflections seen in the data of Figures 9 & 10 seem to occur at the same velocity level as those of the simulation. This seems to indicate some change in compressibility, although not the sharp discontinuity of a volume collapse, in the range of the observed phase transition under static conditions.

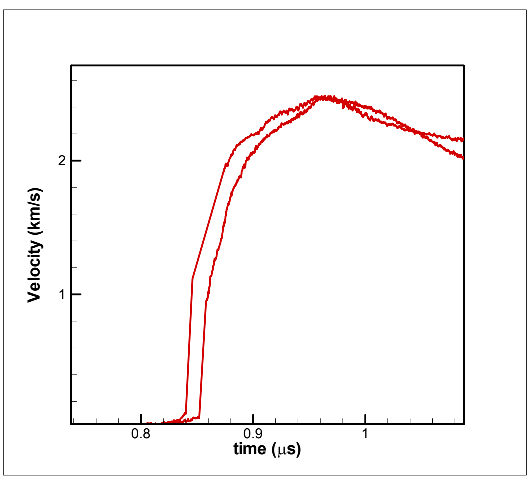

Figure 7. Velocity profiles of HMX/LiF interface.

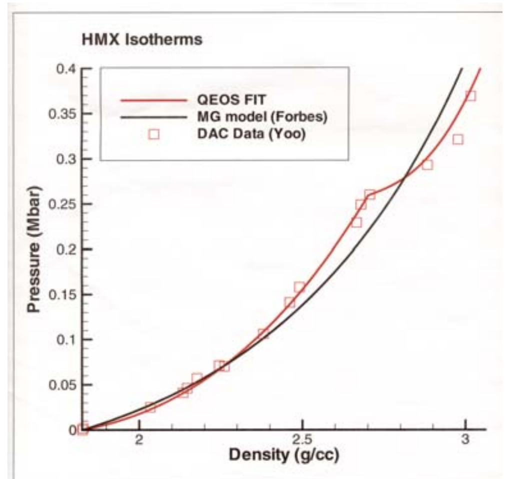

Figure 8. EOS fit used for performing HD calculations. Also shown is a Mie-Grüneisan model derived from a Us-Up fit to experimental data.

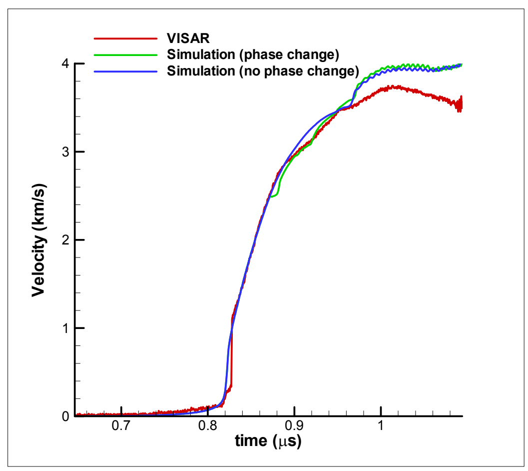

Figure 9. Velocity vs. time calculations performed with and without phase transition compared to experimental data of HMX.

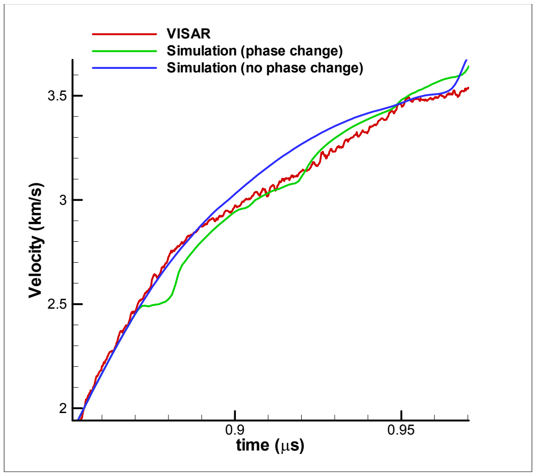

Figure 10. Close up of region of interest in figure 9. Note that calculations without phase transition do not display any inflections

CONCLUSIONS

We have performed isentropic compression experiments on LX-04, LX-17, and (010) oriented single crystal HMX. We were able to extract an accurate isentrope for LX-04 and LX-17 and validate the unreacted EOS model in the Ignition and Growth reactive flow model for LX-17.

Experimental results on (010) oriented HMX single crystals suggest an anomalous compressibility behavior at pressures of around 270 kbar, the region where DAC data suggests a phase transition occurs. However, a distinct two-wave structure under a fast volume collapse phase transition such as that observed in iron[1] was not observed for HMX. We speculate that this is due to slow kinetics of the transition or an unknown artifact of the experiment.

Future experiments on single crystals of HMX will use an independent pressure drive measurement – a LiF window placed on the aluminum driver plate. This will allow us to determine conclusively if there was a change in compressibility around 270 kbar. We also plan to use NaCl windows on several samples. The good impedance match between NaCl and HE will allow us to eliminate possible wave-reverberation effects as the source of the inflections in the velocity waveforms.

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