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DETONATION WAVE PROFILE IN PBX 9501

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Abstract. Measurements of a CJ-detonation wave in PBX 9501 with a VISAR technique have shown a classical ZND profile for the reaction zone. This is compatible with one-dimensional simulations using realistic equations of state and an Arrhenius reaction rate fit to available data from other experiments. Moreover, the reaction zone width is less than the average grain size in the PBX. In contrast to initiation, which requires hot spots, the reaction rate from the bulk shock temperature is sufficiently high for propagating a detonation wave. This raises questions with burn models used for both ignition and propagation of detonation waves.

Keywords: PBX 9501, reaction zone profile, CJ detonation wave, ZND model
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INTRODUCTION

A planar detonation wave can be promptly initiated with a projectile from a gas gun. Several experimentalist have measured the wave profile using a VISAR technique. Here we study the underdriven or CJ-detonation wave profile in PBX 9501 — an HMX based plastic-bonded explosives pressed to within 1.5 % theoretical maximum denstiy — from the experiments by Gustavsen, Sheffield and Alcon [1, 2]. The experiments used two VISARs with different fringe constants in order to determine velocity jump at shock front. The estimated timing resolution is between 1 and 3 ns; the better resolution when velocity jump corresponds to an integer number of fringes.

The VISAR record displays an abrupt rise within the time resolution. Thus, despite the heterogeneities in the PBX and the laser spot size of many grains, the wave front is a shock. Moreover, the shape of the profile corresponds to a classical ZND reaction zone. We note that similar results have been found by Fedorov [3]. The measured ZND profile motivates us to run one-dimensional simulations with an Arrhenius reaction rate to compare with the VISAR data.

We begin in the next section with a discussion of equations of state for both the reactants and the products of PBX 9501. Since chemical reaction rates are temperature sensitive, a complete EOS is needed. A key parameter for the shock temperature is the specific heat. Next we discuss the Arrhenius rate parameters. Some parameter sets for HMX in the literature and commonly used in simulations yield a rate at the von Neumann spike which is unreasonably large by several orders of magnitude. The next section describes the simulations. Simulated VISAR data is compatible with the experimental data. The reaction zone width is substantial less than the average grain size. Thus, in contrast to initiation, which requires hot spots, the reaction zone of a propagating detonation wave is dominated by the reaction rate from the bulk temperature.

EQUATIONS OF STATE

Data for the unreacted PBX 9501 Hugoniot and several fits are shown in fig. 1. The data up to 15 GPa is compatible with several linear fits in the literature. Extrapolated to detonation velocity (D=8.8 km/s) gives a large difference for the particle velocity (2.8 to 3.9 km/s), and would have a large effect on the von Neumann spike pressure and temperature. An

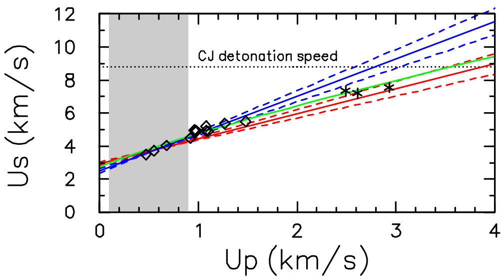

FIGURE 1. Unreacted Hugoniot for PBX 9501. Red and blue lines are from [8, §7.3, p. 116]; dashed lines are error bars and gray region is domain of fit. Green curve is fit to isothermal data [4, 5]. Black dotted line is CJ detonation velocity (8.8 km/s). Diamonds are data points from [9] and stars are single crystal HMX data from [10, p. 595].

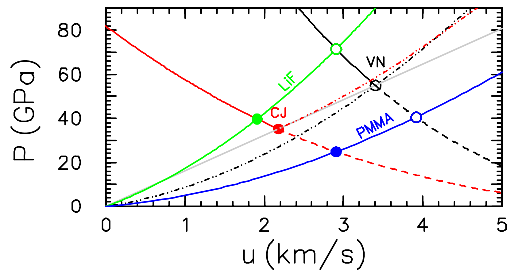

FIGURE 2. Impedance match for detonation wave in PBX 9501 with window. Green and blue curves are Hugoniot loci for LiF and PMMA, respectively. Black and red curves are for reactants and products, respectively. Gray is Rayleigh line corresponding to CJ detonation velocity. Labels VN and CJ denote von Neumann spike and Chapman-Jouguet state, respectively. Open circles are match from VN spike and solid circles are match from CJ state.

equation of state fit to HMX isothermal compression data to 27 GPa [4, 5] is compatible with Hugoniot data, including high pressure (40 GPa) single crystal HMX data. This is the basis for a complete EOS described in [6]. It uses the specific heat determined from molecular dynamics simulations [7].

In the range of interest for the reaction zone profile ($T = 2000$ to 3000 K) $C_V \approx 2.0 \times 10^{-3}$ (MJ/kg)/K. This is compatible with the pseudo-classical limit for lattice vibrations; $C_V = (3N - N_H)R/M$, where N is the number of atoms per molecules, N_H is the number of hydrogen atoms, M is the molecular weight, and R is the gas constant. For HMX ($C_4N_8O_8H_8$), $N = 28$, $N_H = 8$ and $M = 0.296$ kg/mole gives $C_V = 2.1 \times 10^{-3}$ (MJ/kg)/K. We note that this is larger than published data [8, §5.3, p. 112] which extends only up to β–δ transition temperature; $C_P = 1.57 \times 10^{-3}$ (MJ/kg)/K at $T = 450$ K.

For the equation of state of the reaction products a SESAME table [11] is used. The products EOS is fit to data on overdriven detonations and release isentropes in PBX 9501 [12, 13]. The thermal part of the EOS is based on the assumptions that the CJ temperature is 3000 K and the specific heat is 0.5 cal/g (2.07×10^{-3} (MJ/kg)/K).

From the equations of state, impedance matches with the VISAR windows (PMMA and LiF) can be used to estimate the end states of the reaction zone; von Neumann spike and CJ states. This provides a consistency check on both experiments and simulations. The graphical solution to the impedance match is shown in fig. 2.

REACTION RATE

We assume a first order Arrhenius rate; $(1-\lambda)k\exp(-T_a/T)$. For PBX 9501 we use an activation temperature $T_a = 17922$ K and multiplier $k = 5.6 \times 10^5\,\mu\mathrm{s}^{-1}$ based on the "global rate" of Henson *et al.* [14]. The temperature in the ZND profile — based on the EOS of PBX 9501 — varies from 2100 K at the von Neumann spike to 3000 K at the CJ state. In this temperature range, the inverse reaction rate varies from 1 to 10 ns.

A set of Arrhenius parameters commonly used for simulations is based on differential scanning calorimetry experiments of Rogers [15]; see also [8, §5.7, p. 113]. The reaction rate for these two sets of parameters is shown in fig. 3. We note that the rates differ by several orders of magnitude.

Henson's rate and Rogers' rate cross at 470 K. We note that Rogers' calorimetry experiments covered a narrow temperature range about the melting temperature of HMX; from 544 K to 558 K [15, fig. 11]. Rogers' rate would give a sub ps reaction time for a CJ detonation. Since the time for a detonation wave to cross a unit cell in an HMX crystal is 0.1 ps, Rogers' rate is unphysically large. In contrast, Henson's rate is compatible with the 3 high pressure data points for single crystal HMX; experiments by Craig reported in [16, p. 1065] and [17, p. 218].

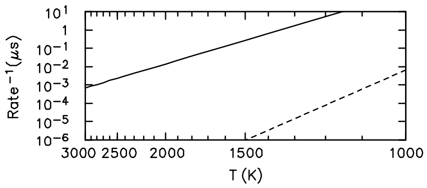

FIGURE 3. Inverse reaction rate vs temperature. Temperature is plotted on inverse scale. Dashed curve uses Arrhenius parameters in [8, §5.7, p. 113] ($T_a = 26522$ K, $k = 5.0 \times 10^{13}\,\mu\text{s}^{-1}$) and solid curve based on [14] ($T_a = 17922$ K, $k = 5.6 \times 10^5\,\mu\text{s}^{-1}$).

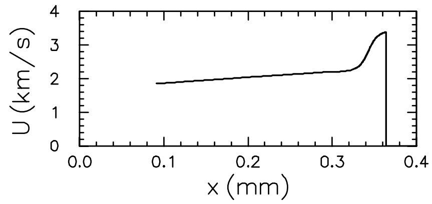

FIGURE 4. Wave profile immediately before detonation wave impacts window.

REACTION ZONE PROFILE

Simulations have been run with the Amrita environment of James Quirk [18, 19] using an adaptive mesh algorithm in order to resolve fully the reaction profile. Simulations are initialized with a steady ZND profile and use a piston boundary condition. The piston is given a ramp velocity to simulate a Taylor wave following the CJ state. The velocity profile immediately before the detonation wave impacts the VISAR window is shown in fig. 4. The reaction zone width is 24 μm at 90 % burn fraction, substantially less than the average grain diameter (140 μm) in PBX 9501.

We note that a propagating detonation wave is subject to a pulsating instability; see [20, chpt. 6A] and references therein. This is a generic property of an Arrhenius reaction rate. The calculations are over a sufficiently short distance of run that the instability does not have time to develop. Presumably the instability would be ameliorated by hot spots and three-dimensional effects.

Comparison of simulated VISAR profiles with experimental data is shown in fig. 5. The simulated profiles are consistent with the impedance match, fig. 2, for the equations of state being used. Moreover, they are compatible with the experiments using a PMMA window. The von Neumann spike is clipped in the VISAR experiment with a LiF window. This is due to the time resolution. For the fringe constants used, the PMMA experiments have a 1 ns resolution while the LiF experiments have a 2-3 ns resolution.

Similar experiments with an HMX based PBX and LiF window were performed by Fedorov. With 1 ns time resolution, the VISAR record [3, fig. 2a] shows a von Neumann spike amplitude close to the value in the simulation, fig. 5. Thus in order to resolve the von Neumann spike, 1 ns or better time resolution is needed. We note that the reaction time in Fedorov's experiment is nearly twice as large as shown in fig. 5. This is due to initiating the PBX with a detonator rather than a flyer plate. A detonator results in a curved detonation wave and a detonation velocity lower than the CJ speed for a planar wave. This in turn lowers the temperature behind the von Neumann spike and due to the sensitivity of the Arrhenius rate can have a large effect on the reaction time.

Finally, we note that similar experiments have been done with PBX 9501, PBX 9404, EDC 37. These PBXs have similar high percentage of HMX and low porosity. But they have different binders and initiation sensitivity. Nevertheless, their reaction zone profiles are nearly the same [1, fig. 2]. This is consistent with the reaction rate being dominated by the bulk shock temperature.

Most burn models for coarse resolution engineering simulations use pressure dependent reaction rate motivated by homogenization or a volume average over small region containing many hot spots. Since the underlying reaction mechanism for propagating detonation waves appears to be different, this raises the question as to why burn models work as well as claimed when applied to applications involving the curvature effect and corner turning.

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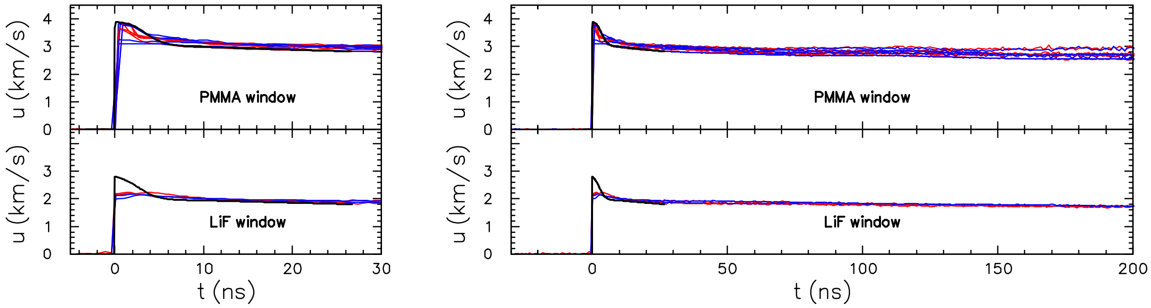

FIGURE 5. Comparison with VISAR data from [1, 2]. Top figures are for PMMA window and bottom are for LiF window. Left and right figures are on 30 and 200 ns time scale, respectively. Red and blue curves are experiments and black is simulations. VISAR used two laser beams with different fringe constants per experiment. Experiments varied drive pressure for initiation and the length of PBX sample.

wave profile experiments.

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