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PROGRESS IN MEASURING DETONATION WAVE PROFILES IN PBX9501

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We have measured detonation wave profiles in PBX9501 (95 wt% HMX and 5 wt% binders) using VISAR. Planar detonations were produced by impacting the explosive with projectiles launched in a 72 mm bore gas gun. Particle velocity wave profiles were measured at the explosive/window interface using two VISARs with different fringe constants. Windows with very thin vapor deposited aluminum mirrors were used for all experiments. PMMA windows provided an undermatch, and LiF (Lithium Fluoride) windows provided an overmatch to the explosive, reacted and unreacted. While the present experiments do not have adequate time resolution to adequately resolve the ZND spike condition, they do constrain it to lie between 38.7 and 53.4 Gpa or 2.4 and 3.3 km/s. Accurate knowledge of the CJ state places the reaction zone length at 35 ± 12 ns (≈ 0.3 mm). The present experiments do not show any effect of the window on the reaction zone; both window materials result in the same reaction zone length.

INTRODUCTION

According to the ZND model, the detonation process consists of a shock wave which takes the unreacted explosive from its ambient state to a "spike" state on the unreacted Hugoniot. Chemical reactions then begin and a "reaction zone" is traversed by proceeding down the detonation Rayleigh line from the spike state to the CJ state. The CJ state, most accurately described as the state at which the shock velocity and Lagrangian sound speed are equal is often thought to represent the fully reacted state. The ZND model indicates that the pressure and particle velocity decrease between the spike and CJ states, even though energy is being released by the chemical reactions in this region. From the CJ state, the explosive products expand in a Taylor wave. A very good discussion of these details can be seen in Engelke and Sheffield (Springer Verlag chapter)

Difficulties in interpreting detonation profile measurements are as follows. First, because explosives react at high shock pressures, there is no reliable unreacted Hugoniot data in the high pressure regime. The result of this is that the spike state cannot be predicted with accuracy, and it is difficult to determine if a measurement has adequate time resolution to actually measure this state. Secondly, most measurements show no distinct end to the reaction zone indicating a CJ point.

Thus, from the detonation profile measurement, one cannot obtain the CJ state. Further, because a confusing variety of CJ state estimates and explosive product Equations of State have been reported in the literature, one cannot with confidence place the CJ state on a measured profile using impedance matching techniques. Fortunately, this latter situation is changing. There is now a very reliable CJ state and reaction product EOS for PBX9501 based on overdriven Hugoniot and sound speed measurements. (Fritz Hixson Shaw and McQueen paper.)

Experimentally, detonation profile studies have been carried out using a number of different techniques. Fundamental difficulties are that inserting any kind of plate (no matter how thin), gauge, or interferometer window into (or onto) the material to try to measure the detonation profile will disrupt or perturb the flow. This is partly because the plate, gauge, or window does not react as does the explosive and partly because it is a different mechanical impedance than the explosive or its products. Obviously, thicker and more massive plates or gauges will perturb the flow more than thin ones. The second difficulty in making reliable measurements is obtaining adequate time resolution to resolve the spike point. Time resolution is dependent on two things; the resolution of the recording instrument and the thickness of the gauge or mirror. For instance, the reaction zone measurements reported by Sheffield, Bloomquist and Tarver, used

subnanosecond recording instruments (an ORVIS interferometer) but 12 – 25 μm thick mirror foils. The metal foils limited the time resolution to 10 – 30 ns. Additionally, time resolution must often be traded for precision in the measurement of the particle velocity or pressure.

In our opinion, the most fruitful experimental technique has been laser velocity interferometry. In this method, a window with a thin mirror is placed in contact with the explosive which is detonated. Laser light reflected from the mirror is Doppler shifted when detonation reaches the interface and the mirror moves. The interferometer and analysis software transform this Doppler shifted light into mirror (or interface) velocity versus time waveforms. Several studies have used this technique in various interferometer setups (ORVIS and Fabry-Perot) to estimate reaction zones in various explosives. Time resolution can range from 10 ns to less than 1 ns depending on the interferometer and recording technique. The present study is an application of the VISAR (Velocity Interferometer System for Any Reflector) to the study of detonation profiles. Because we have used very thin vapor deposited mirrors, we believe that the instrument is the limiting factor in the time resolution.

EXPERIMENTAL DETAILS

Overall Experimental Setup

The experimental setup for measuring detonation wave profiles is shown in Figure 1. Gas gun driven projectiles faced with vial (a pressed aluminum oxide ceramic) were used to obtain planar, sustained-shock inputs to the PBX9501 explosive. PBX9501 consists of (by weight) 95% HMX, 2.5% estane, and 2.5% of the mixture bis(2,2-dinitropropyl) acetal and bis(2,2-dinitropropyl) formal (BDNPA/BDNPF). Sample densities varied between 1.826 and 1.838 g/cm^3 . Impact was directly on the explosive sample which was 50.8 mm in diameter and of various thickness. Inputs for the experiments are listed in Table 1 and ranged from 3.9 to 6 GPa. Full detonations traveled 8 – 17 mm before interacting with the mirror/window. These inputs are well below any estimate of the CJ pressure in PBX9501, and thus the detonations are underdriven. In fact, these experiments were add-ons to modest pressure initiation experiments. VISAR windows were of either PMMA (polymethylmethacrylate, Rohm and Haas type II UVA Plexiglas) or LiF (Lithium Fluoride, single crystal oriented [100] obtained from Optovac).

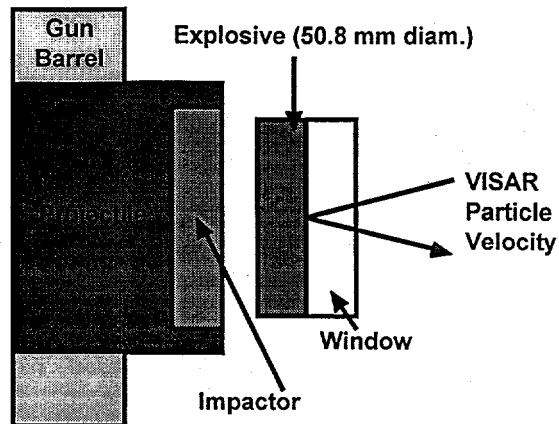


FIG. 1 EXPERIMENTAL SETUP FOR MEASURING DETONATION PROFILES IN PBX9501. VISAR WINDOWS WERE EITHER PMMA OR LiF. THE EXPLOSIVE WAS A DIFFERENT THICKNESS FOR EACH EXPERIMENT. DIFFERENT IMPACTOR MATERIALS AND IMPACT VELOCITIES PRODUCED VARYING INPUTS INTO THE EXPLOSIVE.

Window Preparation

Windows and mirrors were prepared as follows: first, the surface on which the mirror was to be made was determined to be flat using an optical flat. Some PMMA windows needed to be lapped flat and then polished back to an optical finish. Second, the diffuse mirror surface was prepared using an eraser (Faber Castell "ParaWhite" model 7041). This is a technique developed by Howard Stacy at Los Alamos. Next, about 0.4 μm of aluminum was vapor deposited on the diffuse surface. Finally, an 8 μm thick sheet of kapton was epoxied on top of the aluminum to protect it from the hot reaction products. The explosive was glued to the window using Arelhex glue. The combined thickness of all glue bonds for a typical experiment was several microns.

VISAR Setup and System Time Resolution

Interface velocity measurements were made using two VISARs set at different velocity per fringe (VPF) constants. Approximate fringe constants (VPF) for each VISAR, corrected for the window material are listed for each experiment in Table 1. The particular VISARs used in this study were made by Valyn International and were models VLNV-04 and VLNV-03. These VISARs use photomultiplier tubes to convert light intensities into electrical signals. Electrical signals were recorded using a Tektronix TDS684 digitizer, recording at either 0.4 or 1.0 ns/point, depending on the shot. The Tektronix TDS684 has a bandwidth of 1 GHz.

Table 1 Summary of Experimental Details, Results, and Analysis

Shot num.	Input (GPa)	Thickness (mm)	Window	VPF (km/s/fringe)	Measured Spike (km/s)	Fringe count	Calculated spike (km/s/GPa)	Calculated Spike is	Time to CJ state (ns)
1083	6	12.70	PMMA	0.517	3.15	6.1	2.58, 41.6	Low	25
				1.826	3.68	2.0	3.14, 50.6	High	20
1133	5.15	23.0	PMMA	1.281	3.84	3.0	3.31, 53.4	High	50
				1.917	3.77	2.0	3.23, 52.2	High	45
1134	5.17	23.0	PMMA	1.281	3.84	3.0	3.31, 53.4	High	42
				1.917	3.80	2.0	3.27, 52.7	High	35
1156	5.19	23.0	PMMA	1.281	3.83	3.0	3.30, 53.2	High	25
				1.917	3.82	2.0	3.29, 53.1	High	30
1150	3.87	26.1	LiF	0.657	2.16	3.3	2.41, 38.9	Low	19
				2.155	2.23	1.0+	2.50, 40.4	Low	20
1154	5.20	23.0	LiF	0.657	2.15	3.3	2.40, 38.7	Low	40
				2.155	2.25	1.0+	2.53, 40.8	Low	60

Several experiments were done in order to determine the time resolution of this system. Particularly useful were those in which a Cu flyer impacted a thin sapphire plate backed by a PMMA VISAR window. Sapphire is completely elastic at modest stresses, and in the above configuration the first wave propagates as a very sharp shock. The particle velocity of the transmitted shock is monitored at the interface of the sapphire and a PMMA window. (The system input is thus a sharp step function.) The system time response is the time from when the output (the recorded signal) begins to change and the time a steady level is reached. We found that if the velocity jump was recorded using a fringe constant yielding close to an integer number of fringes for the jump, the time response was about 1 ns. For example, if the jump contained 3.02 fringes a steady level would be reached about 1 ns after the first change. If the velocity jump was recorded using a fringe constant yielding, for example $3\frac{1}{2}$ fringes, the response time could be 2 – 3 ns. This is a peculiarity of PM tube based VISAR measurements and is important in evaluating the results of the present study.

RESULTS

Six successful detonation wave profile experiments were completed on PBX9501; four with PMMA windows and two with LiF windows. Figure 2 shows typical interface velocity profiles obtained with each window type. The amplitude of the wave obtained with the LiF window is smaller because this material is higher impedance. These waves have a spike state followed by a reaction zone and then a following Taylor wave. CJ states, estimated in the analysis section are also shown.

In all of the experiments with PMMA windows, the window eventually went opaque. This is almost certainly due to the chemical reaction that occurs in PMMA at

particle velocities greater than 2.9 km/s(ref). The reflected light intensity observed in shot 1156 and shown in Figure 2 is typical. Within about 50 ns the intensity is halved, and after 400 ns the intensity drops to less than 10% of the original amount. (Shot 1083 had records which were useful out to 1.3 μ s.) In experiments with LiF windows, records were useable until the shock broke out at the window free surface.

Along with the experimental parameters, Table 1 lists

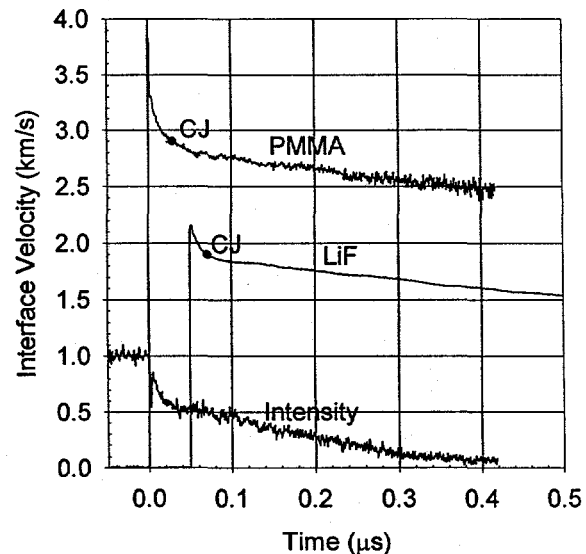


FIG. 2 TYPICAL WAVE PROFILES OBTAINED WITH PMMA AND LiF WINDOWS IN SHOTS 1156 AND 1150 RESPECTIVELY. THE LiF WAVE PROFILE HAS BEEN DISPLACED BY 50 NS. THE INTENSITY CURVE IS FOR THE EXPERIMENT WITH THE PMMA WINDOW AND ILLUSTRATES HOW DECREASING INTENSITY LIMITS THESE EXPERIMENTS.

the measured spike state obtained with each VISAR in each experiment. The fringe count for each VISAR is also listed. Note that the fringe count column shows that all experiments using PMMA windows had spike states which were equivalent to an integer number of fringes. (The 0.517 km/s/fringe VISAR record on shot 1083 is an exception.) In addition, after the initial jump, the particle velocity for these records was always declining. The integer number of fringes to the spike, and the always declining particle velocity are indications that the spike state is overestimated in these records.

With the 0.517 km/s VISAR record for shot 1083, and the LiF window shots, the recorded spike point was more than an integer number of fringes. Additionally, after the initial jump the particle velocity increased before starting to decline, all within a few ns. Because of these facts and because of the 2-3 ns time resolution of the VISAR under these conditions, the spike point is probably underestimated in these records.

ANALYSIS

In our attempt to estimate reaction zone parameters from the measured detonation profiles, we have analyzed the results in the context of the pressure particle velocity plane. Figure 3 displays the relevant Hugoniot, isentropes, and their intersections. Briefly, the spike point is determined by the intersection of the Rayleigh line and the unreacted PBX9501 Hugoniot. This is matched onto the PMMA or LiF window Hugoniot. Similarly, the product isentrope is used to match from the estimated CJ point to the window Hugoniot. This gives the points S:M and CJ:M in the particle velocity plane. These data are then used to determine the nearness of the measurement to the spike point anticipated and to determine the reaction zone time based upon when the particle velocity goes below the CJ:M condition. Details used to calculate each of the curves follows.

Detonation Rayleigh Line

In the Pressure particle velocity ($P-u_p$) plane the equation for the Detonation Rayleigh line is

$$P = \rho_0 D u_p, \quad (1)$$

where ρ_0 is the initial density (1.83 g/cm³), and D is the detonation velocity (8.812 km/s Ref. Fritz).

Unreacted PBX9501 Hugoniot

The unreacted Hugoniot (as well as the inert window materials) have the form obtained from a linear $U_s - u_p$ relation.

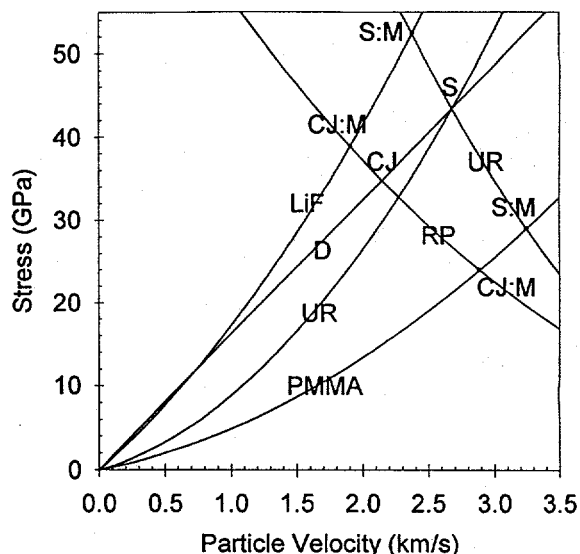


FIG. 3 HUGONIOTS AND ISENTROPES USED FOR CALCULATING REACTION ZONE PARAMETERS. THE CURVES AND STATES ARE LABELED AS FOLLOWS: (LiF) THE LITHIUM FLUORIDE WINDOW HUGONIOT. (D) THE DETONATION RAYLEIGH LINE. (UR) UNREACTED EXPLOSIVE HUGONIOTS. (CJ) THE CHAPMAN-JOUGUET STATE. (RP) THE REACTION PRODUCT ISENTROPE THROUGH THE CJ STATE. (PMMA) THE PMMA WINDOW HUGONIOT. (S) THE SPIKE STATE. (CJ:M) AND (S:M) CJ AND SPIKE STATES MATCHED ONTO WINDOW HUGONIOTS.

$$P = \rho_0 (C + S u_p) u_p, \quad (2)$$

Various reports list various values for C and S for unreacted PBX9501. Gibbs and Popalloto list $C = 2.50$ km/s, $S = 2.26$. Jerry Dick with the addition of some accurate low pressure data has found $C = 2.40$ km/s, $S = 2.39$. These Hugoniot are not very different and will pass through most of the available data up to $u_p = 0.9$ km/s.

Window Hugoniot

Eq. (2) above also describes the Hugoniot for the PMMA and LiF windows. For LiF the parameters are $\rho_0 = 2.638$ g/cm³, $C = 5.15$ km/s, and $S = 1.35$ (LASL Shock Hugoniot Data). For PMMA, the parameters are $\rho_0 = 1.186$ g/cm³, $C = 2.59$ km/s, and $S = 1.52$ (Carter and Marsh Plastics report). Strictly speaking, because a chemical reaction occurs at high pressure, we should only use these parameters for PMMA up to a particle velocity of 2.9 km/s. However we use them here because this reaction causes the PMMA to become opaque thereby

reducing our VISAR signals. Thus, while we have enough light to make the VISAR measurement, this EOS should be valid.

The Spike Point

The spike point is determined by the intersection of the Rayleigh line and the unreacted PBX9501 Hugoniot. (See Fig. 3). It lies in the neighborhood of 2.7 km/s and 45 GPa. The exact point is uncertain because it is well beyond the extent of any available Hugoniot data. Furthermore, because of the shallow crossing angle of the Hugoniot and Rayleigh line, small differences in the unreacted Hugoniot lead to larger differences in the spike parameters. With the two Hugoniot listed above, the spike parameters are 2.79 km/s, 45.3 GPa (Gibbs and Popolato) and 2.68 km/s and 43.5 GPa (Dick et. Al.). When this point is matched onto the PMMA or LiF Hugoniot as shown in Fig. 3, these disparities are magnified even further.

Using the spike point as measured at the explosive/window interface (Table 1), we have calculated the spike point in the explosive. This was done by using an unreacted Hugoniot, crossing it through the measured spike point on the window Hugoniot, and finding the intersection point on the detonation Rayleigh line. The average spike point measured with PMMA windows was found to be 3.26 km/s, 52.7 GPa. The average spike point measured with LiF windows was found to be 2.46 km/s, 39.7 GPa. For the reasons discussed in the results section, spikes measured with PMMA windows are overestimated, while spikes measured with LiF windows are underestimated. If we average these two measurements, we predict the spike to be at 2.86 km/s, 46.2 GPa. This result is in general agreement with that predicted by the unreacted Hugoniot of Dick et. al and Gibbs and Popolato. In summary, our measurement of the spike is not accurate enough to reject extrapolating either one of these Hugoniot to the ZND spike state.

Reaction Products Isentrope

The single most important item in accurately determining the CJ state and thus the end of the reaction zone is an accurate equation of state for the reaction products. Recent work by Fritz and co-workers(ref) has led to such an EOS. This EOS is based on the overdriven Hugoniot and measurements of sound speed at overdriven conditions. According to this study, the Hugoniot of the detonation products is given by,

$$\ln P_s = a + b \ln \frac{1}{V} \quad (3)$$

where P_s is the pressure on the Hugoniot, V is the specific volume, and a and b are constants equal to 0.802 and 3.096 respectively. Units are GPa and cm^3/g in this formulation.

The Gruneisen parameter on the Hugoniot was found to have the form

$$\Gamma(P_s, V) = \Gamma = 0.445 \text{ cm}^3/\text{g} \quad (4)$$

We find the isentrope, indicated by the subscript s , by rearranging and integrating the Hugoniot differential equation

$$\frac{dP_s}{dV} = \left[\frac{dP_s}{dV} + \frac{\Gamma}{2V} P \right] / \left[1 - \frac{\Gamma}{2V} (V_0 - V) \right] \quad (5)$$

The principal isentrope is the solution of equation (5) from the initial condition $P = P_{CJ}$, $V = V_{CJ}$, where the subscript CJ indicates the CJ condition. Solution of equation (5) for the principal isentrope was obtained using Mathematica® and is

$$P_s = \left\{ \begin{aligned} &be^* \left(\frac{1}{V} \right)^{1/b} \left((2+A)(2+\Gamma)V - \Gamma V_0 A \right) V_{CJ} \\ &+ \left(\frac{V_{CJ}}{V} \right)^{1/b} \left(-be^* \left(\frac{1}{V_{CJ}} \right)^{1/b} \left((2+A)(2+\Gamma)V_{CJ} - \Gamma V_0 A \right) \right) \end{aligned} \right\} \quad (6)$$

$$A(2+A)V_{CJ}$$

where $A = 2b - \Gamma$. This isentrope is transformed from the $P-V$ to the $P-u_p$ plane using

$$u = u_{CJ} + \int \sqrt{\frac{dP}{dV}} dV \quad (7)$$

where the integration is begun at V_{CJ} . Equation (7) was evaluated numerically using Mathematica®. Copious use of the results of Fritz and co-workers (ref) were used for the CJ constants.

$$\begin{aligned} P_{CJ} &= 34.8 \text{ GPa} \\ V_{CJ} &= 0.411 \text{ cm}^3/\text{g} \\ u_{CJ} &= 2.15 \text{ km/s} \\ D_{CJ} &= 8.812 \text{ km/s} \end{aligned} \quad (8)$$

After evaluating equation (7) and plotting it along with the other relevant Hugoniot in Figure 3, we find the PBX9501 product principal isentrope crosses the PMMA

Hugoniot at 2.89 km/s, 24.0 GPa, and the LiF Hugoniot at 1.91 km/s, 38.9 GPa. Using these values, we were able to determine the end of the reaction zone for each of the measured wave profiles. The time to reach the CJ state is summarized in the last column of Table 1, and varies between 19 and 60 ns, with a mean of 35 ns. The reason for this wide variance in reaction zone time is likely the shallow angle with which the CJ state is approached. The shallow angle is likely due to a slower reaction rate.

CONCLUSIONS

Reaction zone measurements in quickly reacting explosives such as PBX9501 are very challenging. From our measurements we see that the ZND spike is very narrow and short, and the steeply falling particle velocity near the spike tip is indicative of a very high reaction rate. This makes accurately measuring this state very difficult, and we are not doing exceptionally well with our PM tube based VISARs and their 2-3 ns time resolution. Perhaps an optically recorded VISAR or ORVIS system would be better.

As the CJ state is approached, the reaction slows considerably. Thus, the particle velocity approaches the CJ state at a shallow angle. Because of this, as well as the compromise in particle velocity accuracy we have made in order to get high time resolution, the time at which the particle velocity reaches CJ particle velocity is difficult to determine. Our measured reaction zone time has considerable uncertainty and scatter.

On the positive side, these experiments show that for detonations in PBX9501 there is nothing inconsistent with the ZND model. A spike state well above the CJ state is reached. This state is probably on the unreacted Hugoniot, although we neither know this Hugoniot accurately nor have we measured the state accurately. Furthermore these experiments do not give us any reason to reject (or correct) current unreacted Hugoniots for this material. Finally, these experiments do set limits on reasonable values for the reaction zone.

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