Shock initiation of the TATB-based explosive PBX 9502 cooled to 77 Kelvin

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Abstract. We present gas-gun driven plate impact shock initiation experiments on the explosive PBX 9502 (95 weight percent triaminotrinitrobenzene, 5 weight percent Kel-F 800 binder) cooled to liquid nitrogen temperature, 77K. PBX 9502 samples were cooled by flowing liquid nitrogen through a sample mounting plate and surrounding coil. Temperatures were monitored using embedded and surface mounted thermocouples. Reactive flow was measured with embedded electromagnetic particle velocity gauges. Wave profiles from the particle velocity gauges show that, even at 77K, shock initiation in PBX 9502 retains a heterogeneous or “hot-spot” character. The “Pop-plot,” or distance to detonation, $x_D$, vs. impact pressure, $P$, is $\log_{10}(x_D) = 4.9 - 3.3 \log_{10}(P)$. 

1. Introduction
Triaminotrinitrobenzene (TATB) based explosives, such as PBX 9502, are known to change shock initiation sensitivity dramatically with temperature [1]. When unconfined and heated to 250°C, PBX 9502 becomes significantly more shock-sensitive, comparable to HMX based explosives. When cooled, the shock sensitivity monotonically decreases.

Recently we reported on 26 shock initiation experiments for PBX 9502 cooled to -55°C or 218K [2]. Cooling from 23°C to -55°C causes a large reduction in shock sensitivity. For example, at 12 GPa, the distance to onset of detonation, $x_D$, is 9.1 mm at 23°C and $x_D = 14.0$ mm at -55°C. To further evaluate the changes in shock sensitivity with temperature and relate these data to chemical reaction rates, we have performed a series of gas-gun driven plate impact experiments at temperature much much less than -55°C. The lowest temperature attained was liquid nitrogen temperature, 77K or -196°C. In this paper, we describe the experiments and results.

2. Experiment
PBX 9502 samples were shock initiated by gas-gun driven plate impact. A Lexan (polycarbonate plastic) projectile faced with a flyer plate made of Kel-F 81 was launched to high velocity using the 50.8 mm bore two-stage gas-gun located at Los Alamos National Laboratory (LANL) Technical Area 40 [3]. This gun is capable of launching projectiles with masses ranging from 90 - 250 g to velocities ranging from 1.2 - 3.6 km/s. Projectile impact generates a planar shock wave beginning the initiation process.
Shock initiation of PBX 9502 cooled to liquid nitrogen temperature, 77K.

- **What is the impact of understanding the role of temperature in shock initiation?** Data for calibration or validation of temperature-aware reactive burn models.
- **How many shots were fielded on the 50 mm bore gas-driven two-stage gun?** Instrumentation is thermocouples, embedded magnetic velocity gauges, and PDV.
- **Why is it significant?** First ever shock initiation experiments at liquid N2 temperature, 77K. "Hot spots" remain the dominant shock initiation mechanism even at this low temperature. Shock sensitivity continues to be reduced at 77K.
- **Handoff:** APS SCCM paper. Data has been posted on LANL "small scale" database. It will provide a challenge for ASC reactive burn models such as WSD, CREST, Ignition & Growth, SURF+, etc.
- **What’s next?** C2 funding ended with FY13 and we’re looking for another sponsor. Other explosives (PBX 9501, TNT, Composition B, etc.) should be studied as a function of temperature.

**Figure 1.** Schematic of the experiments used to study shock initiation in a cooled explosive. The PBX 9502 sample was cooled to liquid nitrogen temperature by flowing liquid nitrogen through channels in the target mounting plate and the surrounding copper coil. The cover minimizes radiative heating of the sample.

### 2.1. Overall experimental configuration

The overall configuration for the experiment is shown in figure 1. The Kel-F 81 flyer, ≈ 6 mm thick, impacts the PBX 9502 sample at a velocity measured to within ≈ 0.1% using an optical method. The target consists of a pair of PBX 9502 wedges with an embedded electromagnetic particle velocity gauge package glued between them. A complete description of the embedded magnetic velocity gauging method is given by Sheffield et al. [4]. The gauges measure mass or particle velocity at up to 11 discrete distances from the impact face in the PBX 9502. Shock tracker gauges measure the position of the shock front as function of time.

Most of the experiments were also equipped with Photonic Doppler Velocimetry (PDV) [5] to measure detonation wave profiles after shock transit through the ≈ 23 mm thick PBX 9502 sample. Velocimetry windows included Lithium Fluoride (LiF), Poly-Methyl-Methacrylate (PMMA) and poly(chlorotrifluoroethylene-co-vinylidene fluoride) (Kel-F 800). Reflectors were either 6 µm thick Al foils (diffuse) or ≈ 0.08 µm thick vapor plated Al (specular).

### 2.2. Sample cooling

The PBX 9502 sample was cooled to liquid nitrogen temperature in two stages. The method closely follows that which we described in reference [2]. In the first cooling stage, chilled nitrogen gas from the liquid nitrogen Dewar flows through channels in the aluminum target mounting plate and the copper coil. This cools the target to ≈ -80°C. In the second cooling stage, liquid nitrogen flows through the channels in the target plate and coil. After liquid nitrogen begins flowing through the channels, cooling is very rapid; ≈ 6°C/minute. The heaters we used to reheat the nitrogen gas and thus control the cooling rate in [2] could not be used in these experiments. Passing liquid nitrogen through the heaters rendered them inoperable.

Temperature was monitored using 6 type E (chromel-constantan) thermocouples embedded in the sample and attached to the sample surface. Our temperature measuring system was not very accurate at 77K. For example in more than one experiment the terminal temperature reported by one or more thermocouples was as much as 10K lower than liquid nitrogen temperature. (We think it is impossible for liquid nitrogen to cool the sample below 77K.) In other experiments, the terminal temperature was 10K higher than liquid nitrogen temperature. To estimate final temperature, eventually we settled on observing the flow at the exit of the plumbing system. When liquid nitrogen temperature was reached, there was a switch from cold gas blowing to liquid nitrogen sputtering from the exit.

### 2.3. PBX 9502 samples

All PBX 9502 samples used in this study were from lot # HOL86A891-004. This is a recycled lot made from 50% new material and 50% material in which the TATB was reclaimed from PBX.
9502 machine cuttings, out of tolerance parts, etc. The PBX 9502 was manufactured by Holston Army Ammunition Plant, Kingsport TN, and was pressed at LANL. Further details about this lot can be found in [2, 6].

Densities of individual samples were measured at \( \sim 23^\circ C \) by water immersion. The density at 77K was then calculated using

\[
\rho(T) = \frac{\rho(23^\circ C)}{1 + (T - 23^\circ C) \alpha_V}
\]

where \( \alpha_V \) is the volume coefficient of thermal expansion. For PBX 9502 at 77K, we used \( \alpha_V = 14.2(10^{-5})/^\circ C \) as reported by Hill et al. [7]. Hill’s measurement of \( \alpha_V \) was for \( T = -55^\circ C \) and its extrapolation to 77K introduces uncertainty.

2.4. Impact condition calculations

Initial shock pressure, \( P \), and particle velocity, \( u_p \), were calculated using standard impedance matching methods [8]. These methods use the conditions of constant pressure and constant particle velocity across the flyer/target interface, equations of state for the flyer and target, and the flyer velocity, \( u_{flyer} \), to determine the impact conditions. A linear \( U_S - u_p \) equation of state was used for the Kel-F 81 flyer.

\[
U_S = C_0 + S u_p.
\]

The pressure, \( P \), in the flyer is then given by

\[
P = \rho_0[C_0 + S(u_{flyer} - u_p)](u_{flyer} - u_p)
\]

Parameters used for Kel-F 81 in equations 2 and 3 are listed in reference [6]; \( \rho_0 = 2.14 \text{ g/cm}^3 \), \( C_0 = 2.05 \pm 0.02 \text{ km/s} \), \( S = 1.65 \pm 0.01 \).

The equation of state for the PBX 9502 was taken to be the reactant EOS outlined in sections IIC and IID of Wescott, Stewart, and Davis [9]. Shock velocity, \( U_S \), was calculated using equation 38. Particle velocity, \( u_p \), was calculated using equation 39, and \( P \) was calculated using equation 40 from reference [9]. Impact conditions were determined by plotting \( P(u_p) \) for the flyer (equation 3) and \( P(u_p) \) for the PBX 9502 target and finding the intersection of the two curves.

3. Results

The result of each experiment, identified by the Shot no., is a set of particle velocity versus time, \( u_P(t) \), wave profiles measured in the explosive at 10 or 11 different distances from the impact plane (Figures 2 - 6). The position of the shock front with time, \( x(t) \), is also recorded. Table 1 summarizes the data for all experiments. The results are ordered by increasing flyer velocity, \( u_{flyer} \). The density at 77K, pressure, and particle velocity were calculated. Density at

| Shot no. | \( \rho_0(23^\circ C) \) (g/cm\(^3\)) | \( \rho_0(77K) \) (g/cm\(^3\)) | \( u_{flyer} \) (km/s) | \( P(t_0) \) (GPa) | \( u_p(t_0) \) (km/s) | \( x_D \) (mm) | \( t_D \) (\( \mu \)s) |
|---------|-------------------------------|-----------------------------|-----------------|-----------------|-----------------|----------------|----------------|
| 2s-751  | 1.889                         | 1.950                       | 2.634 ± 0.004   | 12.7            | 1.26            | 18.2 ± 2.0     | 3.06 ± 0.25   |
| 2s-656  | 1.890                         | 1.950                       | 2.815 ± 0.002   | 14.1            | 1.35            | 13.9 ± 2.0     | 2.33 ± 0.25   |
| 2s-750  | 1.888                         | 1.949                       | 2.991 ± 0.002   | 15.4            | 1.43            | 9.36 ± 0.40    | 1.48 ± 0.05   |
| 2s-655  | 1.889                         | 1.950                       | 3.229 ± 0.001   | 17.3            | 1.55            | 6.72 ± 0.40    | 1.02 ± 0.05   |
| 2s-749  | 1.886                         | 1.946                       | 3.506 ± 0.005   | 19.6            | 1.69            | 4.30 ± 0.40    | 0.64 ± 0.05   |
23°C, flyer velocity, $u_{flyer}$, and coordinates for onset of detonation, $x_D$ and $t_D$, were measured. $x_D$ and $t_D$ were estimated either directly from the wave profiles (shots 2s-655 and 2s-749) or from $x(t)$ trajectories using the method of Hill et al. [10] (shot 2s-750). For shots 2s-751 and 2s-656 detonation was not observed on either the particle velocity or the shock tracker gauges, however, detonation was observed at the back of the sample using PDV. For these two shots, $x_D$ was estimated using the particle velocity of the last wave profile.

3.1. Wave profiles from embedded gauges
Figures 2 - 6 show wave profiles for the five experiments making up this study. Each of the plots is composed of a set of ten or eleven $u_p(t)$ wave profiles, each set obtained in a single experiment. Each $u_p(t)$ wave profile in a plot was obtained from a gauge located at the distance from the impact plane indicated beside the wave profile. The shot number, the calculated initial pressure and the distance ($x_D$) and time ($t_D$) coordinates for onset of detonation are listed in the caption. Wave profiles where the PBX 9502 is detonating are characterized by a triangular shape with a peak particle velocity above 2.1 km/s. Examples are seen in figures 5 and 6 at the later times and deeper depths.

Wave profiles prior to detonation are characterized by a “shock” followed by a “hump.” Both the shock and the hump increase in amplitude as the wave travels deeper into the explosive. The amplitude increase is known as growth at the shock front and growth behind the front, respectively. Also, the time lag between the shock and hump decreases; the hump is catching up to the shock. Eventually the hump overtakes the shock marking the onset of detonation. This behavior is typical of heterogeneous explosives such as PBXs. The behavior has been observed in PBX 9502 at all temperatures, including, from this work, 77K.

3.2. The PBX 9502 Pop-plot at 77K
Figure 7 shows the Pop-plot for PBX 9502 at all temperatures thus far studied. Data from the present study are shown in blue along with associated error bars. In log – log space, a line lying above and/or to the right of another line represents a less sensitive explosive; distance to detonation is longer for the same pressure. For example at $P = 12$ GPa, $x_D = 0.9$ mm at 250°C, 5.4 mm at 75°C, 9.1 mm at 23°C, 14.0 mm at -55°C, and 22 mm at 77K or -196°C. Parameters associated with Pop-plot fits are given in table 2. Both the Pop-plot figure and the table show that the shock sensitivity decreases monotonically with decreasing temperature.

3.3. Detonation reaction zone wave profiles
Detonation reaction zone wave profiles, figure 8, were successfully measured using PDV [5] in four of the five experiments. These wave profiles should be useful for testing PBX 9502 reactive burn models such as [9].

Shots 2s-655 and 2s-656 used PMMA windows. PMMA has lower shock impedance than PBX 9502. The linear $U_S - u_p$ EOS is $\rho_0 = 1.186$ g/cm³, $C_0 = 2.59$ km/s, $S = 1.52$ [12]. The window correction factor at 1550 nm for PMMA has not been reported and was assumed to be 1.0. It is nominally 1.0 for green light, 532 nm. Shot 2s-749 used a Kel-F 800 window, which
Figure 2. 2s-751, $P = 12.7$ GPa, $x_D = 18.2 \pm 2.0$ mm, $t_D = 3.06 \pm 0.25 \mu s$.

Figure 3. 2s-656, $P = 14.1$ GPa, $x_D = 13.9 \pm 2.0$ mm, $t_D = 2.33 \pm 0.25 \mu s$.

Figure 4. 2s-750, $P = 15.4$ GPa, $x_D = 9.36 \pm 0.40$ mm, $t_D = 1.48 \pm 0.05 \mu s$.

Figure 5. 2s-655, $P = 17.3$ GPa, $x_D = 6.72 \pm 0.40$ mm, $t_D = 1.02 \pm 0.05 \mu s$.

Figure 6. 2s-749, $P = 19.6$ GPa, $x_D = 4.30 \pm 0.40$ mm, $t_D = 0.64 \pm 0.05 \mu s$.

Figure 7. Pop plots for PBX 9502 at different initial temperatures. Citations for the different data sets are given in table 2.

is a very good impedance match to PBX 9502. The Kel-F 800 EOS is $\rho_0 = 1.99$ g/cm$^3$, $C_0 = 1.838$ km/s, $S = 1.824$ [13]. The window correction factor for Kel-F 800 has not been measured and was also assumed to be 1.0. The Kel-F 800 was obtained from Afton Plastics and was Compression molded. Shot 2s-751 used a Lithium Fluoride (LiF) window, which has a higher shock impedance than PBX 9502. The LiF EOS is $\rho_0 = 2.638$ g/cm$^3$, $C_0 = 5.15$ km/s, $S = 1.35$
4. Conclusions
We have completed a series of gas-gun driven plate impact experiments to study the shock initiation of PBX 9502 cooled to liquid nitrogen temperature, 77K. Results indicate that shock sensitivity decreases monotonically with decreasing temperature. At 77K the wave profiles retain characteristics of heterogeneous or hot-spot initiation. The results should be useful for calibrating temperature dependent or temperature aware reactive burn models.

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