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SHOCK INITIATION OF PBX-9502 AT ELEVATED TEMPERATURES

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The shock sensitivity of PBX-9502 is known to change with temperature. Both volume expansion and increased internal energy may contribute to this phenomenon. PBX-9502 was heated and its initiation and detonation behavior was examined, using MIV and shock tracker gauging on a light gas gun. Sensitivity and reactive wave profiles were measured. Complementary experiments were done on PBX-9502 made to undergo "ratchet growth", or non-reversible anisotropic thermal expansion, under carefully controlled thermal cycling. This process causes noticeable size changes and significant changes in sensitivity. Sensitivity and reactive wave profiles are discussed in terms of density and microscopic material morphology.

INTRODUCTION

TATB-based explosive PBX-9502 exhibits interesting thermal behaviors that may be expected to influence the sensitivity of the material at elevated temperatures. Both morphological and chemical changes may be important in understanding the response of PBX-9502 at temperatures above ambient.

Morphological changes include alteration in size and distribution of hotspots and a decrease in initial density with increasing temperature. On heating, TATB-based explosives undergo "ratchet growth," or non-reversible thermal expansion, as a result of grossly anisotropic thermal expansion of TATB crystallites in the material. The phenomenon is complicated, with the material exhibiting several different growth regimes at different temperatures,¹ due to interactions of crystallites and binder behavior. As the crystallites and binder are expanded and redistributed, intercrystalline porosities are believed to decrease, while intracrystalline porosities are believed² to increase, both of which may be expected to influence initiation properties.

Dependence of reaction rate on temperature almost certainly increases the reaction of the TATB at elevated temperatures, although extensive experiments by Buntain³ have shown the increase in reactivity with increasing temperature to be small.

EXPERIMENTAL

Experiments are done on a single stage light gas gun, using in-material magnetic (MIV) gauging^{4,5,6} in a target specially modified to allow controlled heating and temperature monitoring.

The target is heated front and back using silicone rubber flexible heaters⁷ rated to 45W and 220°C. The entire target was insulated with household fiberglass insulation. Temperature was monitored at 4 locations including the gauge plane, using both wire and fine foil⁸ copper constantan thermocouples. Thermocouple mortality was high during heating, possibly due to expansion of the PBX material onto which the TC was fastened. One minute before firing, the front heater and insulation was jettisoned using a spring assembly, to provide an unimpeded planar surface for projectile impact. Heating rates and temperature uniformity within the target were thoroughly studied, as was cooling after ejection of the front heater. Heating was performed at between 0.5 and 1°C/minute, to prevent thermal stresses, distortion, or cracking in the PBX material.

The gas gun generates reliably well-supported shock waves with a well-characterized wave shape. The square pressure pulse simplifies consideration of the time-dependent behavior of the growth of the reactive wave. The gas gun can reach

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projectile velocities of up to 1.4 mm/usec, corresponding to pressures of up to about 10.5 GPa in full-density PBX materials when single crystal sapphire impactors are used. This maximum pressure is insufficient to detonate cold PBX-9502 within the observable time, but if the material is hot or has undergone non-reversible thermal expansion to a lower density, then the run to detonation will be short enough to provide good data. Particle velocity u_p was measured directly using ten nested magnetic gauges, and shock velocity U_s was obtained from time of arrival at the different gauges and from a shock tracker gauge.⁶

Figure 1. The hot MIV target

DISCUSSION

Heated material exhibits quite different reactive behavior from other detonable (HMX-based) PBX materials studied at these pressures. Waveforms observed in PBX-9502 at 80°C are shown in Figure 2. The mechanism for increased reactivity at high temperatures consists of two factors, temperature dependence of the reaction, and material morphology. These data confirm that significant changes in material morphologies occur between ambient and 100°C, as has been proposed to explain anomalous thermal expansion and contraction data¹.

Between 25°C and the pressing temperature of the material, which varies between 80°C and 100°C, TATB crystallites undergo thermal expansion to reoccupy the voids created when they cooled and contracted after pressing. This process produces some net volume expansion, but is

generally recognized² to result in a decrease in porosity of the PBX material. Above the pressing temperature, further expansion of TATB crystallites pushes them apart. This "graphitic" growth along with increased flow of the binder increase the void fraction as the net volume increases rapidly. Porosity discussed here refers only to intercrystalline voids. It is anticipated that intracrystalline voids increase uniformly with temperature, but data is inconclusive.

Figure 2. MIV gauge records for PBX-9502 reacting at 80°C. Baseline drift due to an electronic artifact that has been corrected.

Figure 3. The x-t diagram drawn from experimental data shows the overtake of the shock () by the superdetonation. ()

Material near the pressing temperature can thus be expected to have quite low intercrystalline porosity, resembling a very dense solid or a liquid. The development of detonation in the absence of hot spots relies on homogeneous initiation,⁹ in which the reactive wave or "superdetonation" arises in the hot shocked material after the shock front has passed, and then accelerates to join the shock front as a full detonation wave. The process is chemical, rather than mechanical. This "superdetonation" is clearly visible, marked "A" in the u_p records shown in Figure 2. The location of the reactive wave maximum is measured in each record, and the position of this wave is shown in the $x-t$ diagram shown in Fig. 3. Further examples of this behavior in liquids may be found in the literature.¹⁰

At higher temperatures, the (intercrystalline) void fraction increases, restoring the material to a typical porous PBX, and the initiation to a homogeneous mechanism. The data shown in Figure 4 was taken at 100°C, and shows u_p profiles typical of heterogeneous initiation, comparable with that of room temperature HMX-based PBX materials.

Figure 4. Detonation behavior of PBX-9502 at 100°C.

These records are consistent with the expectation that the material morphology is altered in different ways as the material passes through different temperature regimes, and that detonation behavior depends strongly on the material morphology, specifically on intercrystalline voids.

A second explanation should be considered for these data. The detonation behavior of TATB-based materials has been suggested to be homogeneous, because the void size is small relative to the reaction zone of the TATB. In this case, the 80°C data may be viewed as an extension of the room temperature behavior. The records obtained at 100°C may be anomalous, resulting from microcracking during heating. This kind of microcracking has been seen¹¹ in a few of many identical PBX-9502 samples subjected to the same very controlled heating. Microcracking has a marked effect on the growth of the reactive wave. A larger number of experiments will assist in distinguishing between these possibilities.

The apparent irrelevance of intracrystalline porosity to the initiation mechanism lends some support to the assertion that small voids do not assist in the hot spot initiation of the PBX-9502. However, the true behavior of intracrystalline voids with temperature is not well characterized.

The chemical contribution to the increase in reactivity at these elevated temperatures may be evaluated by comparison of the run distance for PBX-9502 at reduced density with the run distance vs. run time points for these heated samples. Plots are shown in Figure 5.

Figure 5. The run distance for the heated PBX-9502 sample (*) is comparable to the run distance for PBX-9502 at initial density of 1.825 (◇), but differs from the run for cold PBX-9502. (□)

The points for reduced density material were obtained from shots done on cold PBX-9502 that had been subjected to ratchet growth by repeated heating in an oven to 211°C, cooling very slowly to room temperature in between cycles. The resulting samples were then machined into MIV wedges and fired cold on the gas gun. Wackerle and Dallman² show that the sensitivity of material expanded in this way does not differ from material pressed to the same reduced density, ruling out chemical decomposition during the heating cycles.

The density of the 80°C sample can be estimated^{1,2} to be 1.848, compared with 1.825 for the expanded material, allowing for the possibility that its reaction may be slightly accelerated by temperature in order for its run distance to fall with that of lower density material. Points for both hot and expanded PBX-9502 fall on the Hugoniot given by Dallman and Wackerle² for PBX-9502 at 75°C.

CONCLUSION

The rate of chemical reaction of TATB is increased with temperature, since the 80°C sample exhibits a shortened run distance despite the decreased porosity of the sample and the homogeneous initiation mechanism, which is independent of hot spots.

In the temperature regime above the pressing temperature, porosity increases and hot spot density and activity dominate the reaction, masking bulk thermal effects on chemical rate. The initiation mechanism in this case is heterogeneous.

The observation of increased chemical reactivity supports Wackerle and Dallman's² proposal that those hot spots that are too small to cause significant reaction behind a given shock will, above a certain temperature, become effective and contribute to the initiation of the explosive.

The limits on the temperatures and porosities at which PBX-9502 will exhibit homogeneous initiation behavior will contribute interesting data to the understanding of hotspot reaction, yielding data on the interaction of chemical reaction rate and hotspot effectiveness.

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TATB Materials have Complicated Thermal Behaviors

- * ratchet growth
- * non-Arrhenius sensitivity increase
- * sensitivity decrease at high T

Morphology and Chemical Rate both affect Initiation of Hot Explosive

MIV Experiments are Done on the Gas Gun

- * planar shocks of long duration
- * u_p to 2%
- * well-known initial states
- * reproducible experiments
- * exact impact velocities

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Densities and Porosities can be Estimated

25oC

80oC

100oC

PBX 9502 VOID PERCENT

Howard Cady's extensive work

Homogeneous and Heterogeneous Initiation are distinguished in MIV records

Homogeneous

Heterogeneous

PBX 9502 Initiation has Homogeneous character at 80°C

Morphology at 80°C may Promote Homogeneous Initiation

80°C : few voids : homogeneous initiation

25°C : big voids : heterogeneous records

Homogeneous Initiation may be typical of
PBX 9502 at Any Temperature

HMX

x reaction zone $<$ x hotspot

TATB

x reaction zone $>$ x hotspot

Initiation at 100°C looks Heterogeneous

* Morphology causes
changes in response below 100°C

Chemical Response changes with Temperature

* unmasked by hotspot activity

* reaction accelerated,
after correcting for density

Mechanism of Reaction Varies between 25°C and 100°C

- * dependent on pressing temperature
- * chemical reaction
increases with temperature
- * porosity minimum may unmask
chemical bulk rate effects