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DEFLAGRATION TO DETONATION EXPERIMENTS IN GRANULAR HMX¹

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ABSTRACT

In this paper we report on continuing work involving a series of deflagration-to-detonation transition (DDT) experiments in which we study the piston-initiated DDT of heavily confined granular cyclotetramethylenetetranitramine (HMX). These experiments were designed to be useful in model development and evaluation. A main focus of these experiments is the effect of density on the DDT event. Particle size distribution and morphology are carefully characterized. In this paper we present recent surface area analysis. Earlier studies demonstrated extensive fracturing and agglomeration in samples at densities as low as 75% TMD as evidenced by dramatic decreases in particle size distribution due to mild stimuli. This is qualitatively confirmed with SEM images and quantitatively studied with gas absorption surface area analysis. Also, in this paper we present initial results using a microwave interferometer technique. Dynamic calibration of the technique was performed, a 35Ghz signal is used to increase resolution, and the system has been designed to be inexpensive for repeated experiments. The distance to where deformation of the inner wall begins for various densities is reported. This result is compared with the microwave interferometer measurements.

INTRODUCTION

DDT tubes, initiated thermally or by low-speed impact of a piston, have been used for years to study the DDT process in granular explosives (Griffiths and Grocock 1960; Korotkov, Sulimov et al. 1969; Bernecker, Sandusky et al. 1981; Samirant 1981; Bernecker, Sandusky et al. 1985; Samirant 1985; Bernecker 1986; Bernecker 1989; Samirant 1989; Luebcke, Dickson et al. 1995; Luebcke, Dickson et al. 1996). Much has been learned concerning the DDT process in granular materials from these experiments, however significant gaps remain between the desired data base and available data for model development and evaluation.

The run-to-detonation can be a strong function of density (or porosity), and is therefore of significant interest. The effect of density on run-to-detonation for granular HMX was examined in the early experimental study of Griffiths and Grocock (1960). For several reasons, however, these data are of only qualitative value. Other data also have been reported for run-to-detonation as a function of density (e.g., Korotkov, Sulimov et al. 1969; Luebcke, Dickson et al. 1995; Luebcke, Dickson et al. 1996), but thermal ignition was used and granular HMX was not considered. Interestingly, some recently reported data do not show a U-shaped density dependence in granular CP (Luebcke, Dickson et al. 1996).

Because of the complex nature of the ignition, and early conductive and convective burning in thermally ignited DDT tubes, piston-driven experiments have been used here at Los Alamos National Laboratory (LANL) (McAfee, Asay et al. 1991) and elsewhere (Sandusky 1983). In the LANL experiments a combustion-driven piston ignites particles in the granular bed by compaction, which starts all the processes in a single plane at one end of the tube. These experiments were heavily diagnosed using a variety of pins, transducers, and x-radiography and provide a detailed picture of piston-initiated DDT for a narrow set of conditions (e.g., only a single density was considered). Although these results

give a detailed picture of the DDT event for the conditions considered, little information is obtained concerning the effect of several parameters that are known to be important, including the effect of density. This makes the development and evaluation of improved models difficult. This effort is an attempt to address this issue.

In this paper we present further characterization of the granular HMX used, initial calibration and use of a microwave interferometer technique, and further DDT experiments.

EXPERIMENTAL

The apparatus used in the DDT tube experiments is shown schematically in Fig. 1. The DDT tube used was a thick walled 77.6 mm o.d. stainless steel 304 tube with a 96.5 mm length and 6.4 mm i.d. The 143:1 inner to outer cross sectional area provided enough inertial confinement to contain the DDT event radially. The piston was loaded into the bottom end of the tube and set flush with the bottom surface. HMX was incrementally loaded directly into the tube in 3 mm lengths, and a lexan plug was set in the end to temporarily confine the bed.

The ignitor block was made of vascomax 250 maraging steel. A PYROFUSE® was set into the bottom of the chamber with a thin mixture of titanium and boron, a gasless pyrotechnic, covering the bridge wire. A load of loose HMX in the burn chamber was ignited by the burning titanium and boron and the burning HMX powder raised pressures enough to drive the piston into the bed of HMX leading to initiation. A pressure transducer (PCB 109A02) was mounted into the side of the burn chamber, and in the first two tests, side closure pins were positioned at four locations near the bottom of the tube. The mating lip between the ignitor block and main tube was sealed with a thin copper washer to prevent

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Authors' URLs: <http://www.et.byu.edu/~burnsidn>, <http://steve.son.org>, <http://sonhp.lanl.gov/asay/blaine.html>

venting, and the entire assembly was held together with four one inch thread bolts. A small hole in the upper confinement plate allowed visual inspection of tube for residual explosive

Fig. 1 Diagram of DDT tube experiment.

before disassembly. On the four later shots, a capped pin was set in the top of the tube as an indicator of when the detonation had reached the end of the tube. All pressure data were collected using a pair of LeCroy 9400A Dual channel oscilloscopes. The PYROFUSE® was ignited by a low voltage CDU, and the capped pin data were collected by a time interval meter.

{add background here??} A composed 35GHz Microwave interferometer was used to determine detonation speed and position. In various experiments. The interferometer apparatus consists of two oscillators (8 GHz and 27GHz), mixed to get an overall frequency of 35 GHz. The output signal is guided to the experiment through a waveguide (rolled teflon). The top plate of the experiment has been modified to have a conical hole to minimize reflections in the transition from the waveguide to the circular hole of the DDT tube. The only part of the microwave system not reusable is the teflon waveguide which minimizes shot-to-shot costs. A critical aspect of this diagnostic is the measurement of the dielectric constant for the materials used. This was measured

Fig. 2 Diagram of microwave interferometer.

dynamically using a detonation in various densities. This is presented in a latter section.

To examine the effect of pressing, eight samples of Class A HMX were prepared starting with poured density, about 64% TMD, and increasing by increments of 5% TMD from 65% TMD to 95% TMD (100% TMD=1.903 g/cc). To reduce density gradients, the samples were pressed at 3 mm increments in a 0.25 in diameter die, and samples were removed from the die after every three increments. Samples that formed pellets (above 80% TMD) due to high density pressing were carefully deconsolidated to powder by hand. The pressures needed to deconsolidate the pellets were very small compared to pressures experienced in the pressing procedure, and were therefore considered to have little effect on particle characteristics. Butler et al. (4) made this assumption for low density sugar samples, but did not test higher density samples (>72.2% TMD).

Particle size analysis was done using a Coulter LS 230 Particle size analyzer which uses light scattering of particles to measure size distributions. Samples were taken from solutions of about 0.1 g HMX in a bath of 10 ml of distilled water. Because of quick settling of the larger particles, a magnetic stir bar was used to obtain samples representative of the entire distribution. Samples of approximately 1 ml were quickly transported from the solution to the particle analyzer using a dropper.

Two sets of experiments were performed with the particle size analyzer. In the first set, the 8 samples of HMX which had been pressed then deconsolidated were analyzed for particle size distributions. In the second set of experiments, HMX from the same batch of samples was first put into a low power ultrasonic cleaning bath, 1.8w/in² at 48kHz, (7) for one minute, then introduced into the analyzer. A magnetic stir bar was once again used to ensure uniformity in the samples.

Surface area analysis was performed on the samples using a Quantachrome AUTOSORB-1 Surface area analyzer, which measures quantities of gas adsorbed and desorbed on a solid surface. This instrument performs a multipoint Brunauer-Emmett-Teller (BET) analysis using nitrogen as the adsorbate.

Finally, structure of the HMX was qualitatively analyzed using a Scanning Electron Microscope (SEM). Samples were observed and photographed extensively. These images are available at: http://sonhp.lanl.gov/sem_jpg.

PARTICLE CHARACTERIZATION

Works by Elban et al. (1) and Coyne et al. (2) have focused on the compaction process of very coarse (~900 μm) granular HMX, and have found fracture at very low pressures. Hardman et al. (3) also observed fracturing of other granular material at low pressures. Because of sample consolidation at high densities, however, many past studies have very little particle characterization of high density samples (4). In this section we present characterization of the 8 pressing densities considered, beginning with the original material.

Unpressed HMX

The original unpressed HMX (a typical particle is shown in Fig. 1) shows well formed crystal structures with

identifiable facets, and few cracks or flaws. Particle size analysis at this density shows that the unpressed HMX has a mean particle diameter of 193 μm (see Fig. 2). The largest volume percent of the sample is grouped around 178 μm with a relatively low volume of small diameter particles. A fairly good comparison of this measured size distribution with a sieve analysis was achieved.

Pressed HMX

Figure 3 shows a typical HMX crystal after having been pressed to 70% TMD. Large cracks run throughout the structure of the crystal, however, the bulk of the original particle clings together. Particles at 80% TMD, see Fig. 4, are heavily damaged with increased evidence of fracturing and shearing. Finally, particles at 90% TMD, Fig. 5, are crushed to fine pieces which cling together in larger agglomerates of about 100 μm

As seen in Fig. 2, the mean particle diameter decreases with increasing density from 192 μm to 131 μm at 95% TMD. With increased pressures due to high-density pressing, many of the particles are cracked and sheared, leaving a much larger volume percent of particles in the 20 μm to 40 μm region. It does appear, however, that although highly fractured, (see Fig. 3) a large volume of the particles cling together, leaving the distributions of even the high density samples in the 100 μm to 180 μm range.

Sonication Effects

One minute of sonication showed little effect on the mean particle diameter of the original HMX (Fig. 6). Because of the low power of the ultrasonic bath, and the unfractured state of the particles, the distribution was almost unchanged. (Compare "original" distributions from Fig. 2 and Fig. 6). As density increases, however, and the state of the particles becomes increasingly fractured, the effects of sonication become apparent. Distributions of samples from 65% and 70% TMD show increasing volumes of particles in the <100 μm range. At 75% TMD many of the large, but fractured particles are deconsolidated by the mild stimulus and we begin to see a transition to a bimodal distribution between 40 μm and 180 μm . This new 40 μm mode becomes more prominent with increasing densities until the 180 μm distribution completely disappears, and the remaining distribution tends toward 40 μm .

BET Surface Area Analysis

Increasing density due to pressing results in highly fractured particles with an increased surface area. Surface area per volume analysis, using the AUTOSORB-1, shows that surface area increases approximately linearly with density.

By assuming spherical particles, the size distributions obtained with the light scattering particle analyzer (Fig. 2) were converted to total surface area per unit volume, and plotted along with the AUTOSORB-1 data. Because of the mechanism used by the size analyzer, however, it is incapable

of detecting fine cracks in fractured particles (see Figs. 3-5), thus underestimating the overall surface area. The data, however, display a linear increase with surface area as also seen in the AUTOSORB-1 data. A similar conversion using the size distributions from the sonication experiment (Fig. 6) is also plotted on the same axis. As seen, the deconsolidation of fractured particles by sonication shifts the conversion closer to the actual surface area analysis. It is also nearly linear with TMD, except at low TMD.

FIGURE 4. HMX 80 % TMD (bar=10 μm).

DDT TUBE EXPERIMENTS

Table 3 lists the DDT tube experiments performed and list the conditions considered. Figure 6a is a photo of a cross-sectional view of the base case (shot #1). There is a change from dark smooth walls to lighter walls, followed by darker pitted walls downstream. The color changes are visible in the photo, but are more identifiable visibly. Past analyses at LANL and elsewhere (see Luebcke, Dickson et al. 1996) have correlated the color changes to events in the transition process by comparing pin and slit-camera records with color changes on recovered walls. For example, the first dark to light change appeared to correspond to the intersection of a rearward traveling wave (driven by the detonation) and the initial burning wave in DDT experiments (Luebcke, Dickson et al. 1996). The second color change (light to dark) corresponded to the onset of detonation. Our analysis uses both color change and wall profile to define points that can be correlated to the DDT process (see Fig. 6b). Light intensity analysis with a reflective light microscope was used to accurately define the point of transition between light and dark regions, while digitized data from scanned images of the cross sections yield the wall profile. The wall profiles obtained have many of the same characteristics as previous thermally ignited tubes (Luebcke, Dickson et al. 1996). One distinguishing feature of these tubes is the widely expanded region followed by a region where the expansion decreases. This likely indicates that detonation occurred in the pre-compacted region, producing higher pressures, as well as, in the initial low density material. This scenario is consistent with highly diagnosed DDT experiments by (McAfee, Asay et al. 1991) who, under similar conditions, observe detonation beginning within the material compacted by the initial compaction wave. This indicates there are some qualitative

differences between thermally ignited and piston-initiated experiments.

Closure pin data was obtained near the initial location of the piston in some of the experiments. This is shown in Fig. 7 for shots #1 and #2. There was a long ignition delay (about 400 ms compared with delays <100 ms in the other experiments) due to tight packing of the titanium and boron in the ignitor block. This delay was probably accompanied by some burning in the chamber which pushed the piston into the bed slightly. Nevertheless, by shifting the initial time to correspond to the closure of the first pin in both shots #1 and #2 reasonable comparison between the two sets of pins is obtained. A wave speed of about 300 m/s is observed which probably corresponds to the initial compaction wave. The ignition delay, and probable small movement of the piston from its original position before full ignition appeared to shift identifiable points further downstream in the tube by 3-4 mm consistently. This is seen in Table 4 where the identified points are listed for the six experiments reported here.

Burn chamber pressure was also measured in some of the tests (see Fig. 8). The pressure rises quickly as the loose HMX burns in the chamber. The pressure attained in the high chamber load (shot #4) is slightly more than a factor of two higher than the base case (shot #1). The pressure is slightly higher in the 85% TMD case (shot #6) than in the base case (shot #1) due to the increased resistance to the movement of the piston by the higher density bed. Also shown in Fig. 8 is the time when detonation was indicated by a closure pin near the end of the tube.

An important objective of this work was to have a very repeatable experiment. Consequently, we repeated shot #1 as closely as possible (shot #3) and compared the results. We found that the two experiments were very close as indicated by the wall deformations reported (see Fig. 9) and other points of reference (see Table 3). These results are an indication that the experiments are fairly repeatable. Additional tests will hopefully verify this further.

Figures 10a,b summarize the results obtained so far. Figure 10a shows the location of the first color change and Fig. 10b shows the second color change for the six cases. These color changes have been correlated to the transition event, as discussed above. It is clear that increasing the burn chamber pressure (piston speed) yields a much shorter run-to-detonation, as expected. A slight decrease is observed at 75% TMD compared with 65% TMD, however, at 85% TMD there is a significant increase in the run-to-detonation. Although more data is needed to better define this curve, results appear qualitatively similar to those obtained by (Griffiths and Grocock 1960). Interestingly these results were obtained in piston-initiated DDT experiments that circumvent the initial convective burning mode. Convective burning issues have often been used to explain much of the U-shaped plot, however, convective burning is very likely irrelevant in these experiments. Increasing the density yields more rapid pressurization because more explosive is available in a given volume leading to a shorter run-to-detonation. There is a limit, however, after which further increases in density inhibits the formation of ignition sites. This type of effect has been explored to explain plug formation (Son, Asay et al. 1995).

SUMMARY

Initial results from a new series of experiments in heavily confined granular HMX were presented. These experiments were designed to be useful in model development and evaluation. The effect of density (3 densities so far), piston speed (2 speeds), and ignition delay on the DDT event have been considered. We have measured the particle size distribution of the original granular explosive and pressed (higher density) samples. Scanning electron microscope (SEM) pictures were presented for the materials used. These pictures were used to interpret the size distribution measurements and to more fully characterize the initial condition of the materials used. The size distribution changes significantly and the particles become increasingly fractured with pressing. Further, we found that sonication can significantly affect size distribution measurements. This effect becomes more apparent for the more fractured materials (higher pressed densities). For the base case (65% TMD) two DDT experiments were performed yielding nearly identical results. This indicates that reproducibility was good. Run-to-detonation, as indicated by color change and deformation of the on the inner wall, for the three densities (65%, 75%, and 85% TMD) considered show a slight decrease in run-to-detonation going from 65% to 75% TMD and a significant increase at 85% TMD. Increasing the piston speed decreased the run-to-detonation. The effect of the ignition delay (tighter packing of titanium and boron) extended the run-to-detonation.

Surface area analysis of class A HMX shows a nearly linear relationship between density and surface area per volume. Converted particle size analysis, however lacks the ability to account for fractures which leads to an under-prediction of the surface area. The increase of nearly an order of magnitude in the AUTOSORB-1 data corresponds to widespread fracturing and breaking of HMX particles by pressing. These results have significant implications on the modeling used to describe the burning and transition to detonation of granular HMX.

Several additional experiments are planned. The initial density will be varied over a broader range to better define the "U-shape" run-to-detonation plot. A few thermally ignited experiments, using the same tubes used here, will be performed to further examine the differences from piston-initiated experiment. RDX is very similar to HMX, but burns very differently. For example, the "stand-off" distance of gas phase reactions in burning HMX is much larger than for RDX, at least at low pressures. To investigate this effect on the DDT event, RDX (with nearly the same size distribution) will be used in place of HMX also. As we press the granular HMX we are changing the density as well as the particle size distribution. To separate these two effects we plan to press to a higher density, deconsolidate the material and test the pre-pressed material at our standard low-density (65% TMD). To better understand the effect of particle size on DDT, nearly monosized distributions of coarse and fine HMX particles will be tested. These experiments will use the same HMX as planned for use in gas gun experiments. Also, to determine the significance of wall effects, similar tests will be run with

tubes of larger i.d., while longer tubes may be implemented to confirm the development of a steady detonation wave. There remains some ambiguity concerning the interpretation of the pin and wall records. To clarify these issues we plan to use microwave interferometry to continuously measure piston velocity (at least part of it), compaction wave, and finally the detonation wave.

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FIGURE 1. Unpressed HMX (bar=10 μ m).

FIGURE 2. Particle size distributions of pressed HMX (No sonication).

Figure 3. HMX 70% TMD (bar=10 μ m).

FIGURE 4. HMX 80 % TMD (bar=10 μ m).

FIGURE 5. HMX 90% TMD (bar=10 μ m).

FIGURE 6. Particle size distributions of pressed HMX after one minute of sonication.

FIGURE 7. Surface area data taken with AUTOSORB-1 gas adsorption unit. Coulter LS data converted from particle size to surface area assuming spherical particles. Earlier data taken with Quantasorb and Microtrac systems.

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