PARTICLE CHARACTERIZATION OF PRESSED GRANULAR HMX*

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It is widely accepted that particle size and morphology in granular beds of HE plays a large role in combustion and detonation events. This work reports the characteristics of coarse granular HMX (Class A) at a range of densities from stock density to 95% TMD. We report measurements of the particle size distribution of original granular HMX, as well as the size distribution of pressed (higher density) samples. Scanning electron microscope (SEM) pictures are presented and are found to be useful in interpreting the size distribution measurements of the granular HMX, as well as in helping to more fully characterize the state of the particles. We find that the particle size distribution changes significantly with pressing. Particles are observed to be highly fractured and damaged, especially at higher pressed densities. Also, we have found that sample preparation can significantly affect size distribution measurements. In particular, even short duration ultra-sonic or "sonication" treatment can have a significant effect on the measured size distributions of pressed HMX samples. Surface area measured by gas absorption is found to be much larger than inferred from light scattering.

INTRODUCTION

It is widely known that damaged explosives can be more sensitive to initiation than undamaged materials. Granular explosives have often been used as a simulant of damaged explosives because it is far easier to characterize the materials than actual damaged explosives and the "damage" is isessentially uniform. However, little material characterization is generaly reported in studies that use granular explosives, such as deflagration-to-detonation transition (DDT) experiments. This lack of characterization makes modeling and interpretation of the experiments difficult. Further, very little is known about how particle size changes with compaction processes, even for quasi-static pressing.

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).

As a part of the explosives safety program at LANL our group has worked to develop models to describe the DDT of granular HMX explosives. A main goal of this effort is to develop truly predictive models. DDT experiments by McAfee *et al.* (5) and Burnside *et al.* (6) have extensively used the same batch of Class A HMX as examined in these experiments, however particle characterization is limited for this material. In this work we try to address this void.

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EXPERIMENTAL SETUP

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/in^2 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.

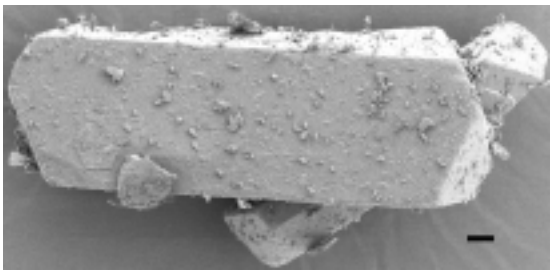
Finally, structure of the HMX was qualitatively analyzed using a Scanning Electron Microscope (SEM). Samples were observed and photographed extensively. These images are available at: http://sonhp.lanl.gov/sem_jpg.

RESULTS AND DISCUSSIONS

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



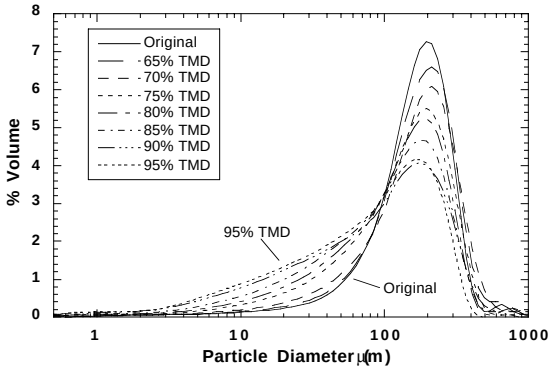


FIGURE 2. Particle size distributions of pressed HMX (No sonication).

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.

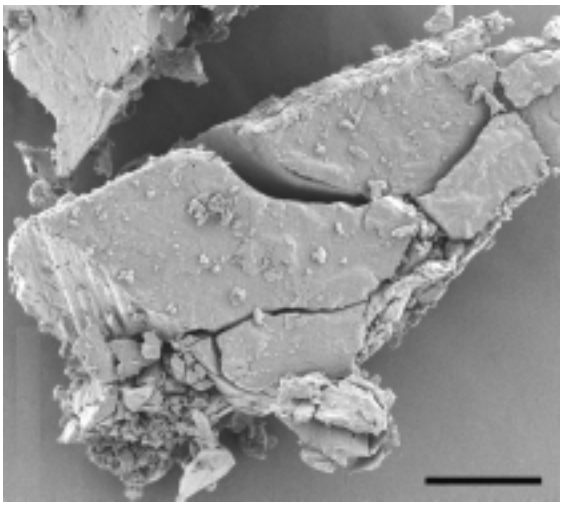


Figure 3. HMX 70% TMD (bar=10 μm).

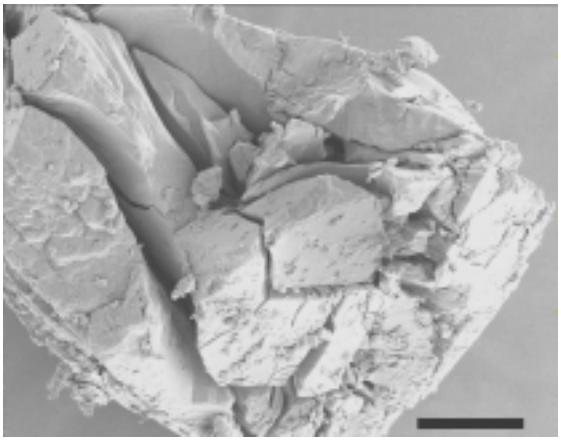


FIGURE 4. HMX 80 % TMD (bar=10μm).

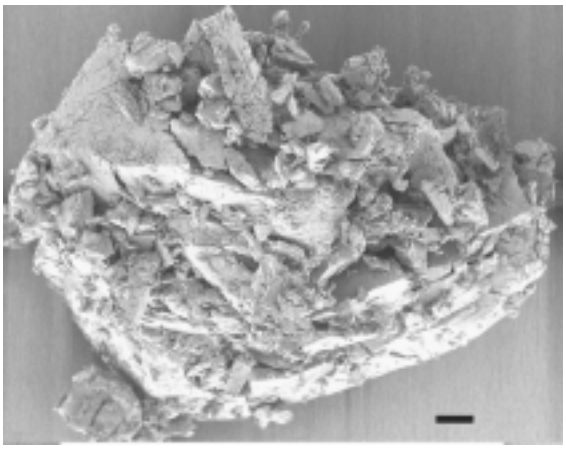


FIGURE 5. HMX 90% TMD (bar=10μm).

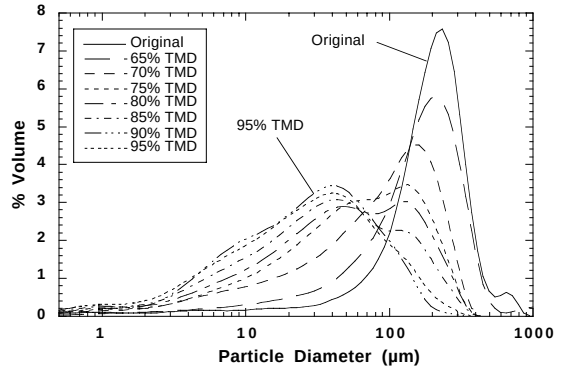


FIGURE 6. Particle size distributions of pressed HMX after one minute of sonication.

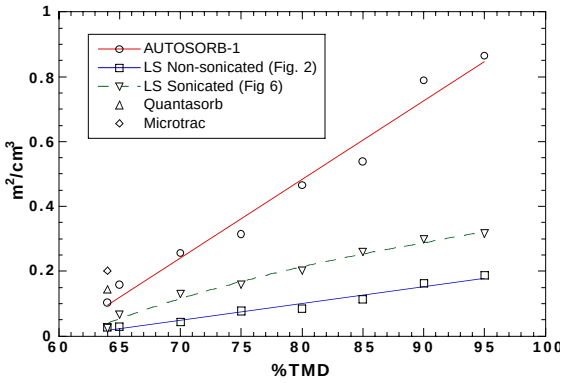


FIGURE 7. Surface area data taken with AUTOSORB-1 gas adsorbtion unit. Coulter LS data converted from particle size to surface area assuming spherical particles. Earlier data taken with Quantasorb and Microtrac systems.

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.

CONCLUSIONS

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.

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