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The Relationship between Shock Sensitivity and Morphology in Granular RDX*)

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Abstract: It is known that batches of the secondary explosive RDX from different manufacturers show significant variation in their shock sensitivity. No obvious correlation between shock sensitivity and either chemical composition or morphology has previous been identified which explains this. We use a range of techniques to study the microstructure of RDX crystals and the bulk morphology of granular beds in order to assess which hotspot mechanisms tend to be dominant. Crystals were characterized using mercury porosimetry, environmental scanning electron microscopy (ESEM) and optical microscopy. This range of methods yields quantitative and qualitative data on internal void size and number and surface structure. Shock sensitivity is quantified using small-scale gap tests, and this demonstrates the clear differences in sensitivity between batches from different manufacturers. The samples used are from three manufacturers, produced by both the Woolwich and Bachmann processes, and all have an average particle size of approximately 1200 μm.

Keywords: RDX, gap tests, sensitivity, morphology

Introduction

Explosives can be found in many different forms such as liquids, pressed powder compacts, polymers, and two-phase mixtures such as polymer-bonded explosives. These can be divided into homogenous and heterogeneous materials and this division provides a basis for describing the details of their response to mechanical stimuli. A homogenous explosive is simpler to understand since

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it is a continuous medium; each region has the same material properties as those adjacent to it and parameters such as temperature, pressure and density vary continuously across the bulk. In general the mechanisms of ignition and initiation in these materials are well-understood. For example, ignition in a liquid containing a number of bubbles is likely to start as the bubbles collapse to form hotspots [1].

Most commonly used explosives fall into the second category. Solid explosives such as RDX and HMX perform very well, releasing a considerable amount of energy per unit volume and having high detonation pressures and velocities. However, they are produced in a crystalline form and are usually either pressed into high-density compacts or are mixed with a polymer binder to produce a polymer-bonded explosive. In either case, the heterogeneities in the system make the details of ignition or initiation much harder to understand than is the case for homogenous explosives. These heterogeneities lead to a highly non-uniform temperature and stress field in the shocked material, and it is the highest-temperature regions within this distribution which will form critical hotspots [2] and lead to the reaction of the bulk.

The commonly used secondary explosive RDX is produced by many different manufacturers and it has been found that there are considerable variations in its sensitivity [3]. The batches are generally found to be chemically identical and so the explanation for these differences seems to lie in the crystal and bed morphology. In this research, four batches of RDX of a single size from three different manufacturers were examined. Their sensitivity to shock was assessed using a small-scale gap test, and their morphology was examined using a wide range of techniques. The exact processes causing ignition and initiation in granular explosives are not yet completely understood and so this study should shed light on these details. We have the opportunity to compare a wide range of large crystals with varying morphologies.

The processes which occur when a shock wave passes through a granular material do not depend only on the features of individual crystals. A granular bed consists of individual particles, supported by each other, with a small number of contact points per particle. Low level stresses are not supported by all the particles equally, but are transmitted by force chains which depend on how the particles fit together. Considerable work has been done on the structure of granular materials and how they transmit stress, but almost all of it relates to the quasi-static regime. During a shock, there will be a qualitatively different response [4] which will significantly modify the material behaviour, with the result that quasistatic studies may be of limited use.

In the past, many studies have been done to try to link particular RDX particle features, for example closed internal pores, to sensitivity. There has been some success [5-8], but a complete explanation has not been found. It is possible that part of the explanation relates to how the particles interact with each other through the contact points and not only how particles respond individually. In general, the mechanisms which occur will contribute to the inhomogeneity of the temperature field and critical hotspot formation are friction, visco-plastic work, jetting, and adiabatic compression of gas-filled pores [9].

The approach that will be taken here is to examine the morphology in a general sense, including how particles are arranged and how they interact as a bulk material, as well as studying the features of individual particles.

It should be noted that the sensitivity differences between different batches of RDX have mostly been observed for material cast in a polymer binder. We have chosen to test the sensitivity of free-poured material in order to study the simplest possible system, and to investigate how much the reduced sensitivity effects are due to the behaviour of the as-received material and not due to interaction with the binder or damage caused by pressing.

Experimental Methods and Results

Samples

Four batches of granular RDX from three different manufacturers in a single size class were acquired. All the batches had an average size of 1200 μm and the complete size range was 600-1500 μm. Laser particle size analysis confirmed the size distributions. The details of the different crystal morphologies are discussed in subsequent chapter.

To make the samples, as-received crystalline material was poured into the confinements in small increments and tapped. This resulted in a very reproducible porosity for each sample batch. For the purposes of this paper, samples are labelled in order of sensitivity within each size class, with (1) being the most sensitive material.

Sensitivity tests

Shock sensitivity was measured using a small scale gap test [10] (see Figure 1). The detonator generates a reproducible shock wave which is then attenuated by a PMMA gap. By conducting tests with various gaps, the “critical gap” can be found – the largest gap (corresponding to the lowest pressure) at which ignition will occur.

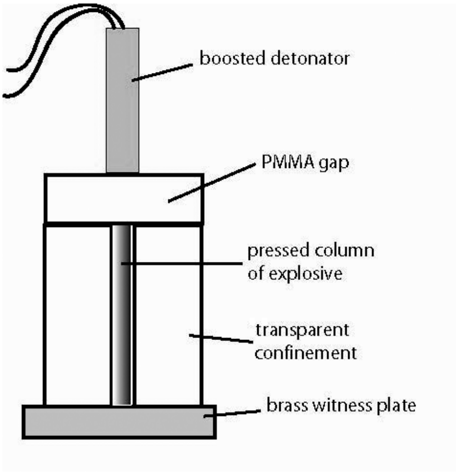


Figure 1. Diagram of the small-scale gap test. The charge was 9 mm in diameter and 25 mm long, contained within a 25 mm diameter PMMA confinement. The shock pressure from the detonator alone was approximately 15 GPa.

The gap test results are shown in Figure 2. It can be seen that there are significant variations in shock sensitivity and that some samples require twice the input pressure of others in order for reaction to start.

The criterion that must be satisfied for a "go" result in a gap test is that part of the charge detonates. In these materials, almost all the "no go" samples ignited, but did not transition to a detonation. From the streak photography used to follow how the reaction progressed with time, it can be seen that there was a steady increase in the burn speed up to the point where the material actually detonated. All detonations were prompt detonations and the input pressure necessary to cause a "go" result is therefore the detonation pressure. This is "shock-to-detonation" or SDT. It is likely that the sensitivity hierarchy would be the same if longer charge lengths had been used to allow deflagration-to-detonation processes to occur, but this has not been demonstrated for these samples.

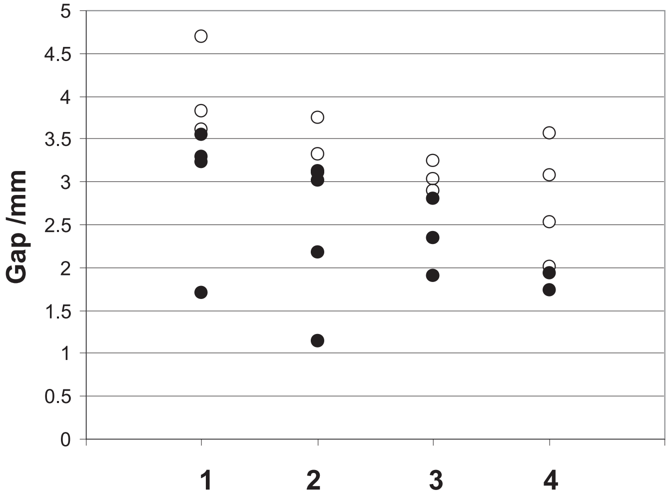


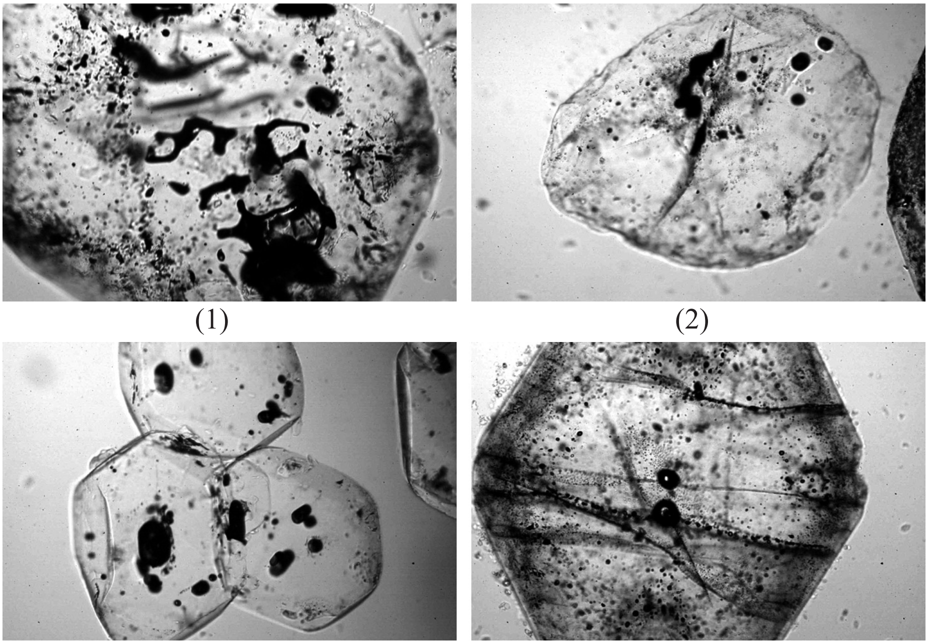
Figure 2. Gap test results for the four batches. Filled symbols represent "go" results and open symbols represent "no go" results, as discussed in the main text. An additional 2 mm of PMMA gap attenuates the shock by approximately a factor of 2. These results show significant differences in shock sensitivity, since nearly twice the critical pressure for sample (1) is required to initiate sample (4).

Investigation of morphology

A range of techniques are available to study individual particle morphology. Environmental scanning electron microscopy (ESEM) yields information on surface features of 1 μm and above in size. In contrast to normal scanning electron microscopy, ESEM allows samples to be viewed in a gaseous environment and non-conductive samples such as explosive crystals do not have to be coated with a conducting material. This reduces the sample preparation time and the possibility of introducing artefacts in the coating process. Optical microscopy of particles which are surrounded by a refractive index-matched fluid can be used to observe closed internal voids down to 1μm in size, and also shows the general particle morphology. Mercury porosimetry provides surface roughness data in the form of specific surface area (with a resolution down to 0.01 μm).

Figures 3 and 4 show typical crystals viewed using optical microscopy and ESEM. No discernable surface or internal features correlate with sensitivity in the material.

The contents of the closed internal voids seen is unknown, but it has been shown [5] that it is likely to be a mixture of water, solvent and air. Gas-filled voids are expected to affect shock sensitivity the most as the gas is compressible and allows the void to collapse and the material around it to plastically deform. Previous studies [11] have shown that shock sensitivity can decrease with void content, *i.e.* a *higher* pressure is required for the system to detonate. This can be understood if a variety of mechanisms can cause critical hotspots and there is no single ignition process for all particle morphologies.



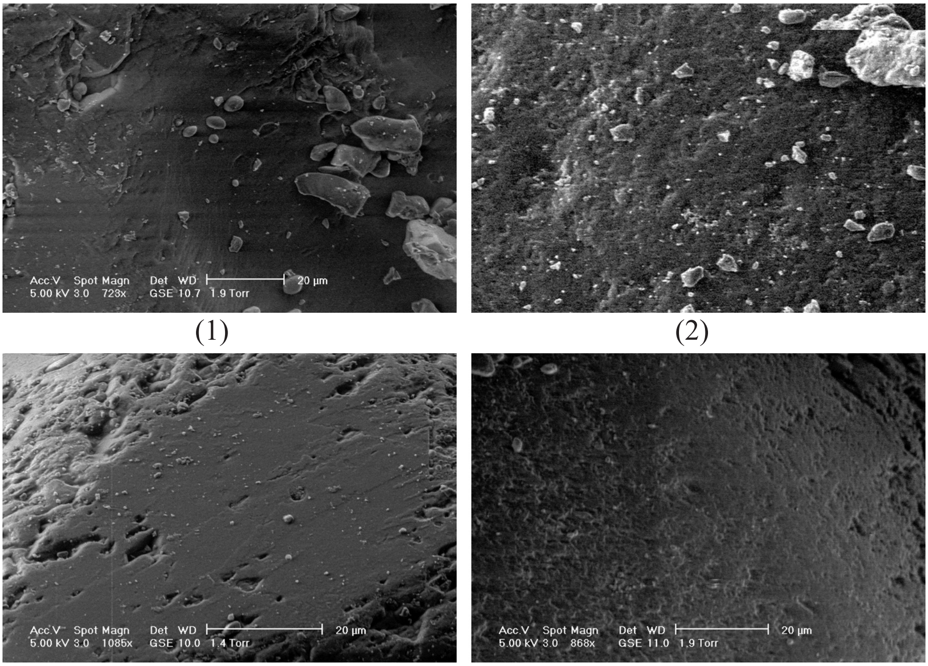
(1) (2)

(3) (4)

Figure 3. Optical microscopy showing typical crystals from the four batches. The field of view in each case is 1.3 mm wide.

Quantifying bulk morphology is far more difficult, but an indication of intergranular friction (which may affect the packing and coordination number) is provided by measuring the angle of repose. The angle of repose is the angle between the horizontal and the slope of a poured pile of the granular material (see Figure 5). Particles which interlock more and have greater intergranular friction will be able to support a steeper slope.

For all of these samples, the measured bulk properties (porosity and angle of repose) were very similar in spite of the large differences in overall shape. However, they are very different to the measured quantities for batches of RDX with smaller particle sizes [5], so this may be relevant in a wider context.



(1) (2)

(3) (4)

Figure 4. ESEM images of the surface of each of the four crystal batches. Since any critical hotspot is expected to be 0.1-10 μm in size, it might be expected that features of approximately that size would be potential sites for critical hotspots. However, no correlation is seen here between any surface feature and sensitivity. (1) is the most sensitive and (4) is the least sensitive material.

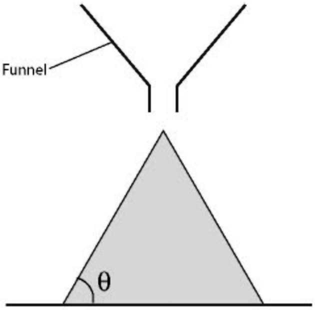


Figure 5. Angle of repose of a powder.

Table 1. All quantifiable data for each batch. Samples are shown with the most sensitive at the top of the list and the least sensitive at the bottom. Samples from different manufacturers were supplied with up to 4% of HMX as an impurity. No average void content is shown because this data does not capture the highly irregular void shape and size in some samples, and such a comparison would be misleading. The percentage of the theoretical maximum density (% TMD) data relates to the free-poured and tapped samples which were used for the sensitivity tests. No trends can be seen in any single parameter as the shock sensitivity decreases

Sample	Critical Gap mm^{-1}	% HMX	Specific Surface Area $m^2 g^{-1}$	Angle of repose	% TMD
(1)	3.6 ±0.2	0	0.0044	35 ±2 °	62 ±2
(2)	3.2 ±0.4	4.1	0.0087	35 ±1 °	62 ±1
(3)	2.9 ±0.3	0	-	35 ±2 °	59 ±1
(4)	1.9 ±0.4	0.3	0.0018	32 ±2 °	63 ±1

Discussion

An empirical study of the morphology and shock sensitivity of a material cannot prove in absolute terms whether any one particular mechanism alone is responsible for sensitivity. Direct observation of these mechanisms causing critical hotspots in a shocked material is not possible in these systems. However, any correlation seen would be a strong indication of the importance of a particular feature and a corresponding mechanism.

These results show that there are significant differences in sensitivity between different RDX batches. Some manufacturers claim to produce a “reduced sensitivity” product, but no causal explanation of these effects has been found. The batches tested here are a mixture of “reduced sensitivity” products and standard products from manufacturers. Much of the current effort to quantify and explain the difference has made the assumption that there is one dominant property or mechanism which extends across all the products. No clear correlation is seen here between any measured property and the shock sensitivity.

Large particles are more likely to fracture than smaller particles, which tend to plastically deform, and it seems that some of the stress in the shock could be relieved by particle fracture before ignition. The samples studied here contained large grains of millimetre dimensions, and so are also likely to contain large internal cracks and defects initially. This means that fracture is more likely than plastic deformation to relieve the initial stress. Fracture surface energy in these materials is not sufficient to cause critical hotspots, but the fractured morphology could respond differently to the continued loading.

Jetting from surfaces concave to the incident shock can be a source of critical hotspots. As the radius of curvature of the concave region increases, the resulting jet increases in speed. Jets causing critical hotspots are most likely from cavities with a radius of a few microns or less. Significant numbers of external particle features which meet this criterion were not observed in any one sample more than the others and only the least sensitive material had significant numbers of closed internal voids of this size. It seems that jetting alone is not a likely critical mechanism in this case.

There are large closed internal voids in the most sensitive material examined, but their contents is not known. Hotspots caused by adiabatic heating of trapped gas are more likely when the gas spaces are larger, but in this case the closed internal voids are extensively interconnected. No other sample contained significant numbers of larger internal voids and there is no correlation between internal void content and sensitivity here. The most important factor here could be the number of gas-filled spaces only, since closed internal voids could also contain water or solvent.

No clear correlations were seen between the bulk material properties and the sensitivity. Quantities such as coordination number are difficult to measure directly, but could be very relevant, since the lower the number of interparticle contacts, the higher the stress concentration at each contact. The importance of such parameters has not been investigated, but it seems very likely that the nature of the packing and the interparticle contacts must play a large role in localising stress. Contacts such as these will experience considerable stress and plastic deformation at such regions could cause very high temperatures locally, making these potential sites for critical hotspots.

The fact that the “go” results were all prompt detonations implies that the input pressures necessary to cause a “go” were above or equal to the detonation pressure. However, it seems likely that the resulting sensitivity hierarchy will be the same as one measured with longer charges.

Conclusions

In the case of free-poured granular samples, there is no correlation between any individual morphological feature and shock sensitivity for these large particle sizes. Previous work [7] has shown that there is no such correlation for smaller particle sizes either. This suggests that there is no single quality leading to the differences in sensitivity between different RDX products.

The critical pressure for these samples varies by a factor of two between the most and least sensitive, so there are significant sensitivity differences between samples of the raw material from different manufacturers. The observed variation in shock sensitivity may be explained by qualitative differences in how a shock interacts with the bulk material as well as with individual particle features. It may be the case that the combination of material properties in RDX mean that several critical hotspot mechanisms become significant at a similar shock pressure. Separating out specific mechanisms and quantifying their contribution would be extremely difficult at this stage. For the samples examined here, we cannot identify which mechanisms are dominant in causing critical hotspots, but we can make some suggestions.

Bulk morphology is difficult to quantify since properties such as average coordination number cannot be directly measured. Only angle of repose and the particle size analysis provide quantitative data on bulk morphology and neither of these quantities correlates directly with sensitivity. However, it seems likely that the interaction of particles in the granular bed must make some contribution to the inhomogeneity of the stress field. A non-uniform stress field will cause a non-uniform temperature field, because of the micromechanical response of the material to shock. There are many processes that could cause critical hotspots (viscoplastic deformation, jetting, friction, adiabatic gas collapse) and all are directly affected by both the microstructure and how the particles are arranged. Further investigation of how the interaction of bulk and individual particle morphology might affect shock sensitivity in free-poured granular beds is required. Future papers will deal with samples which has smaller particle sizes than those reported here.

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