Aspects of material insensitivity

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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 been identified. 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 characterised using mercury porosimetry, environmental scanning electron microscopy (ESEM) and optical microscopy. This range of methods yields quantitative data on internal void size and number and surface structure. Shock sensitivity is quantified using small-scale gap tests, and demonstrates the are clear differences in sensitivity between batches from different manufacturers. Data is presented here from seven samples in two size classes (10-30 μm and 100 – 300 μm) which were sourced from three different manufacturers.

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 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 mechanism of ignition and initiation of 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 products are found to be chemically similar (some contain a few percent of HMX as an impurity) and so the explanation for these differences is thought to lie in the crystal and bed morphology. In the research presented here, seven batches of RDX 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. No other study to date has had access to such a large sample set, and here we have the opportunity to compare a wide range of 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 supporting 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 particle features, for example closed internal pores, to sensitivity(5,6,7). There has been some success, 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. In general, the mechanisms occur and contribute to the inhomogeneity of the temperature field and critical hotspot formation are friction, viscoplastic work, jetting, and adiabatic compression of gas-filled pores(8)

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 in the 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. Some links between morphology and sensitivity in cast materials have been found, but no one feature has emerged as most dominant for RDX.

Samples

Granular RDX from three different manufacturers and in two size classes was acquired. The two size classes were class 5 (10-30μm), class 1 (100-300μm), and laser particle size analysis confirmed that the batches fell within these ranges.

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(9) (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.

The 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. In all these cases, if the material ignited it would progress to a detonation before the end of the charge. No charges were observed to ignite but not detonate. The relevant criterion here is therefore the appearance of a single critical hotspot to start reaction in the bulk.

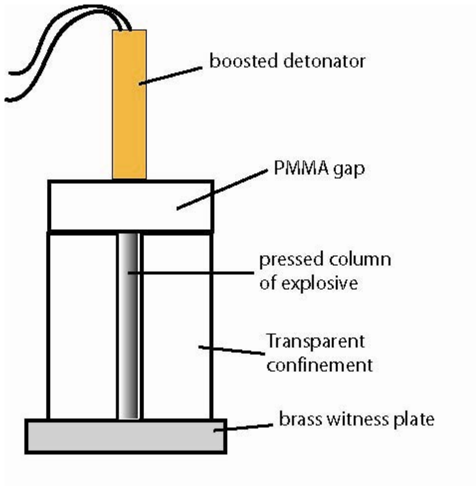


***Figure 1**. Diagram of the small-scale gap test. The charge was 5mm in diameter and 25mm long, contained within 25mm diameter PMMA.*

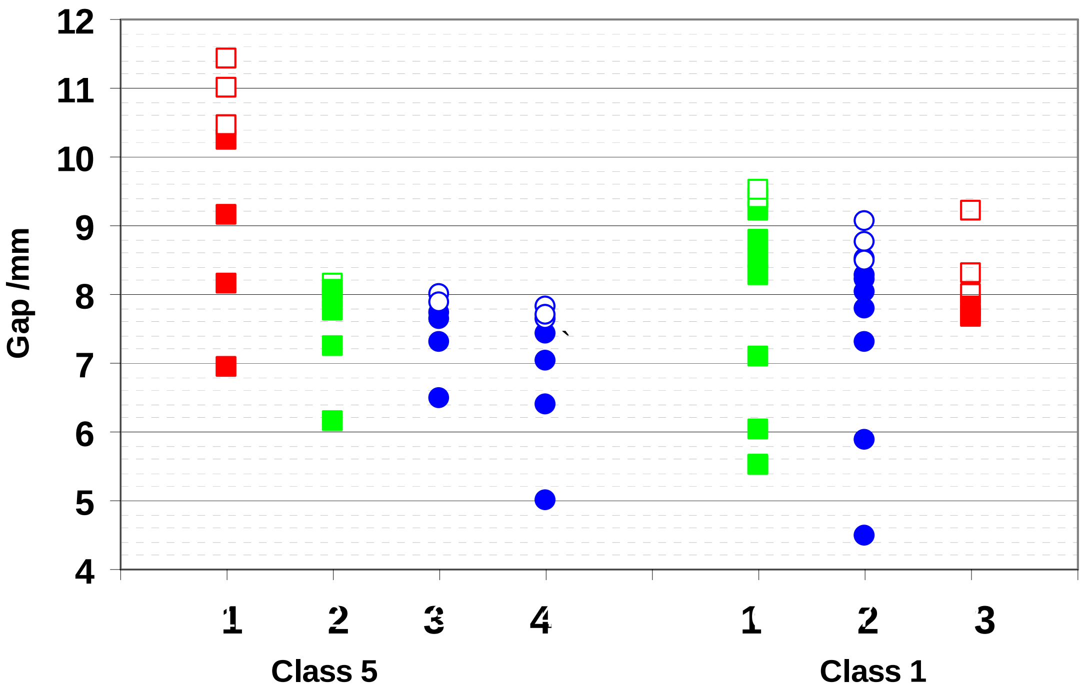


***Figure 2**: Gap test results for class 5 (10-30μm) andr class 1 (100-300μm) batches. Open symbols represent "no go" events and filled symbols represent "go" events.*

Investigation of morphology

A range of techniques are available to study individual particle morphology. Environmental scanning electron microscopy yields information on surface features of 1μm and above in size. 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 the specific surface area (with a resolution down to 0.01μm).

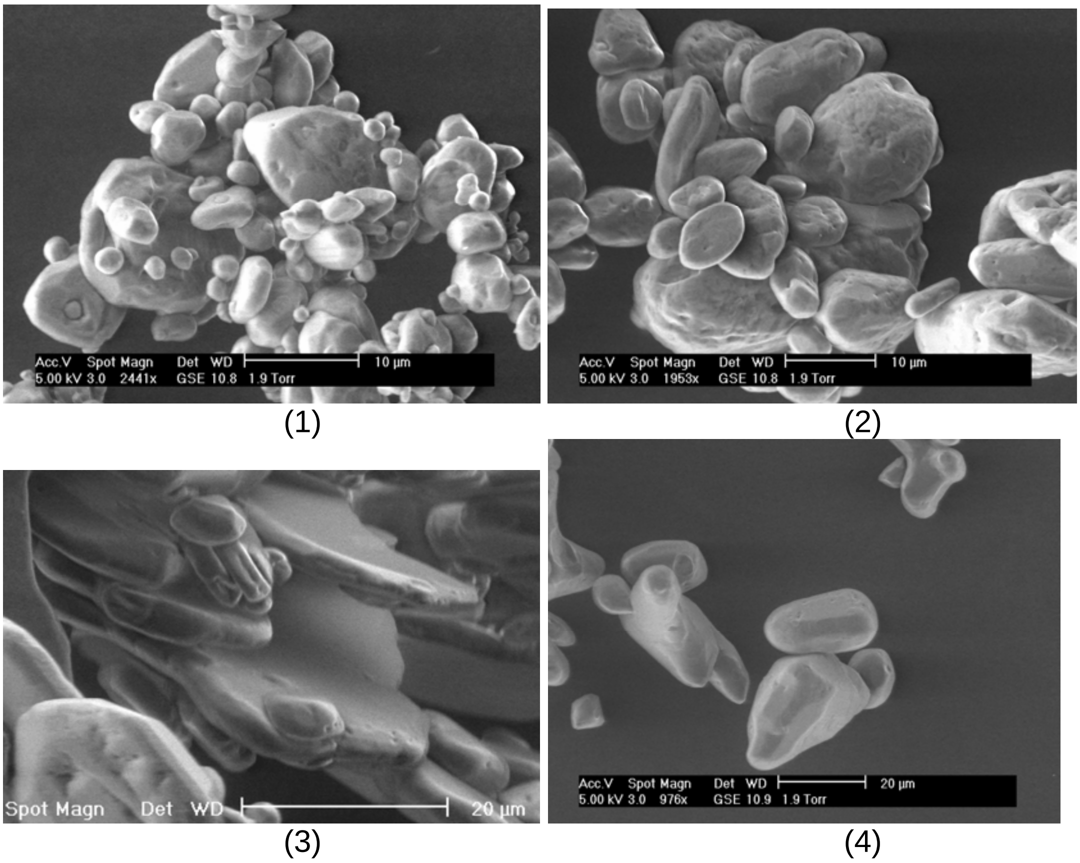
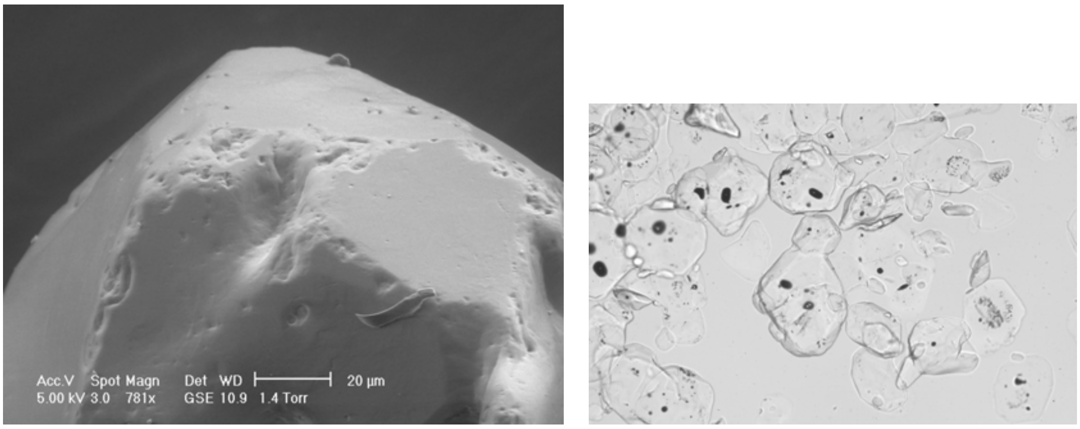
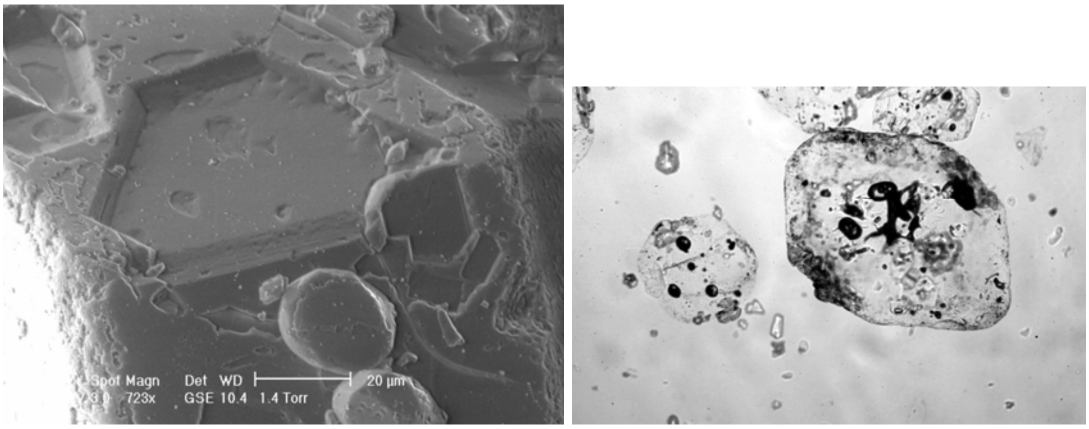


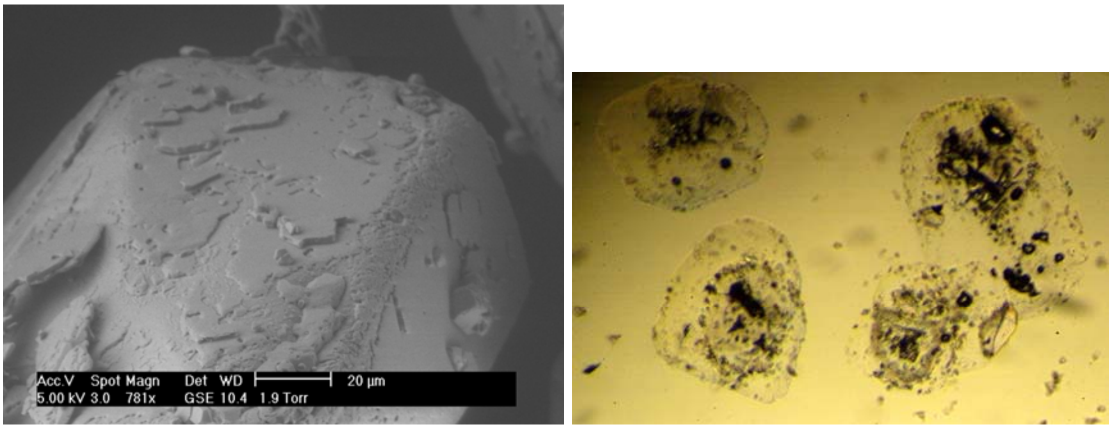
Figure 3 *ESEM images of the four RDX products of the smallest size class. (1) is the most sensitive and (4) is the least sensitive. (1) has many micron-sized dimples, which may be the cause of its high sensitivity.*



(1)



(2)



(3)

Figure 4 *ESEM and optical images of the medium-sized (class 1) material. The most sensitive (1) is shown at the top and the least sensitive (3) is at the bottom. The field of view of the optical images is 0.6mm wide.*

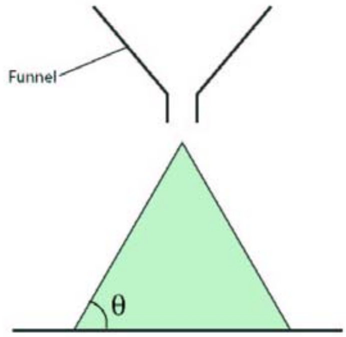


***Figure 5**: Schematic representation of the angle of repose. The material is poured though a funnel to form a conical pile and the angle between the horizontal and the side of the pile is measured.*

Sample	Critical Gap /mm	% HMX	Mean void number per crystal	Specific Surface Area m^2/g	Angle of repose	% TMD
Class 5 (1)	10.3 ± 0.9	0.9	4.1	0.45	48 ± 2 °	50 ± 3
Class 5 (2)	8.1 ± 0.2	0	1.5	0.64	52 ± 5 °	38 ±1
Class 5 (3)	7.8 ± 0.2	7.3	3.1	0.11	60 ± 4 °	32 ±1
Class 5 (4)	7.5 ± 0.3	0	0.1	0.2	48 ± 4 °	44 ±1
Class 1 (1)	9.3 ± 0.4	1.0	9	0.032	50 ± 3 °	61 ±1
Class 1 (2)	8.5 ± 0.2	8	21	0.034	41 ± 3 °	62 ±1
Class 1 (3)	7.9 ± 0.3	0	35	0.027	46 ± 4 °	61 ±1

***Table 1**: All quantifiable data for each batch. Within each size class, samples are shown with the most sensitive at the top of the list and the least sensitive at the bottom. Samples from different manufacturers had various amounts of HMX as an impurity. 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, except that for the class 1 material, the more sensitive particles have the fewer voids.*

Optical photographs and ESEM images are shown for some of the particles in figures 3 and 4. In the class 1 materials, the most sensitive particles have fewest closed internal voids, smoother surfaces and most a more angular shape.

The contents of the closed internal voids seen is unknown, but previous studies[3] have shown 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.

Quantifying bulk morphology is more complicated, but an indication of intergranular friction (which may affect the packing) 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.

In spite of a detailed inspection of the morphology of the individual crystals, no other clear correlation was seen between shock sensitivity and any individual particle feature (including overall shape, surface roughness at different scales, closed internal void content or percentage HMX content) for both classes.

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 features actually 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.

These results show that there are significant differences in sensitivity between different RDX batches in the same size class. 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" 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.

The particle sizes of the materials tested in this research differ by an order of magnitude. There is no reason to make the starting assumption that the same mechanism is responsible for ignition in both cases. Indeed, it would be surprising if this were the case. In the time a shock could completely cross one of the smallest class 5 particles, the plastic deformation at a contact point between much larger particles would only just have started, and the rest of the particle would not yet be shocked. The timescale of deformation and the resulting shock front shapes and intensities would be very different in the different size classes.

In the case of the smallest size class, the only feature which appeared to correlate with sensitivity was micron-sized surface dimples. Concave surfaces of this size could generate jets of shocked material fast enough to generate a critical hotspot when they hit another particle. The more sensitive materials were observed to have more dimples in the ESEM images. This could not be quantified with the specific surface area as it is almost impossible to separate out the contribution of surface roughness from that of the general shape.

For the medium-sized material (class 1) the more closed internal voids there are, the less sensitive the material. This suggests that either closed internal voids damp the shock wave and so de-sensitise the material or that they are irrelevant in this case. They could be mostly filled with solvent, and thus be less likely ot sensitise the material. The most sensitive particles in this size class were the most angular and so it seems that plastic work at contact points between particles might be the dominant cause of critical hotspots.

No clear correlations were seen between the bulk material properties and the sensitivity. Quantities such as co-ordination 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 yet 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. .

Conclusions

In the case of free-poured granular samples, there is no correlation between any individual morphological feature and shock sensitivity for both particle sizes. 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. This 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.

In the samples examined here, it seems that plastic deformation at contact points between particles may be the dominant critical hotspot mechanism for material that has particle sizes of 100-300μm. For the 10-30μm particles, it seems that dimples on the particle surfaces may cause jetting which contributes significantly to critical hotspots.

Bulk morphology is difficult to quantify since properties such as average co-ordination 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 in relation to one another. We think that an explanation of the sensitivity differences observed may be found by further investigation into how both bulk and individual particle morphology affect the micromechanical response to shock in free-poured granular beds.

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