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REVERSAL OF PARTICLE SIZE/SHOCK SENSITIVITY RELATIONSHIP AT SMALL PARTICLE SIZE FOR PRESSED HETEROGENEOUS EXPLOSIVES UNDER SUSTAINED SHOCK LOADING

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ABSTRACT

Shock initiation of pressed heterogeneous explosives has been reviewed. The key processes of ignition and buildup and their relative importance under sustained and short duration shock are described. Particle size effects on shock sensitivity are shown to depend on both density (%TMD) and shock duration. A series of RDX samples of narrow particle size range were subjected to sustained shock (gap test). Sensitivity increased over the series 250 < 138 < 100.0 < 21.2 μm median particle size but decreased for a 3.9 μm sample. These results combined with earlier published data support published modelling studies which predict decreased shock sensitivity at small particle size.

INTRODUCTION

The initiation and buildup to detonation of solid heterogeneous pressed explosives is a key process for proper functioning of explosive trains. Despite the importance of knowledge of this process to ordnance design, it was not till the early 1960s that the first detailed studies were published[1-4]. Investigation over the last twenty-five years, both experimentally and via theoretical modelling, has considerably increased our understanding of many aspects of these processes[5,6].

One of the key parameters controlling the macroscopic behaviour of shocked pressed explosives is particle (grain) size. Indeed, much of the insight into the microscopic processes which are occurring during initiation and buildup is derived from studies of particle size effects. Despite a substantial body of experimental evidence, the relationship between shock sensitivity and particle size is still poorly understood, although Price[7] has advanced our understanding in a recent report. The short but succint statement by Stresau and Kennedy[8] best summarises available knowledge: "fine-particle powders are often harder to ignite than coarse powders, but reactions in fine powders grow to detonation more rapidly once ignited."

We have been investigating the use of very fine particle size explosives in fuze trains, where there may be considerable advantages in decreased impact sensitiveness. However, Taylor[9,10]

has predicted substantial decrease in shock sensitivity to sustained shock at very small particle sizes, and has cited limited experimental evidence in support[11-13]. We accordingly undertook a study to investigate the effect of explosive particle size on shock sensitivity. In this, the first part of the study, we review the literature and present current understanding of the physical processes occurring during shock initiation and buildup in heterogeneous pressed explosives, with particular emphasis on the effect of explosive particle size. A study of the effect of particle size on shock sensitivity of binderless RDX from < 5 μm to 250 μm particle size is also described.

INITIATION BY SHOCK

It is generally agreed that initiation of heterogeneous solid explosives occurs at inhomogeneities in the pressed solid, where interaction with the incident shock wave produces small localised regions of high temperature ("hot spots"). Energy released by growth of chemical reaction from the hot spots can subsequently become self-sustaining, reinforcing the incident shock wave and leading to detonation. A number of possible mechanisms for formation of the hot spots have been proposed and largely fall into two types.

(i) Shock induced collapse of voids within the filling. Mechanisms for energy generation include impact of the void front onto the void rear and associated

microjetting[1,5,9,14], and viscoplastic work done on explosive at the void peripheries[5,9,15,16,17]. The stagnation (of microjets) theory proposed by Seely[18] and elaborated by Stresau[8] is closely related to these mechanisms.

(ii) Impact[8] and friction[5,8,9] between grains, and viscous friction within deforming grains[9]. The related process of shear banding has also been proposed[16,19].

Howe et al[20] have concluded on the basis of theoretical models that the relationship between particle size and threshold pressure for initiation can only be explained if both void collapse and frictional processes are operating. It is generally agreed that shock interaction with voids is the primary mechanism, with friction playing a secondary role. Note that Lee et al[21] have concluded that pore (void) collapse may not be the dominant mechanism for producing reaction sites during shock initiation of TATB at densities below 1.65 Mg/m^3. Experimental evidence has been variously interpreted to mean that shock sensitivity depends principally on the number[1], volume[22] and surface area[20] of voids, but the relative importance of each contribution is not known. Theoretical modelling of shock initiation in heterogeneous explosives has largely centred on the void collapse mechanism[9,14,23] but more recently other mechanisms have been treated[16]. Adiabatic compression of interstitial gases, which is the most important mechanism for initiation under impact[24], is not

normally operative under shock conditions[2,25].

The molecular processes occurring during shock initiation are not known. Walker et al in a series of papers[26,27] have proposed that the shock initially causes cleavage of the (explosive) molecules into ions and free radicals. If these reactive species are formed in sufficiently high local concentration they could grow to produce a self-sustaining exothermic decomposition. Isotope labelling studies[28] have indicated that the same bonds (C_α-H) are broken in the rate determining step for both thermal decomposition and shock initiation of TNT. An extension of this concept is that the incident shock does not need to be degraded to thermal energy, i.e. hot spots, but initiation can occur directly by bond shear[27]. A similar "tribochemical" mechanism was proposed some years previously[24], but both these mechanisms are currently considered to be unimportant.

BUILDUP TO DETONATION

Initiation (ignition) is followed either by buildup to self-sustaining exothermic reaction and ultimately detonation, or by failure to propagate due to energy losses. Chemical energy release in the buildup and subsequent detonation occurs by grain burning as elaborated by Eyring[29]. However there is considerable disagreement on whether the rate of grain burning or physical processes such as permeability, thermal conduction, convection and diffusivity are the key parameters controlling this process.

Most of the evidence for grain burning being the dominant parameter in buildup following shock initiation comes from study of particle size effects[6,8,16,20,22]. In essence, the higher shock sensitivity of explosive compacts pressed from small particle size materials to very short duration pulses has been attributed to their higher rates of grain burning. Although the burning rate of materials such as HMX is faster for small particle fractions[30], particle size effects on both deflagration-to-detonation transition (DDT)[31] and strand burning[32] of low density high explosives are not consistent with this picture: larger particle size materials show more rapid buildup. One explanation of this apparent contradiction is that there is an abrupt increase in the pressure exponent at higher pressures, and there is some evidence to support this[33]. Large increase in burn rates in closed bomb tests due to crystal breakup at higher pressures has been observed[30], and could also contribute to this disparity. Lee and Tarver[5] concluded that the growth of reaction from the ignition sites apparently proceeds at rates that exceed the linear burn rate-pressure dependence of laminar deflagration in explosives.

A basically different picture has been proposed by Mader[14,34]; energy released close to the shock front by thermal explosions of the hot spots can result in shock acceleration. The strengthened shock then produces increasing numbers of hot spots till the shock ultimately builds to a detonation wave.

In summary, the effect of a number of key parameters which could define the key processes in buildup, and which could be used to predict buildup/propagation success, has not been defined with any certainty.

THE RELATIVE IMPORTANCE OF INITIATION AND BUILDUP

Experimental evidence is consistent with the ignition process and the buildup to detonation process being separate[22]. Their relative importance in the overall formation of self-sustaining detonation has two aspects. In terms of energy release, it is now accepted that energy contribution from the hot spots formed at initiation is very small relative to the energy released by grain-burning during buildup. In other words, initiation occurs by formation of hot spots at the shock front, then these hot spots grow and burn in the reaction zone to generate the energy necessary to build to and sustain detonation.

Whether initiation or buildup will be the key process leading to detonation is critically dependent on the nature of the incident shock. Walker and Wasley[35] proposed, on the basis of experiments using short duration/high pressure shocks delivered to nearly voidless explosives by flyer plates, that there existed a "critical energy" criterion for initiation. This criterion states that there is a critical energy per unit area which must be delivered to an explosive to obtain detonation. The relationship

$$\frac{P^2 t}{\rho_o U} = \text{constant} \quad \text{or} \quad Put = \text{constant} \ (\text{since } P = \rho_o uU)$$

can be derived[35] where P is the shock pressure in the explosive, t is the shock duration, ρ_o is the initial density of the explosive, U is the shock velocity in the explosive and u is the particle velocity in the explosive. Since $\rho_o U$ changes only very slowly with increasing P, the relationship reduces to P^2t = constant, the form in which it is normally used. de Longueville et al[36] have derived a related "critical time" concept for shock initiation.

The critical energy criterion is now considered to be a useful engineering development guide applicable to voidless explosives of the conventional CHNO composition. It is not a general relation; although the shock sensitivity of some explosives obeys P^2t = constant over a considerable range of t[35,36,38], most explosives do so only at very short pulse durations, typically < 1 μs [36-39], while the behaviour of others does not correlate at all[36,38]. The shock initiation of heterogeneous explosives subjected to long (or sustained) shocks in excess of 1 μs duration depends only on the incident shock pressure. Most standard shock sensitivity tests such as gap tests employ sustained shocks. In addition, lower density leads to increased sensitivity to sustained shock but decreased sensitivity to short duration shock.

Howe et al[20] proposed, on the basis of the different observed behaviour under short duration and sustained shock, that P^2t = constant correlated with buildup, while pressure dependent initiation correlated with the initiation stage.

Tarver et al[40] have recently published a revision of their earlier[5] phenomenological model for shock initiation of heterogeneous explosives. This revision[40] specifically deals with short pulse duration shocks, and the inability of the earlier model to accurately predict behaviour under these conditions. They have now proposed a three step process for initiation by short shock pulses: initiation by formation of hot spots, then slow growth of reaction from the isolated hot spots somewhat analogous to a deflagration in DDT, and finally rapid completion of the reaction by coalescence of hot spots. At longer pulse lengths, where initiation becomes the determining process, it is not necessary to split the buildup into two processes although it may still occur in this manner. Johnson, Tang and Forest[41] have also recently published a numerical model of shock initiation of heterogeneous explosives and have surveyed previously published models, particularly those relating to polymer bonded explosives (PBXs).

The physical picture which emerges from these studies shows a gradation of behaviour depending on the incident shock. Very high pressure/short duration shocks result in up to 20-30% of the explosive being ignited as hot spots[40]. The success or failure to grow to detonation is determined solely in the buildup stage, and the calculations of Tarver et al[40] strongly suggest that this buildup occurs as discrete slow and fast steps. Presumably the critical stage is the slow step roughly corresponding to deflagration in DDT. Low pressure sustained shocks result in

only a few tenths of a percent of the explosive being ignited as hot spots[41], and it is this ignition stage which is crucial to success or failure to grow to detonation. Further support for this proposition can be found in von Holle and Tarver's measurement of hot-spot temperatures in shocked explosives using time resolved infrared radiometry[42]; only relatively small amounts of explosive ignited close to the shock front for sustained shocks.

THE RELATIONSHIP BETWEEN EXPLOSIVE PARTICLE SIZE AND SHOCK SENSITIVITY

The relationship between shock sensitivity and explosive particle size has been reported for quite a number of pure pressed materials: TNT[1,20,22], RDX[36,43,44], HMX[8,25], PETN[18,43-46], tetryl[18,43,47], HNS[11,12], TATB[21,37,48], and HNAB[39]. A number of other studies dealing with particle size effects of some of these explosives in formulations have been published. In general they follow the behaviour of the pure materials, and no attempt has been made to cite them here. It should be noted that most of these studies are restricted to comparison of "coarse" with "fine", ie. only two or sometimes three size fractions are compared.

Analysis of the data in the references cited above plus others not cited reveals two key points:

(1) Particle size effects on shock sensitivity can only be discussed in conjunction with two other parameters: density and shock duration.

(2) The term "shock sensitivity" is used fairly loosely in the literature and the two major uses are not necessarily consistent. Researchers from the atomic weapons laboratories usually equate sensitivity with run distance to detonation, as typified by Pop plots[49]; shorter run distance at the same incident shock pressure is equated with higher sensitivity. The other criterion is threshold pressure for initiation, or the more readily determined shock pressure for 50% initiation probability; lower threshold (or 50% initiation) pressure means higher sensitivity[50]. In the remainder of this paper we will be largely using this definition.

Particle size effects on heterogeneous explosives pressed to high relative density, eg. > 98 %TMD typically achieved by isostatic pressing, are clear cut; shock sensitivity increases as particle size decreases. This behaviour is also followed by cast and homogeneous explosives[6,18,51] such as composition B. At these densities approaching crystal density, behaviour under shock is approaching that of homogeneous liquid and cast explosives[51]. One explanation given for the effect of particle size at high density is that these charges have very low permeability and shock sensitivity should therefore be a function of pore surface area which will increase with decrease in particle size[4].

At low (relative) densities of 95 %TMD and less, ie. accessible by normal pressing operations, pronounced dependency on particle size is often observed. The following discussion compares behaviour at identical %TMD.

For sustained shocks such as in gap tests, reaction thresholds as measured by incident shock pressure are nearly always lower for larger particle size materials[7,8,20,22,43,47] while pressures for 50% detonation probability[52] are also often lower[7,18,43,45,47]. One explanation is that in relatively low density charges the area of surface exposed to reaction products, which will determine shock sensitivity, will be a function of permeability which will decrease as the particle size decreases[4]. Alternatively the smaller grain size materials will have smaller voids and hence smaller hot spots, thus energy losses to thermal conduction will be higher[37].

As pulse duration decreases and shock pressure increases, there is usually a reversal of behaviour and smaller particle size materials are more shock sensitive[8,11,20,22,37,39,48]. Initiation under these conditions was discussed in the previous section: a relatively high proportion of the shocked explosive is ignited by the incident shock, and Howe et al[20] have suggested that it is the higher rate of grain burning which enhances the buildup success of small particle size materials. Another explanation is that under these higher incident shock pressures the hot spots are hotter, the reactions are consequently faster, and a state is reached

where the number rather than the size distribution of hot spots is important[37].

An illustration of the relative behaviour discussed in the preceding two paragraphs is shown below in Fig. 1. The curves suggest that threshold shock pressure is lower for larger particle size materials but the initiation energy (at short shock duration) is lower for smaller particle size materials. A similar figure can be seen for PBXN-5 (HMX/Viton A 95:5) in ref. 8 and data for RDX and TATB are listed in Refs. 21,36,48.

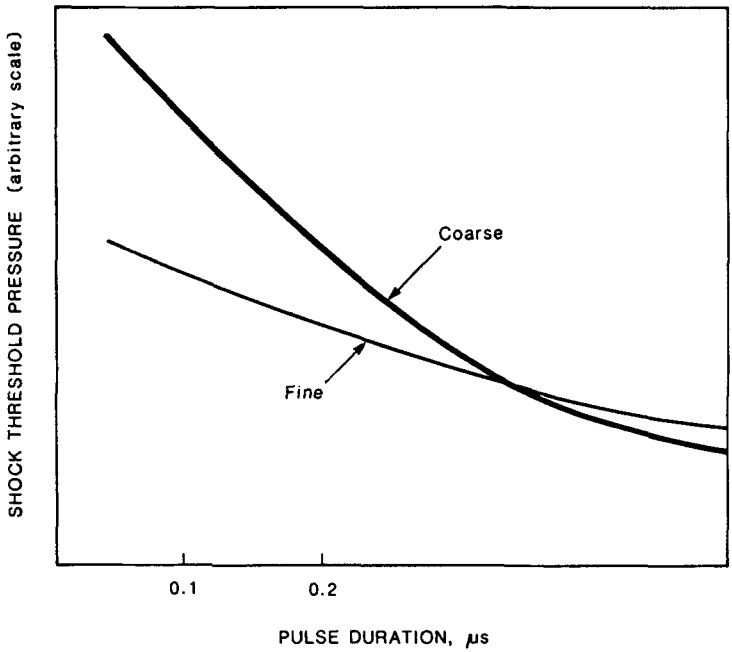


FIGURE 1 The relationship between threshold shock pressure and shock duration for coarse and fine particle size explosive pressed to the same density.

The relationship between shock sensitivity and explosive particle size has been modelled by Taylor[9,10]. Taylor assumed that hot spot formation occurred by void collapse and calculated hot spot temperatures and thermal energies for materials at the same density but with decreasing particle size. It was found that hot spot temperature remained roughly constant as particle size decreased till a critical pore size was reached; here the temperature achieved decreased substantially. This decrease in hot spot temperature was assumed to result in a decrease in ability of the hot spots to grow (to buildup). Mader has also arrived at similar conclusions for explosives at crystal density[34] and 90 %TMD[53] on the basis of numerical modelling. Interestingly, de Longueville[36] had earlier suggested that hot spot temperature determined shock sensitivity.

Taylor[9] then used this calculated data in conjunction with two published observations,

(1) explosives under sustained shock display higher chemical reactivity as particle size decreases[1,13,54].

(2) coarser grained explosives exhibit lower reaction thresholds during sustained shocks[8,20,36,38].

to suggest that a reversal of shock sensitivity for sustained shock should occur. That is, as particle size decreases, shock sensitivity increases due to increased surface area which enhances

buildup, but below an (unspecified) particle size initiation probability decreases due to decrease in hot spot temperature not compensated for by increased surface area.

Taylor's calculations[9] are limited to void collapse as the sole mechanism for hot spot formation and Howe[20] has shown previously that this is not normally the case. In addition, the higher chemical reactivity of smaller particle size material, cited as (1) above, is not necessarily normal behaviour. While it might be tempting to dismiss Taylor's predictions because they are possibly based on incorrect assumptions, if they do predict trends correctly they could have profound importance on future design of fuze trains via reduced shock sensitivity for very small particle size materials. With the exception of HNS and TATB, most of the published data on particle size/shock sensitivity cited earlier refer to "large" particles, eg 75–500 + μm.

EXPERIMENTAL

Preparation of RDX Samples

Grade A Sieve Fractions

RDX Grade A, Class 1, was supplied water wet from Albion Explosives Factory. The material as received was passed successively through a series of sieves under water by gentle brushing. The sieves as used sequentially were 355, 300, 250, 212, 180, 150, 125, 106, 75 and 45 μm. The three samples used in this study, 250–300 μm, 125–150 μm and 75–106 μm consisted of

material retained on the smaller pore size sieve and passing through the larger. Material retained on the sieve was rinsed off with distilled water and dried by suction filtration.

Grade E

RDX Grade E, prepared by aqueous acetone precipitation of Grade A[55], was supplied water wet by Weapons Systems Research Laboratory (WSRL), Salisbury. The material as received was dried by spreading thinly on paper and air drying with break up of agglomerates.

Ball-milled

RDX Grade A was ball-milled at WSRL under inert solvent to nominal size 4 μm. The material was supplied water wet and was dried as described above for Grade E.

Characterisation of RDX Samples

Particle Size Measurements

Samples were prepared by dividing about 1 g of the powder into small fractions (10 to 20 mg) using a rotary sample divider[56] constructed at MRL. Sample splitting of powder slurried in liquid was preferred to splitting the dry powder due to the poor flow of dry RDX powders especially for small particle size fractions.

Particle sizes were determined using a Malvern Particle Size Analyzer Model 2600/3600. The samples as slurries were dispersed

using an ultrasonic bath for 1 minute, then transferred to an optical cell and placed in the laser beam path. The finer particle size materials, Grade E and ball-milled RDX, were dispersed in HPLC grade chloroform. The coarser materials were dispersed in distilled water with a small amount of Decon 90 surfactant.

Data reduction was performed using computer programmes supplied with the instrument. The results are presented as total mass per sampling band. The particle size distribution model which was found to be most applicable was a "model-independent" 16 parameter fit over 15 size bands.

Scanning Electron Microscopy

A Cambridge Instruments Model S250 MkII scanning electron microscope with a tungsten electron gun was used. The instrument was operated at 15-21 kV in the secondary electron mode. The samples were prepared by mounting crystals of the formulation with PVA adhesive onto a stub coated with PVA adhesive. The sample was then sputter coated with a conducting film of gold. Micrographs were generally obtained for tilt angles of 30^{o}.

Shock Sensitivity: MRL Small Scale Gap Test[57]

The MRL small scale gap test (SSGT) has been described previously in detail[57]. The system consists of a donor of a PETN filled exploding bridgewire (EBW) detonator, an acceptor of two

12.7 mm diameter x 12.7 mm height pressed cylinders of the explosive under study, with the gap being of laminated brass shim. Detonation of the acceptor is confirmed by a sharply defined dent in a 25 mm square x 12.7 mm thick mild steel witness block. A typical run consists of 25–30 firings conducted using the Bruceton staircase method[58], the result being expressed as the gap in mm at which detonation probability is 50%. Donor EBWs were UK Mk 3 supplied by AWRE Aldermaston.

RDX acceptor pellets were pressed to the required density on an Instron Universal Testing Machine operated as a press. Complete experimental details of this procedure have been given previously[59]. It was found that the pellets, particularly from the finer powders, possessed poor mechanical strength resulting in cracking or breaking upon ejection from the mould or subsequent handling. Two strategies were used to overcome this problem.

(i) The RDX Grade A powders (2.50 g) were pressed in two sequential stages each of 1 min duration. The pellet was removed from the mould at the completion of the second 1 min period. In the case of the 125–150 μm and 75–106 μm sieve cuts a thin film of oleic acid was applied to the mould prior to addition of the powder; this facilitated ejection of the pellet.

(ii) The Grade E and ball milled RDX were first tamped into the mould then a cavity was formed in the mildly consolidated

powder with a 3 mm diam. plunger. Acetone (0.02 mL) was injected into the cavity and the drift was placed lightly on top in the mould. After leaving the assembly in this condition for about 5 min, it was pressed for a single 3 min period. This treatment was necessary to form pellets strong enough for ejection from the mould; it also lowered the force necessary to press the pellets and facilitated their ejection.

Densities were determined by accurate weighing and dimensional measurements.

RESULTS AND DISCUSSION

Characterisation of RDX Samples

Median particle sizes and 16-84% probability ranges for the five RDX samples are listed in Table 1 and the particle size distributions are depicted as bar charts in Figs 2 and 3. All samples exhibit quite narrow particle size ranges and, with the exception of the 75-106 μm and 125-150 μm samples there is minimal overlap with other samples. All the sieve cuts have a tendency to contain significant amounts of particles smaller than their lower sieve limit; this results mainly from fracture of smaller crystals from the bridged agglomerates during treatment in the ultrasonic bath.

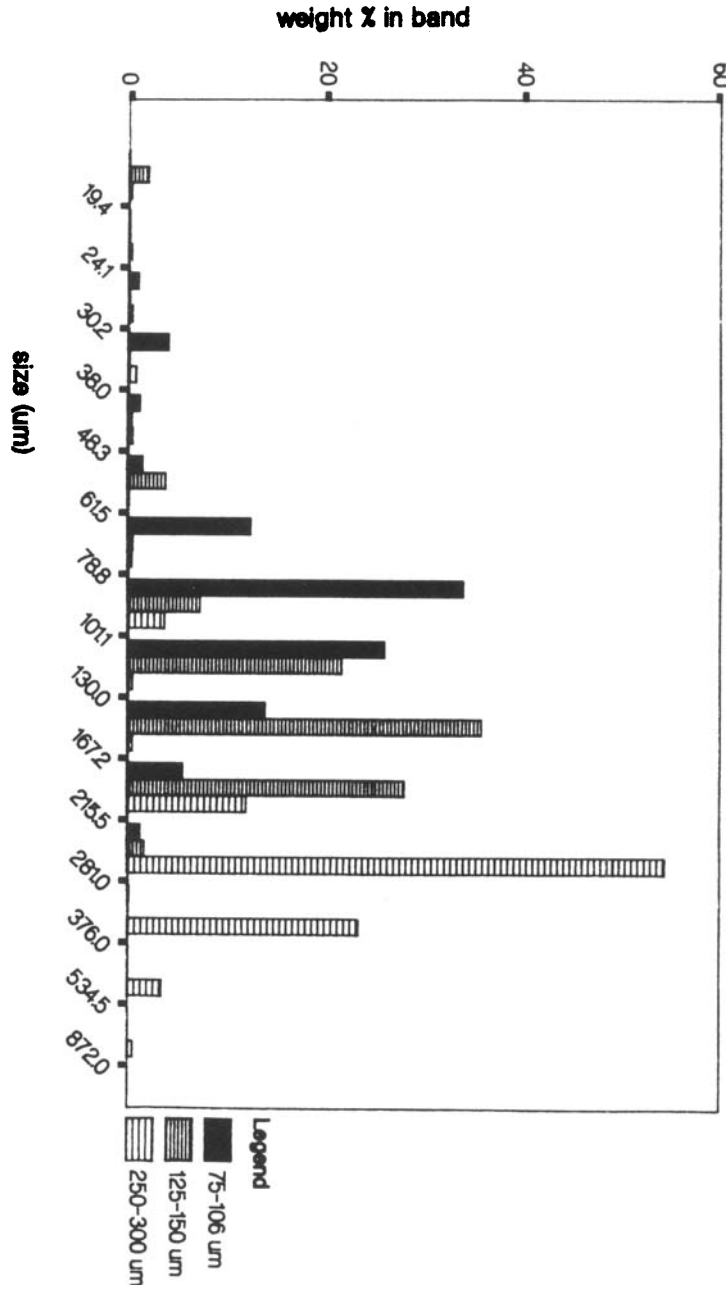


FIGURE 2 Bar chart showing particle size distribution of three sieve cuts of RDX Grade A over 16 sample bands.

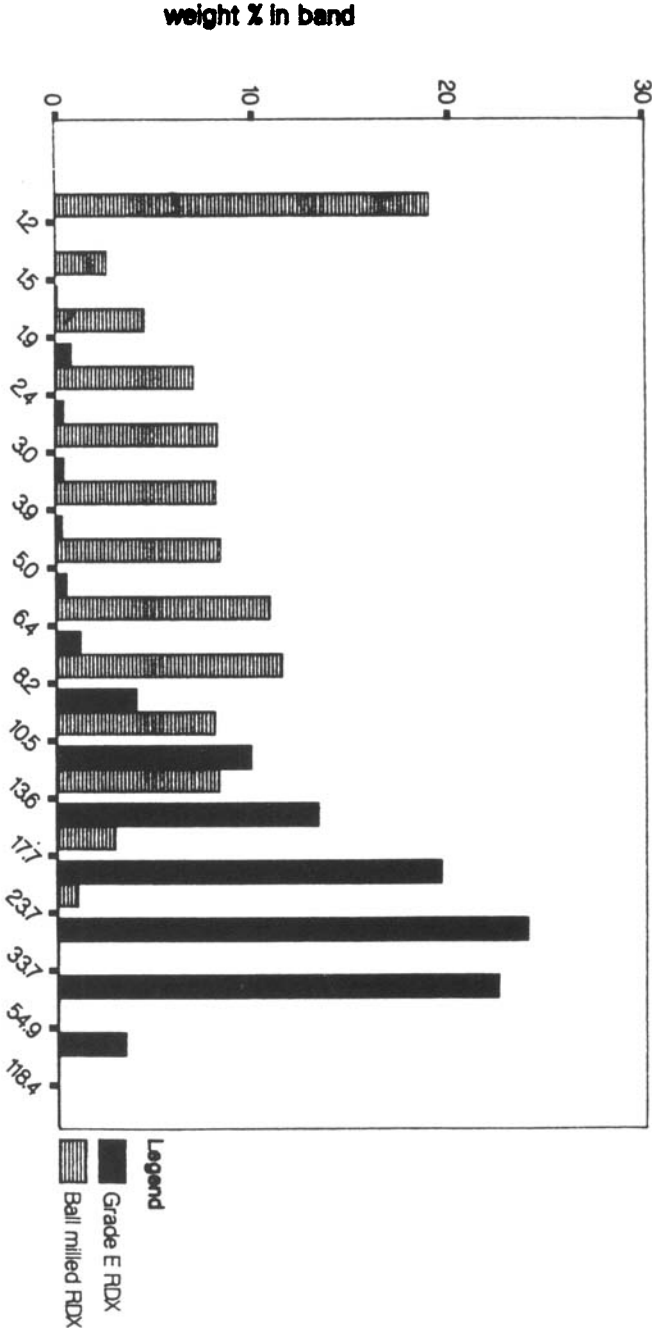


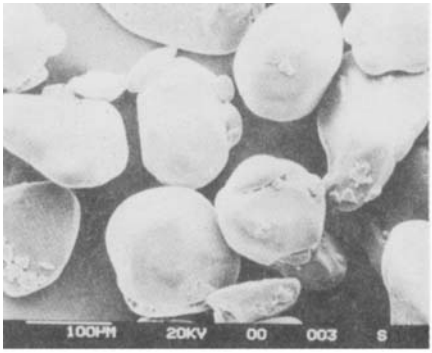
FIGURE 3 Bar chart showing particle size distribution of two samples of RDX over 16 sample bands.

TABLE 1

Particle Size Data for RDX Powders

RDX Sample	Particle Size Data (μm)	
	Median	16%-84% Probability Range
Grade A		
250-300 μm sieve cut	250	220 - 300
125-150 μm sieve cut	138	95 - 179
75-106 μm sieve cut	100.0	73.3 - 138.0
Grade E	21.5	10.8 - 36.0
Ball-milled	3.9	approx. 1 - 9.4

Scanning electron micrographs (SEM) of four of the five samples at magnification X200 are shown in Figs 4a-d. The recrystallised Grade A samples (Figs 4a and b, and the 250-300 μm sieve cut not shown) have the typical rounded irregular appearance. Small crystals growing off the faces, referred to above, can clearly be seen. The Grade E and ball-milled materials exhibit a different structure which becomes more apparent at higher magnification (X1000 and X2000) in Figs 5a-c. The Grade E material consists of a range of shapes from elongated spheres to rounded rods and other unusual forms. Such a proportion of particles with large length to diameter ratio makes accurate determination of particle size very difficult. However the particles are clearly much larger than the rounded ball-milled material and much smaller than the 75-106 μm Grade A sieve fraction.



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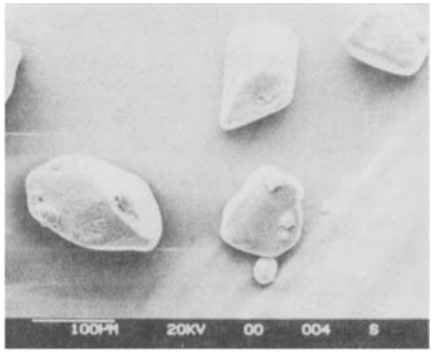

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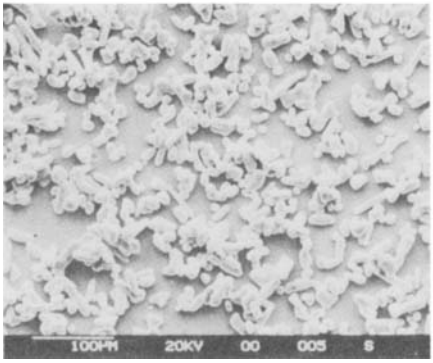

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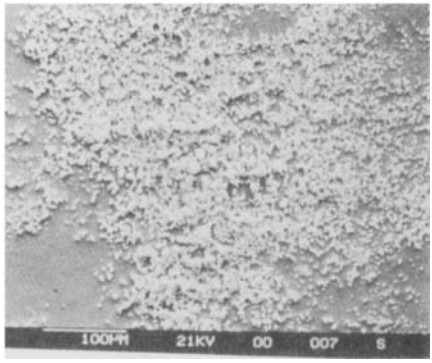

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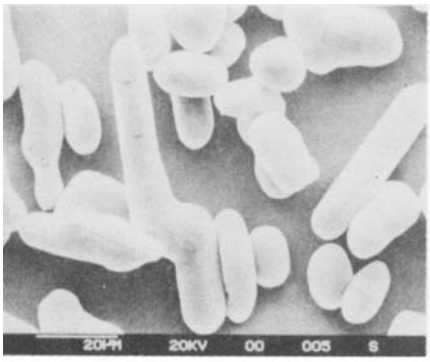
FIGURE 4 Scanning electron micrographs of four of the RDX samples studied, magnification X200.

(a) sieve cut nominally 125-150 µm, Grade A
(b) sieve cut nominally 75-106 µm, Grade A
(c) precipitated, Grade E
(d) ball-milled.



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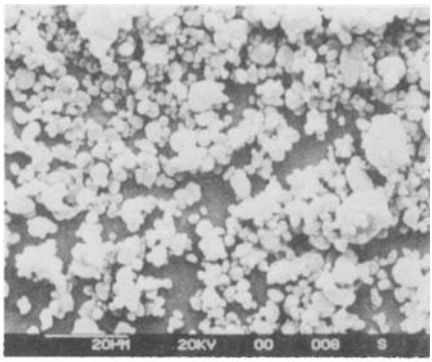

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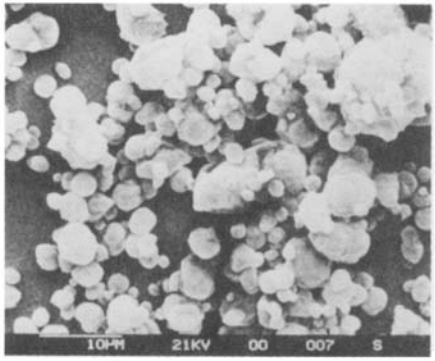

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FIGURE 5 Scanning electron micrographs of the two smaller particle size RDX samples, depicted in the previous figure, but at higher magnification.

(a) Grade E, magnification X1000
(b) Ball milled, magnification X1000
(c) Ball milled, magnification X2000.

Shock Sensitivity

Results for shock sensitivity (SSGT) of compacts of the five RDX samples pressed nominally to 90.0 %TMD (1.622 Mg/m^3) are listed in Table 2. The gaps for 50% initiation probabilities together with calculated standard deviations are plotted in Fig. 6. The results for the 250–300 μm Grade A sieve cut and Grade E RDX had high standard deviations. This is often observed for larger particle sized material and results from the relatively large range of shock pressure from the true threshold through to full detonation[43]; in the MRL SSGT the criteria for "go" is a dent in the witness block and shallow or full dents are not differentiated. In the case of Grade E RDX, the principal cause was probably the increased standard deviation of the acceptor pellet densities resulting from the necessity to use acetone to achieve adequate mechanical strength of the pellets (see Experimental). However, the 3.9 μm ball-milled material, which had a similar standard deviation on pellet density, exhibited a low standard deviation on $M_{50\%}$. This could mean that density dependence on shock sensitivity is decreasing at very small particle size, but further studies would be needed to substantiate this.

The clear trend shown in Fig. 6 is for the shock sensitivity of the pressed materials to increase steadily as median particle size decreases from 250 μm (Grade A 250–300 μm sieve cut) to 21.5 μm (Grade E). There is then a small decrease in shock sensitivity for the 3.9 μm ball-milled RDX, although the large

TABLE 2

Shock Sensitivity (SSGT) of RDX Pellets Pressed Nominally to 90 %TMD

RDX Sample	Pellet Densities Mg/m^3		Shock Sensitivity [a]	
	Mean(std.dev.)	[%TMD]	$M_{50\%}$	Std. dev.
Grade A sieve cut				
250–300 μm	1.622(0.002)	[90.00]	3.360	0.12
125–150 μm	1.622(0.001)	[90.00]	3.513	0.019
75–106 μm	1.620(0.002)	[89.89]	3.551	0.027
Grade E	1.623(0.014)	[90.04]	3.785	0.15
Ball-milled	1.624(0.009)	[90.13]	3.742	0.025

[a] All figures are in mm of brass shim.

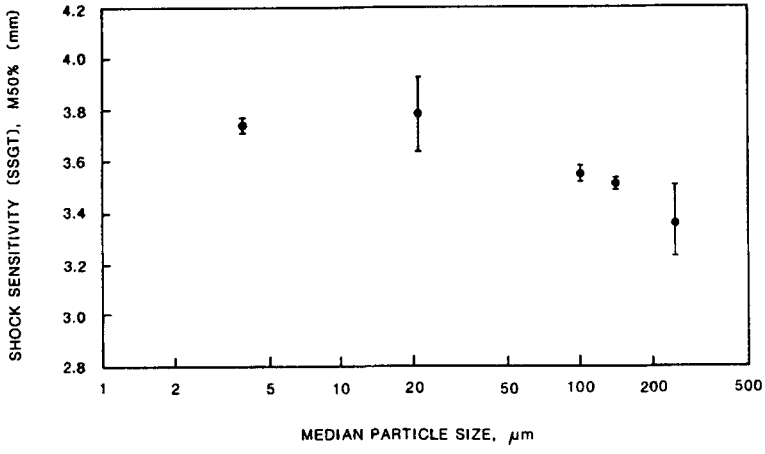

FIGURE 6 A plot of shock sensitivity (SSGT, $M_{50\%}$ in mm) against median particle size (logarithmic scale) for five RDX samples pressed nominally to 90.0 %TMD. Bars represent calculated standard deviations.

standard deviation on the Grade E result makes this comparison not statistically significant. The large uncertainty in the Grade E result made examination of material of intermediate particle size, eg 12.5 μm, of little relevance. Although study of smaller (than 3.9 μm) RDX would have been very useful, the only possibility of obtaining sufficient material for a gap test was from the milling facilities at WSRL; shipment to MRL under water would have resulted in crystal growth to approximately the same size as the 3.9 μm RDX.

Scott[43] examined three RDX sieve cuts pressed to 1.517 Mg/m^3 (84 %TMD); 74–125 μm RDX was more shock sensitive than 177–250 μm RDX, but both were substantially more shock sensitive than sub–44 μm RDX. Similarly Roth[44] found the shock sensitivity trend for RDX fractions pressed to 1.54 Mg/m^3 (85.3 %TMD) to be 330 μm < 54 μm < 25 μm , but 8 μm RDX was substantially less shock sensitive. de Longueville et al[36] studied coarse (200–400 μm) and fine (40–80 μm) RDX pressed to 1.55 Mg/m^3 (86 %TMD). For sustained shock delivered by flyer plate they found the coarse RDX to have a lower shock threshold than the fine RDX. Chick[25] studied shock sensitivity of HMX pressed to 1.14 Mg/m^3 (60.0 %TMD) using a SSGT similar to the MRL SSGT. Coarse (176 μm) HMX had a higher shock threshold than fine (8.8 μm) HMX[25].

Although the trends observed here and in the earlier studies on RDX are similar, we did not observe the substantial decrease in shock sensitivity for our finest material that Scott[43] and Roth[44] detected. Densities in the three studies were similar. There are

two possible explanations for these differences.

(i) The Grade E and ball-milled RDX are clearly different in crystal form from the Grade A RDX sieve fractions (Figs 4 and 5). In particular the ball-milled RDX represents very rough, imperfect material in contrast to the smooth surfaced Grade A RDX. Such crystal imperfections have been shown to substantially increase the shock sensitivity of HMX[60]. Thus the result for the ball-milled RDX could be a compromise between a particle size effect leading to much reduced shock sensitivity while the "rough" crystal form with increased surface area enhances shock sensitivity.

(ii) The SSGTs used by Scott[43] and Roth[44] used a confined RDX acceptor, in contrast to unconfined acceptor pellets used here. Ignition probability will not depend on confinement, but buildup will[61]. The confined tests are largely tests of ignition probability[61], whereas the unconfined MRL SSGT will introduce an additional factor from buildup due to greater energy losses from side rarefactions. Since buildup becomes increasingly more favourable at smaller particle size/higher surface area, this will compensate for decreased ignition probability at very small particle size, producing a curve like Fig. 6.

It is interesting to note that the trend observed here and by Scott[43] and Roth[44] is also followed for buildup time in shocked

low density (0.4 Mg/m^3) RDX[62] and run distance in DDT of HMX[63-65]. These combined results qualitatively support the modelling predictions of Taylor[9,10].

The relationship between particle size and shock sensitivity determined by gap test has been extensively studied for TATB[37]. The clear trend is for decrease in shock sensitivity with decrease in particle size over the range 81-10 μm. The conclusion reached was that shock sensitivity to sustained shock correlated inversely with specific surface area (SSA)[37]. Dinegar[45] had earlier noted only a very small reduction in shock sensitivity for low density PETN as SSA was increased over a very substantial range. The contrast in the behaviour of TATB, a very unreactive energetic material, with very reactive RDX or PETN probably arises because of response to hot spot variation; whereas RDX or PETN will pick up from many hot spots produced by sustained shock, TATB has very specific requirements for hot spot size and intensity. Thus any generalisation of behaviour to sustained shock must take into account the nature of the energetic material.

CONCLUSION

Shock initiation of heterogeneous pressed explosives has been reviewed. Mechanisms for initiation are reasonably well understood but the relative importance of void collapse and frictional processes are not known with certainty. Study of the molecular processes associated with initiation has only just begun. Initiation is followed by buildup which occurs by grain

burning. There is considerable disagreement as to whether the rate of grain burning or physical processes such as permeability, thermal conduction, convection etc are the key parameters controlling this process. It is frequently observed that the critical energy criterion P^2t = constant is obeyed by many explosives for short pulse duration shocks but not sustained shock. This observation coupled with density effects has been interpreted to mean that buildup is the dominant process determining success/fail for short duration shocks while initiation is the key process under sustained shock. These conclusions are supported by modelling studies.

The effect of particle size on shock sensitivity is not straightforward. Although the statement "fine powders are often harder to ignite than coarse powders, but reactions in fine powders grow to detonation more rapidly once ignited[8]" is largely correct, pressing density and shock duration strongly influence particle size effects. Heterogeneous explosives pressed to high relative density (> 98 %TMD) increase in shock sensitivity with decrease in particle size. At lower relative densities (≤ 95 %TMD) results from gap tests (sustained shock) often indicate lower reaction thresholds for larger particle materials but the reverse if $M_{50\%}$ values are compared. Smaller particle size materials invariably are more shock sensitive to short duration shock.

Five RDX powders of narrow particle size range, median 250, 138, 100.0, 21.5 and 3.9 μm, were studied to further define

particle size effects on shock sensitivity. Under sustained shock at 90.0 %TMD shock sensitivity increased with decreasing particle size; 250 < 138 < 100.0 < 21.5 μm, while the 3.9 μm RDX showed a small decrease in sensitivity. These results together with those previously published[43,44] support the prediction of Taylor[9,10] for shock sensitivity under sustained shock to decrease at very small particle size.

We are currently extending these studies to initiation by short duration shocks delivered by flyer plate, including very fine RDX.

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