

EFFECTS OF PARTICLE BEAMS ON EXPLOSIVES

BY J. SHARMA AND B. C. BEARD RESEARCH AND TECHNOLOGY DEPARTMENT



DECEMBER 1991

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FOREWORD

NAVSWC TR 91-682

This report summarizes the results from a series of experiments to determine the effects of proton and electron beams on explosives. The work on electron beam interaction was supported by SPAWAR Command of the Navy (Directed Energy) under the Charged Particle Beam Program. Work on proton beams was supported by Defense Nuclear Agency managed by the Air Force Weapons Laboratory at Kirtland Air Force Base. The authors are thankful to Dr. A. Stolovy of the Naval Research Laboratory for irradiating TATB in the LINAC accelerator during the initial phase of this work, as well as the use of his reaction chamber. The authors would like to thank Dr. D. Loughran of the Los Alamos National Laboratory (LANL) and Dr. A. Renlund of Sandia National Laboratories, for providing samples of explosives at their facilities. The authors are thankful to Dr. H. Cady of LANL for providing us with single crystals of explosives.

Approved by:

Carl E. Mueller

CARL E. MUELLER, Head

Materials Division

ABSTRACT

For application of particle beams as weapons it is essential to know the consequences of beamexplosives interactions. In the present work explosives have been subjected to particle beams of varied parameters below the level of ignition and the consequent chemical and physical changes have been determined. It has been found that in primary explosives (lead azide and lead styphnate) thermal ignition can be achieved regardless of confinement. However, in the case of high explosives such as TNT, HMX and TATB, confinement is required for thermal ignition. Without confinement other changes such as crystal phase transitions, melt-flow, evaporation, spallation, pyrolysis, fragmentation and chemical decomposition occur. These phenomena could conceivably disable a warhead indirectly by adversely effecting the performance of its explosives.

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INTRODUCTION

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Although radiation effects on explosives have been studied by many workers in the last 70 years (for a good review article see L. Avrami [1]), more studies are needed before particle accelerators can reliably be used as weapons to destroy incoming missiles, through an attack on their explosives. Lethality depends on the beam parameters, which can vary over a wide range in terms of beam energy, current density, duration and the energy deposition rate. The mechanism of the beam action can be shock, thermal ignition or molecular decomposition. The results also depend on the chemical and physical properties of the explosive and its confinement. The combination of these variations make the field of study very complex, especially since it is not feasible to extrapolate the results of one explosive on to another.

Rapid delivery of energy can lead to thermal ignition, as has been shown by the work of Stolovy, et al. [2,3,4]. They have reported the thermal threshold for ignition of confined high explosives ranges between 57 cal/gm [238 J/gm or 23.8 Mega Rad (MR)] and 168 cal/gm (702 J/gm or 70.2 MR) [2]. They report similar values for proton beam effects. For primary explosives, such as lead azide and lead styphnate, the ignition thresholds were found to be 32 and 53 cal/gm [3]. As expected on the basis of the Arrhenius equation, a heating rate dependence of ignition threshold was found. This is an important result. It means that ignition can be achieved with less total energy if the energy is delivered at a lower rate.

In the present work the effects of particle beams in the regime below that of instantaneous ignition have been addressed. An unconfined high explosive may undergo many changes without ignition that effect the performance of the explosive. Under unconfined conditions energies much larger than the

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threshold values obtained by Stolovy, et al. [2-4] can be deposited into the explosive without ignition. Due to heating effects of the beam the explosives can crack, spall, melt and run, change crystal phases, chemically alter into more sensitive products and, finally, lose their explosive power. To properly design particle beam weapons for effective use it is necessary to know the response of explosives under various irradiation conditions.

EXPERIMENTAL

Explosives were irradiated at various accelerator facilities. The LINAC at the Naval Research Laboratory (NRL) was used in some preliminary experiments with TATB. In this particular series of experiments, the irradiation was carried out over a period of hours to a dose range of 5×10^7 to 6×10^9 R. The sample temperature never rose more than a few degrees above room temperature. The purpose was to observe changes caused by radiation alone, without the effects of accompanying heat. Other parameters were the same as described by Stolovy, et al. [2].

Subsequent experiments with pulsed electron beams were carried out at ETA, PHERMEX and RADLAC accelerators. Proton beam experiments were carried out at Brookhaven National Laboratory using the 200 MeV accelerator at the Radiation Effects Facility. The samples in all cases were unconfined 5 mm diameter pellets varying in thickness from 1 mm to 5 mm. Pure explosives pressed to $\sim 97\%$ theoretical maximum density (TMD). The explosives studied included both primary (lead styphnate and lead azide) and secondary (HMX, RDX, TATB, HNS, NTO and TNT) explosives as well as the oxidizer ammonium perchlorate. During each experiment the explosive pellets were loosely placed in cylindrical holes (6mm diameter) in graphite. The samples were covered with a 2-3 mils thick chlorostyrene film, which changed color in proportion to the irradiation dose delivered to the sample.

The coloration of the chlorostyrene film was measured with a microdensitometer. The irradiated samples were analyzed for chemical changes with x-ray photoelectron spectroscopy (XPS), thin layer chromatography (TLC) and chemical ionization mass spectrometry (CIMS).

RESULTS AND DISCUSSIONS

LOW RATE ENERGY DEPOSITION - ELECTRON BEAMS

This study for the first time showed that explosive molecules can convert into intermediate solid state products, rather than immediately degrading to gaseous products. Detectable decomposition for TATB appeared at a dose of 10⁷-10⁸ R. TATB was partially converted into mono and difuroxan derivatives (Figure 1). Mono and difurazan derivatives were also found. The furoxan product is far more sensitive than the original explosive. The furazan derivatives are the same in sensitivity as TATB [5], but have less explosive power. The results on TATB indicate that the effect of irradiation without bulk temperature rise is through molecular changes. Radiation damage is intimately connected with "hot spots" of microscopic dimensions, as described by Phung [6]. The dose required for such decomposition is different for different explosives. Similar results were obtained for TNT, RDX and HMX.

HIGH RATE ENERGY DEPOSITION - ELECTRON BEAMS

High explosives were subjected to pulsed mode electron beams at ETA, PHERMEX, and RADLAC accelerators. It was found that at a threshold of 1-10 cal/gm the explosives evidenced cracking

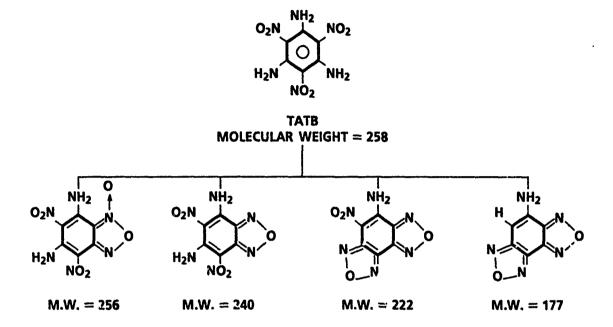


FIGURE 1. MOLECULAR STRUCTURE OF TATB AND ITS REACTION PRODUCTS

(Figure 2). If the yield strength of explosives (ranging between 0.1-1 kbar) and the Gruneisen constant of unity are considered, then this dose range is just about right for physical disintegration of the solid. Experiments with single crystals as well as plastic bonded explosives showed fragmentation in the same range of energy deposition. It was observed that if the beam diameter was smaller than the explosive pellet, the cracks ran radially from the area of the beam. If the beam was larger in area than the explosive pellet, a plain shock wave was apparently generated causing planar spallation of the sample into thin discs as shown in Figure 3. In some cases, a 5 mm thick pellet opened into as many as four

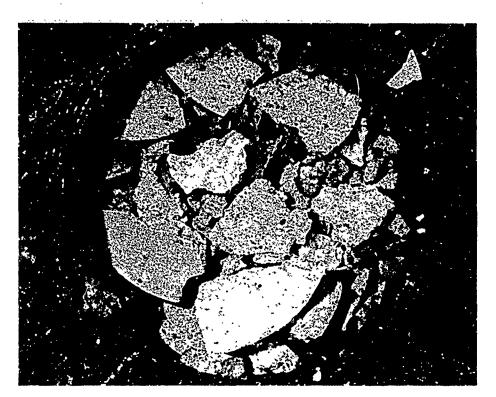
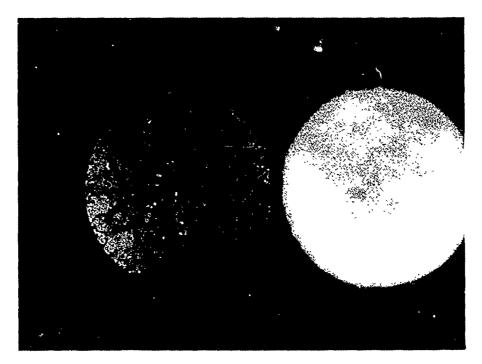


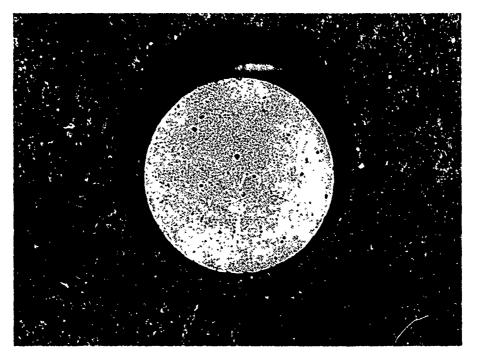
FIGURE 2. FRAGMENTATION OF A 5 mm DIAMETER, 1 mm THICK PRESSED PELLET OF TATB

discs of approximately equal thickness. This effect is of great significance. If the explosive is used to generate a controlled shockwave, the physical break-up of the explosive is likely to cause mission failure.

With a single pulse electron bean, ignition of unconfined TATB could only be achieved at RADLAC. The threshold energy for ignition was bracketed between 125 and 204 cal/gm. The value of 204 cal/gm is rather high compared to the results of Stolovy [2]. Probably during the short duration of the pulse the explosive provides its own confinement. Multiple sub-ignition dose pulses (as many as four) did not show evidence of the formation of more sensitive decomposition products. So far no explosive has evidenced the expected effect of sensitization where previous pulses lower the threshold for subsequent pulses.



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NOTE: DISK SEPARATED AT CRACK

FIGURE 3. SPALLATION OF A 5 mm DIAMETER AND 1 mm THICK TATB PRESSED

PELLET BY A SUB-MICROSECOND PULSED ELECTRON BEAM

Sub-ignited TATB showed color change, spallation and fragmentation. TNT in similar experiments showed crater formation, melting and recrystallization along with partial decomposition into trinitrobenzyl alcohol and anthranil. The trinitrobenzyl alcohol product is more sensitive than TNT. In the case of RDX, gaseous products are most common but as much as 10 percent conversion to the solid nitroso derivative of RDX was observed in irradiated samples.

HIGH EXPLOSIVE - PROTON BEAMS

Proton beam experiments on high explosives (secondary) were carried out with HMX, RDX, TNT, TATB, HNS and NTO. During this study, the rate of energy deposition was approximately 1-1.5 cal/gm-s, yielding a temperature rise of 5-7.5 °C/s. These experiments showed that unconfined secondary explosives would not explode or ignite, even when more than ten times the energy threshold for confined samples was deposited. Some of the samples showed as high as 30 percent decomposition, yet did not undergo thermal explosion (Figure 4). As a result of heating, the samples fragmented, melted, flowed in the sample cavity, evaporated, and decomposed. Foam like samples of RDX and HMX resulted from melting and recrystallization (Figure 5). Analysis of this HMX sample demonstrated partial decomposition (Figure 6). The HMX was also found to have been converted into the delta phase which is more sensitive than normal (beta) HMX. Drop test experiments with C.S. Coffey's Ballistic Impact Chamber machine [7] showed that the rate of burning has also decreased due to degradation of the explosive. In general the effect on the explosive was the same as that of controlled partial thermal decomposition.

From these experiments it was possible to determine the relative role of heat and that of radiation damage. In a typical experiment of Stolovy, when a sample ignites at an energy deposition of say

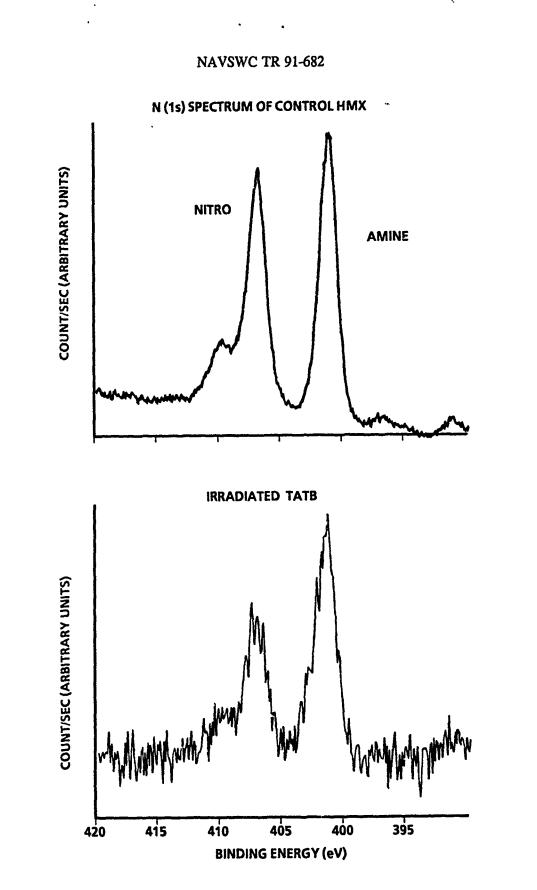


FIGURE 4. X-RAY PHOTOELECTRON SPECTRA OF NITROGEN (1s) CORE LEVEL IN TATB

100 cal/gm, the radiation dose is approximately $4 \ge 10^7$ R. According to Avrami [1] this dose would cause only a few percent loss of weight. Therefore, the major effect on the explosive is by heat. If the rate of energy deposition is decreased, then the relative role of thermal effects will decrease and that of radiation damage will increase.

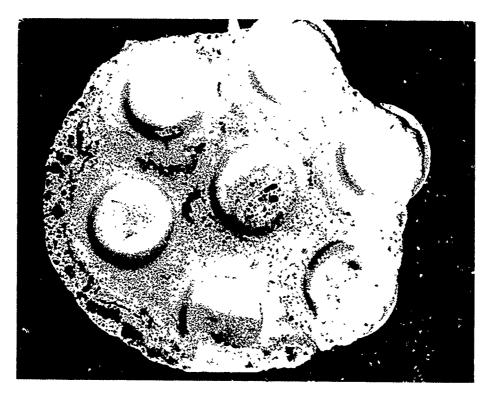


FIGURE 5. MELTED AND RECRYSTALLIZED HMX

PRIMARY EXPLOSIVE - PROTON BEAMS

When experiments on primary explosives such as lead azide, lead styphnate, PETN and ammonium perchlorate were carried out, although the rate of energy deposition was rather low (1.2 cal/gm-s) and the explosives were unconfined, all of them showed ignition. Thus the primary explosives do not demonstrate an ignition energy dependance on confinement. The threshold energies are: Lead azide, 24 cal/gm; Lead styphnate, 25 cal/gm; PETN, 144 cal/gm; and AP, 64 cal/gm.

The values for lead azide and lead styphnate compare well with those of Stolovy, et al. [3]. The important point is that confinement does not make any difference in the case of primary explosives.

In the case of PETN, the results can be deceptive due to the low melting point. When PETN was not allowed to flow out of the irradiation area it ignited at 144 cal/gm. AP ignited at 64 cal/gm, but part of the sample survived reaction, only about half of the pellet was consumed. The left-over sample showed about 3.6 percent of the chlorine to be present in a (+1) chemical state.

In order to determine if sensitization from previous irradiation comes into play, the irradiation was broken up into two installments, each being 80 percent of the measured ignition level. Two 80 percent threshold irradiations were delivered, separated by a cool down period. None of the primary explosives showed sensitization from the first irradiation. In fact, during one experiment lead azide which had been given 80 percent of the ignition dose, required more energy (31 pulses) to ignite than that of a virgin sample (21 pulses). The question of sensitization of the explosives is difficult and will require further investigation. Perhaps the sub-ignition doses thermally degraded the material instead of making it more sensitive.

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N (1s) SPECTRUM OF CONTROL HMX COUNT/SEC (ARBITRARY UNITS) NITRO AMINE HMX AFTER 2 TRAINS OF PULSES 305, 258 FROM BNL-REF COUNT/SEC (ARBITRARY UNITS) 415 410 405 400 395 4**2**0

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FIGURE 6. X-RAY PHOTOELECTRON SPECTRA OF NITROGEN (1s) CORE LEVEL IN HMX

BINDING ENERGY (eV)

SUMMARY OF RESULTS

HIGH EXPLOSIVES

Slow rate energy deposition where the temperature rise is insignificant causes partial chemical decomposition. Two kinds of products have been observed, some make the explosive more sensitive while others decrease the explosive power.

At an energy deposition rate of 8-10 J/s, producing a temperature rise of about 30-60°C/s, unconfined high explosives failed to ignite despite the energy deposition of ten times the ignition threshold of confined explosive. This result demonstrates the strong dependance of ignition threshold on confinement.

The chemical and physical changes resulting from irradiation can profoundly alter the behavior of explosives causing the failure of the explosives in a weapon even in the absence of ignition. The exact consequence would depend on the amount of energy deposited, the explosive and the purpose of the explosives within the weapon.

In experiments of high rate deposition the main decomposition is by heat rather than radiation. The role of pure radiation damage (via ionization effects) is estimated to be a few percent of the thermal effects.

If sufficient energy is deposited in a single pulse of micro-seconds duration, high explosives can be ignited, regardless of confinement conditions. It appears that at such rates of energy deposition the explosive can provide its own confinement.

No sensitization effects in multi-pulse irradiations were observed.

PRIMARY EXPLOSIVES

The primary explosives showed ignition when energy was delivered at 1.2 cal/gm-s regardless of confinement.

Thermal effects dominate in the ignition process. Hardly any difference between the effects of electrons or protons is noticeable, in terms of ignition threshold.

No sensitization effects in multi-pulse irradiations were observed.

CONCLUSIONS

The studies at low rate of irradiation have shown that the changes caused in secondary explosives arise from chemical alterations of the molecules. This finding explains many observations made by previous workers [1] in the field of radiation damage. Shifts in explosion temperature, sensitization and loss of power for TATB can be easily explained as due to the transformation of the molecules into furoxan and furazan. Furoxan derivatives of TATB are more sensitive than TATB. Furazan derivatives are less powerful. Altered molecules have been observed in other explosives such as TNT, RDX and HMX.

The present work has also demonstrated many physical effects caused by intense and heat generating beams. Melting, phase transitions and fragmentation can explain sensitization, loss of power and other changes observed previously.

The dependence of ignition threshold on the confinement of secondary explosives is not surprising, because ignition is known to be controlled by the charge size and strength of external confinement. It is noteworthy that the confinement effect for explosives under irradiation is dependent on the rate of energy deposition.

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The results mentioned show that the effects of irradiation can cover a wide range of consequences, depending on the nature of the beam and the explosive. In order to develop a predictive capability more experiments are needed. The present work was confined to pure explosives; in the future different formulations should be investigated, particularly plastic bonded explosives.

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