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EFFECT OF A HIGH-ENERGY ELECTRON BEAM
ON THE DETONATION VELOCITY OF AN EXPLOSIVE

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October 4, 1976

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EFFECT OF A HIGH-ENERGY
ELECTRON BEAM ON THE DETONATION
VELOCITY OF AN EXPLOSIVE

ABSTRACT

As part of a program to study the mechanism of detonation in explosives, the effect of a pulse of high-energy electrons on an explosive's detonation velocity was examined. A short, intense burst of 1.1-MeV electrons was directed onto a specimen of XTX-8003 explosive in a narrow track as a detonation wave was passing along it. No change in the detonation velocity resulted from an electron energy fluence of 1.4 J/cm^2 .

INTRODUCTION

Walker and Wasley¹ have proposed a model for initiation of explosives, a major feature of which is the generation of ions and free radicals in a shock front by nonthermal processes. This phenomenon is proposed as a primary mechanism in the process leading to detonation. To test this idea, LLL's Organic Materials Division is studying the effect of free-radical-producing additives on the properties of explosives.

In the experiment described in this paper, high-energy electrons were used to generate ions, excited molecules, and free radicals in an explosive. The purpose was to provide an excess of excited species without altering the chemical make-up of the explosive.

The interaction of radiation with matter and the reactions of the products have been extensively discussed.²⁻⁵ Briefly, under the conditions of our experiment, the interactive events to be expected are these: Energetic electrons in the beam will impart some of their energy to electrons in the atoms in the explosive that they pass close to, and some of these initially bound electrons will be removed from their atoms, becoming free electrons in the system along with the beam electrons. As all of the free electrons become degraded to a low energy, they will be trapped or will combine with molecules. Thus, as a result of the passage of the electron beam through the explosive, some molecules will have lost electrons, becoming positive ions, and some will

have gained electrons, becoming negative ions.

These positive and negative molecular ions are free radicals because they have unpaired electrons. These free radicals can dissociate into smaller fragments; also, along with ionization, there will be many lower-energy excitation events that produce atoms and free radicals directly.

The "events" described above are concentrated in a narrow channel along the path of each energetic electron in the entering beam, and the energy of the electrons in the beam determines the depth of penetration and reaction density in a given target.

Both Eyring et al.⁶ and Wood and Kirkwood⁷ have derived expressions for the dependence of the detonation velocity, D , on the reaction-zone length and the radius, R , of a high-explosive charge in which the detonation wave proceeds along the axis of the charge. The expressions from Refs. 6 and 7 are here reduced to a simple formula, from which the "effective" reaction-zone length, λ , may be solved:

$$\frac{D}{D_{\infty}} = 1 - \frac{\lambda}{R}.$$

If the excited species created by the electron beam increase the reaction rate, and if λ is made correspondingly shorter, then the wave-front curvature will decrease as described in Refs. 6 and 7, and the detonation velocity D will increase toward the asymptotic value D_{∞} associated with infinite charge radius. A careful measurement of D/D_{∞} for a specified R will thus yield relative values for λ and the reaction rate that is related to it.

EXPERIMENTAL

In this study, we attempted to measure, by the Dautriche technique,⁸ a difference in detonation velocity between irradiated and unirradiated specimens of explosive in narrow tracks.*

The tracks consisted of XTX-8003 plastic explosive formed into slots of square cross section machined in a brass plate. XTX-8003 is 80% PETN and 20% silicone rubber; some of the properties of PETN and XTX-8003 are given in the Appendix. The means of irradiation was a beam of 1.1-MeV electrons, directed onto the tracks as the detonation wave passed along them.

*In this paper, "track" refers to a rod of explosive. We used a rectangular cross section; the theory and some other work refer to a circular cross section.

Method

A nonelectrical technique for comparing the detonation velocities in the tracks was needed because of the electrical noise that was produced, for an appreciable period after the instants of firing, both by the capacitor bank used for firing the explosive-track detonator and by the electron accelerator. The technique we used was based on the sharp pressure gradient produced at the point where two detonation fronts meet in a track of explosive; the principle can be summarized as follows:

As a detonation wave progresses down an explosive track consisting of a narrow slot in a brass plate, the high pressure at the shock front compresses the metal around the track beyond its elastic limit. This is particularly noticeable at the outer edges of the slot, where small local inhomogeneities in the fracture strength of the brass lead to a jagged edge, whereas the bottom of the slot remains relatively smooth.

Our experimental configuration provided for separate detonation fronts, starting from a common origin, to travel down pairs of parallel pathways; if the detonation fronts have been led down separate tracks, they can also be caused to meet again (see Fig. 1). At the point where they meet, the high pressure at the shock front may quadruple. Because the shock front is very thin, the pressure gradient is sharp, producing a narrow depression in the metal across the bottom and up the sides of the slot. The depression is visible in the sides and outer edges of the slot, but it is most easily observed and measured under the microscope in the smooth bottom of the slot.

By precision machining of the slots, one can know their lengths precisely. If two tracks whose lengths are known precisely are connected so that their detonation waves meet, the point where the waves meet can be used to determine any difference in the detonation velocities of the two tracks. Thus the effect of different cross sections or, as in this experiment, the effect of irradiating one of the tracks can be precisely measured.

The general specimen configuration and procedure chosen for the irradiation experiment are illustrated in Fig. 2. The subject tracks are labeled A, B, C, and D. The wider tracks leading from the detonator to A, B, C, and D and interjoining them at the bottom serve as connectors and as the sites where the detonation fronts converge, leaving witness marks.

Tracks A and B are of a width chosen to provide an experimentally established, known velocity near D_{∞} . Tracks C and D are of a narrower

Fig. 1. Where two detonation fronts meet in a narrow slot, reinforcement of the shocks causes a sharp pressure gradient that produces a narrow depression, or "witness mark," across the bottom and up the sides of the slot.

Fig. 2. General configuration of the specimen for the irradiation experiment. Arrows indicate the directions of propagation of the detonation fronts. See text for a description of the procedure.

width, chosen to reliably provide a velocity as much lower than D_{∞} as practicable.

The beam from the electron accelerator is shaped by an aperture plate so that it strikes only the part of the specimen plate indicated with cross-hatching; thus only tracks B and C are irradiated.

The connecting tracks are arranged to provide the following velocity comparisons:

1. A vs B: unirradiated vs irradiated wide tracks
2. D vs C: unirradiated vs irradiated narrow tracks
3. A vs D: wide vs narrow unirradiated tracks
4. B vs C: wide vs narrow irradiated tracks

The witness marks at the points where the detonation fronts meet in the various connecting tracks are the data for the velocity comparisons; the known velocity of track A is the reference velocity, and the velocities of the other three tracks can be calculated with respect to it on the basis of the locations of the witness marks. The comparison calculations can proceed in either direction around the circuit: $A \rightarrow B \rightarrow C$ or $A \rightarrow D \rightarrow C$.

Slot Dimensions

Before doing the irradiation experiment, we performed several detonation experiments to measure the detonation velocity of XTX-8003 in square slots of various sizes.

A series of plates like that shown in Fig. 3(a) was prepared. After the slots had been filled with XTX-8003, the excess explosive was removed with a razor blade; then after several days the silicone rubber cured to a firm consistency.

EX-12 detonators (43 mg PETN, 23 mg 9407, stainless-steel shell) were used for these velocity-measurement experiments. [In a preliminary experiment, an SE-12 Mod 6 detonator (30 mg PETN, polycarbonate shell) did not detonate the XTX-8003. This demonstrates that a high pressure and temperature are needed to initiate XTX-8003.]

Three techniques were used to measure the detonation velocities:

1. Piezoelectric pins placed at the start and the finish of a track; pins are visible at the ends of the outer tracks in Fig. 3(b).
2. Insulated #40-gage wires placed across and close to the inner tracks; also visible in Fig. 3(b).
3. The reinforcement-wave method, as described above; see Fig. 3(c).

Fig. 3. Typical shot in the preliminary velocity-measurement series. Numbers on the scales indicate centimeters. (a) Brass plate with slots. Plate is 12 mm thick, ductile, machinable. (b) Plate with explosive, crystal pins, and wires. Detonator and cables to pins are still to be added. Connecting tracks are 1.6 mm wide, 1.0 mm deep. Tracks on right are 0.25 mm square; tracks on left are 0.36 mm square. Wires are connected to oscilloscopes through 50- Ω resistors. Base plate bias = 24 V. (c) After the shot. Wave convergence witness marks (barely visible on the original photo) were sharp under the microscope and were measured to within 20 μ m.

The results are plotted in Fig. 4 as detonation velocity vs cross-section side dimension, along with the failure-diameter curve from the original description of XTX-8003, prepared by A. Popolato at Los Alamos in 1964.⁹ The difference between our curve and Popolato's curve is due to differences in the test setups: Popolato used confined cylindrical tracks in plastic, whereas we used square tracks in brass with one face open to the air. Our setup needed a smaller track for a given detonation velocity and entailed a smaller failure dimension.

The data in Fig. 4 show that, under our test conditions, a track having a side dimension of 0.25 mm (0.010 in.) reliably provides a detonation velocity appreciably lower than D_{∞} but is close to the failure dimension. At this dimension, the explosive has to be placed very carefully in the slot to avoid flaws, particularly voids, that would prevent the detonation wave from propagating.

On the basis of these velocity measurements, we chose the 0.25-mm track for use in the irradiation experiment, in conjunction with the 0.64-mm (0.025-in.) track to provide a velocity of 7.26 km/s, which is near D_{∞} , as the reference velocity.

Electron Source

The electron source was a Physics International Pulserad #422. It was focused so that it would uniformly irradiate tracks that were 88 mm long. The intensity had to be adjusted so that the explosive was not overheated; a high temperature would have made the reaction dependent on thermal rather than non-thermal generation of excited species.

The anode-to-cathode distance was 24 mm. The graphite cathode was 10 mm in diameter, with a bullet nose. The titanium anode was 0.13 mm thick, with 0.13-mm-thick Kapton sheet used as a vacuum seal. The drift-vessel pressure was 270 Pa. The front face of the aperture plate was set 152 mm from the outer face of the drift vessel. The beam guide in the drift vessel was removed to provide a wider aperture.

A typical electron-energy distribution is shown in Fig. 5. The burst time is almost 50 ns. Energy fluence deposited in the plane of the explosive tracks was determined separately using a calorimeter consisting of an array of carbon blocks each monitored by a thermocouple. The resulting broad beam was uniform over a 100-mm diameter, and the average energy fluence was 1.4 J/cm^2 .

Fig. 4. Detonation velocity of XTX-8003 vs side dimension of square cross section.

Fig. 5. Typical energy distribution in electron beam from accelerator.

Irradiation Experiment

The brass plate used in the electron-beam experiment is shown, as it appeared after the shot, in Fig. 6(a). Before the shot, it had a pair of 0.25-mm tracks on the right and a pair of 0.64-mm tracks on the left.

The aperture plate, shown in Fig. 6(b), was used to shape the electron beam, thereby defining the lengths of the explosive tracks that were exposed to the beam (see Fig. 2). These were the two inner tracks, each of which was irradiated over a length of 85 mm. The aperture plate was positioned 7.6 mm in front of the specimen plate.

The capacitor discharge unit that fired the detonator was operated via a manual switch. The detonation wave traveled down equal connecting paths to the heads of the four parallel subject tracks. At the head of the inner 0.25-mm track, a piezoelectric pin was actuated by the detonation wave as it passed; this actuated the electron source, after a delay of 1.26 μ s, and the electron beam irradiated 85 mm of the two inner tracks for 50 ns. The detonation traveled 8.9 mm down the 0.25-mm track during the delay and another 0.4 mm during the pulse; then the wave traveled the rest of the 88.3-mm-long track after the electron pulse.

RESULTS

With a traveling microscope, the positions of the reinforcement waves on the brass plate were measured with respect to fiducial marks that had been scribed on the surface. The fiducial marks had been accurately measured with respect to the tracks that were to be compared. If two tracks had exactly the same velocity, the waves would meet exactly at the midpoint between the tracks. Thus a displacement of the meeting point from the midpoint gave the velocity difference between the two tracks.

The velocity of the unirradiated 0.64-mm track, the outside track on the left in Fig. 6(a), was assumed to be 7.26 km/s, the value obtained in the earlier velocity measurements. From this value, on the basis of the locations of the meeting points of the various pairs of detonation waves, the velocities of the other three tracks were calculated. The results are shown in Table 1; they were the same in either direction around the circuit of comparisons from the reference track.

The reinforcement waves are vividly indicated by the marks of the detonation products on the aperture plate, as shown by Fig. 6(b). Although the

(a)

(b)

Fig. 6. (a) Specimen plate from irradiation experiment, after the shot. Numbers on the scale indicate centimeters. (b) Top and bottom portions of aperture plate after the shot. The aperture plate was placed 7.6 mm in front of the specimen plate; seen here is the side facing the specimen. Numbers on the scales indicate centimeters.

resolution is not as great as in the wave marks in the slots of the brass plate, they clearly substantiate the implication of Table 1. The marks between the tracks of the same width are close to the midpoints. The marks between tracks of different widths are displaced toward the narrower tracks, showing that the detonation velocity was greater in the wider tracks.

Table 1. Summary of velocities in tracks.

	Velocity (km/s)	
	0.64-mm tracks	0.25-mm tracks
Control	7.26 ^a	7.06
Irradiated	7.25	7.00

^aReference velocity.

From the energy-spectrum curve of Fig. 5, the part of the beam absorbed by a 0.25-mm thickness of XTX-8003 was calculated to be 0.13 J/cm^2 . This is an energy density of 5.2 J/cm^3 and would cause a temperature rise of 3°C . If each ion pair is formed by $8.0 \times 10^{-18} \text{ J}$ (50 eV), such an energy density would form 6.4×10^{17} ion pairs/cm³. PETN (chemical formula: $\text{C}_5\text{H}_8\text{N}_4\text{O}_{12}$), with a molecular weight of 316, has 2.9×10^{21} molecules/cm³. So there should be one ion for every 2300 molecules. Each ion produced will also be a free radical, since it has an unpaired electron.

DISCUSSION

We have observed that 1.1-MeV electrons, at a fluence level of 1.4 J/cm^2 , have no significant effect on the detonation velocity in explosive tracks. Possible reasons for the lack of an observable change in reaction rate and hence in reaction-zone length are:

1. The electron beam may have low efficiency for producing the appropriate excited species. The calculation cited above indicates that 1.4 J/cm^2 of electrons with energies near 1 MeV will produce one radical per 2300 molecules of PETN.

2. The lifetimes of the ion radicals produced by the 50-ns pulse of electrons may be short compared with the time it takes the detonation wave to traverse the track. However, Refs. 2-5 and 10, in discussing the interactions

of radiation with matter, point out that these ion radicals should have a low recombination rate because of their slow diffusion in the solid matrix.

3. Ion radicals may be ineffective--compared with "molecular fragment" free radicals--in initiating detonation reactions. Walker¹¹ has suggested that the ions are too large and immobile to reach the reaction sites. However, the high-energy electrons impart excess energy to the molecules as well as ionizing them. This energy can subsequently dissociate these excited ion radicals into smaller fragments. The relative importance of these processes and the level of radical concentration to produce significant rate effects will require further research.

4. A concentration of about 4×10^{-4} free radical per molecule of explosive may not be sufficient to change the reaction rate enough to be detected. To be effective at this concentration, a chain reaction, initiated by one of the radicals, would be required. Such a chain reaction ($F_2 + H_2$), initiated by free radicals generated by an electron beam, is described by Gerber and Patterson.¹²

FUTURE WORK

Because of the difficulty of adjusting the beam aperture and recalibrating the beam intensity, the study reported here was limited to a single integrated flux intensity. A logical extension of this work would be to attempt to change the detonation rate by increasing the electron intensity and hence the free-radical concentration. The heating effect of the beam could be compensated for by control experiments in which heat alone is used.

Experiments discussed so far have been based on the use of a single short pulse of electrons. If, as we expect, the lifetime of free radicals and ions is relatively long, multiple pulses might also be used to increase the concentration.

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acknowledge R. Ward's helpful discussion of the production of ions and free radicals.

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APPENDIX:
PROPERTIES OF PETN AND XTX-8003

Source: *Properties of Chemical Explosives and Explosive Simulants*, compiled and edited by B. Dobratz, Lawrence Livermore Laboratory, Report UCRL-51319 Rev. 1 (1974).

EXPLOSIVE: PENTAERYTHRITOL TETRANITRATE	DESIGNATION: PETN
2. STRUCTURE OR FORMULATION	6. THERMAL PROPERTIES (continued)
	T_g ($^{\circ}\text{F}$ (K)): none C_p (cal/g- $^{\circ}\text{C}$ (kJ/kg-K)): Exp.: 0.26 (1.088) Thermal stability (cm^3 of gas evolved at 120 $^{\circ}\text{C}$ (393 K)): 0.25 g for 22 hr: 0.10-0.14 1 g for 48 hr: —
4. PHYSICAL PROPERTIES	8. DETONATION PROPERTIES
Physical state: solid Color: white At. comp.: $\text{C}_5\text{H}_8\text{N}_4\text{O}_{12}$ MW: 316.2 Density (g/cm ³): TMD: 1.77 Nominal: 1.76 m.p. ($^{\circ}\text{C}$ (K)): 139-142 (412-415) b.p. ($^{\circ}\text{C}$ (K)): — v.p. (mm Hg (Pa)): 8×10^{-5} at 100 $^{\circ}\text{C}$ (1.1×10^{-3} at 373 K) $\log_{10} P_{\text{mm}} = 14.44 - [6352/T \text{ (K)}]$ Crystal data: I: tetragonal (P4 ₂ /c) II: orthorhombic (Pcnb) a = 9.38 a = 13.22 b = 9.38 b = 13.49 c = 6.71 c = 6.83 R: —	D (mm/ μsec (km/s)): 8.26 ($\rho = 1.76$) P_{CJ} (kbar (10^{-1} GPa)): Meas.: $\frac{\rho = 1.77}{340}$ $\frac{\rho = 1.67}{300}$ $\frac{\rho = 0.99}{87}$ Calc.: 326 280 100 E_{Cyl} ((mm/ μsec) ² /2 (MJ/kg)): ($\rho = 1.765$) 6 mm: 1.255 19 mm: 1.575
5. CHEMICAL PROPERTIES	9. SENSITIVITY
ΔH_{det} (kcal/g (MJ/kg)): H_2O (l) H_2O (g) Calc: 1.65 (6.90) 1.51 (6.32) Exp: 1.49 (6.23) 1.37 (5.73) ΔH_f (kcal/mol (kJ/mol)): -128.7 (-593) Solubility (s-sol., sl-sl. sol., i-insol.): s—acetone, DMFA, DMSO, ethyl acetate, pyridine sl—benzene, ethyl ether i—carbon disulfide, carbon tetrachloride, chloroform, ethanol, water	H_{50} (cm (10^{-2} m)): <u>12 tool</u> <u>12B tool</u> 11 — Susan test: — Skid test: Impact angle (deg (rad)) Drop ht. (ft (m)) Event — — — Gap test (mils (mm)): Small-scale: 190-220 (4.8-5.6) ($\rho = 1.757$) Large-scale: 2.732 (69.4) ($\rho = 0.81$)
6. THERMAL PROPERTIES	10. ELECTRICAL PROPERTIES:
k: — CTE: $\alpha = 46.1 \times 10^{-6}$ in./in.- $^{\circ}\text{F}$ (83.0 m/m-K) $\alpha = 76.5$ -89.9 $\mu\text{m}/\text{m-K}$ at 244-363 K $\beta = 249.2$ $\mu\text{m}/\text{m-K}$ at 243-343 K	ϵ : 2.447 ($\rho = 1.4$) 2.577 ($\rho = 1.5$) 2.897 ($\rho = 1.7$) 2.727 ($\rho = 1.6$) 2.95 ($\rho = 1.75$)
	11. TOXICITY
	High.

EXPLOSIVE: XTX-8003	DESIGNATION: XTX-8003
2. STRUCTURE OR FORMULATION	6. THERMAL PROPERTIES (continued)
<div> <div>wt%</div> <div>PETN 80</div> <div>Silicone rubber 20</div> </div>	<div> T_g (°F (K)): — </div> <div> C_p (cal/g-°C (kJ/kg-K)): <div>Est.: 0.27 (1.13)</div> </div> <div> Thermal stability (cm³ of gas evolved at 120 °C (393 K)): <div>0.25 g for 22 hr: >0.02 at 100°C (373)</div> <div>1 g for 48 hr: —</div> </div>
4. PHYSICAL PROPERTIES	8. DETONATION PROPERTIES
Physical state: putty curable to rubbery solid Color: white At. comp.: C _{1.80} H _{3.64} N _{1.01} O _{3.31} Si _{0.27} MW: 100 Density (g/cm ³): TMD: 1.556 Nominal: ≈1.53 m.p. (°C (K)): 129-135 (402-408) b.p. (°C (K)): — v.p. (mm Hg (Pa)): — Crystal data: — R: —	<div> D (mm/μsec (km/s)): 7.30 (ρ = 1.53) </div> <div> P_{CJ} (kbar (10⁻¹ GPa)): (ρ = 1.546) <div>Meas.: 170</div> <div>Calc.: 210</div> </div> <div> E_{cyl} ((mm/μsec)²/2 (MJ/kg)): (ρ = 1.554) <div>6 mm: 0.710</div> <div>19 mm: 0.950</div> </div>
5. CHEMICAL PROPERTIES	9. SENSITIVITY
<div> ΔH_{det} (kcal/g (MJ/kg)): <div> <div>H₂O (l)</div> <div>H₂O (g)</div> <div>Calc: 1.88 (7.89) 1.69 (7.07)</div> <div>Exp: 1.16 (4.85) 1.05 (4.39)</div> </div> </div> <div> ΔH_f (kcal/mol (kJ/mol)): -44.4 (-185.9) </div> <div> Solubility (s-sol., sl-sl. sol., i-insol.): — </div>	<div> H_{50} (cm (10⁻² m)): <div> <div>12 tool</div> <div>12B tool</div> <div>Cured: 21 —</div> <div>Uncured: 25 —</div> </div> </div> <div> Susan test: Threshold velocity ~ 160 ft/sec (~ 49 m/s); has very small probability of buildup to violent reaction. </div> <div> Skid test: <div>Impact angle (deg (rad)) Drop ht. (ft (m)) Event</div> <div>— —</div> </div> <div> Gap test (mils (mm)): (ρ = 1.53) <div>Small-scale:</div> <div>Cured: 130-160 (3.3-4.1)</div> <div>Uncured: 160-190 (4.1-4.8)</div> </div>
6. THERMAL PROPERTIES	10. ELECTRICAL PROPERTIES:
k: — CTE: <div> α = 68.8 × 10⁻⁶ in./in.-°F at -22 to 158°F (123.8 μm/m-K at 243-343 K) </div> <div> α = 77 × 10⁻⁶ in./in.-°F at 75 to 150°F (139 μm/m-K at 297-339 K) </div> <div> β = 413.7 μm/m-K at 219-296 K </div>	<div> ϵ: — </div>
	11. TOXICITY
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