TITLE: PRECURSORS IN DETONATIONS IN POROUS EXPLOSIVES

MASTER

AUTHOR(S): ROBERT L. SPAULDING, JR.

SUBMITTED TO: 7TH SYMPOSIUM (INTERNATIONAL) ON DETONATION



By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish of reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos Scientific Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.



LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545 An Affirmative Action/Equal Opportunity Employer

Form No. 836 R3 81, No. 2629 UNITED STATES DEPARTMENT OF ENERGY n an **n n n n** Marian An an an an an an an PRECURSORS IN DETONATIONS IN POROUS EXPLOSIVES

Robert L. Spaulding, Jr. University of California, Los Alamos National Laboratory Los Alamos, New Mexico 87545

Photographs of detonation waves in low-density HMX and PETN, made with an image-intensifier camera, sho a brilliant band of light in front of the pressure jump. The radiation temperature is estimated to be 12,000 K to 14,000 K. The spectrum of this light is continuous. A quartz gauge shows a gradual buildup of pressure from the material producing the light. The material has little effect on the propagation of detonation.

Further observations, using pellets of plastic-bonded HMX and single crystals of PETN, show that the material thrown off the free surface is transparent, with a leading edge moving at approximately 20 mm/us. Collison of this material with polymethyl methacrylate (PMMA) produces a brilliant light with a spectrum that is initially a narrow H_3 line. Quartz gauges measure the rate of pressure buildup of this material.

I. INTPODUCTION

During the past forty years, several investigations (1-3) have been made of the light recorded in photographs of detonating explosives when air shock is suppressed. Paterson (2) showed that in the absence of air shock, very bright light is emitted from detonating granular PETN, and from strongly whocked layers of NaCl, whereas only faint light is emitted by plastic explosive alone. Blackburn and Seely (3) showed that the bright light observed by Paterson in NaCl is not caused by air shock because it is undiminished in intensity when methane is substituted for air.

Later, Seely (4) showed that detonating pellets of pressed HMX and PETN emit a bright burst of light, brighter than the subsequent air shock, and that this burst of light is emitted even when the explosive is detonated in methane, helium or vacuum. He further showed that material is ejected from the surface of a large, detonating PETN crystal which produces a brilliant light when it stagnates against another PETN crystal surface. He postulated

stagnation hot spots against downstream grains to be the ignition mechanism for shock initiation of granular explosives. At about the same time, Lundborg (5) showed that, in a vacuum, some material is thrown off the end of a detonating stick of TNT with a velocity that varies with the amount of residual gas in the vacuum and reaches 20 mm/us at a pressure of 0.13 Pa. Hay, Peters, and Watson (6) then showed from spectroscopic studies that the material moving with the highest velocity is hydrogen, and it is followed by other heavier atomic species. The work discussed in this paper was undertaken to further characterize this material and to study its connection with the detonation process.

II. EXPFRIMENTS

Our observations were made with image-intensifier cameras (1^2C) (7) with each camera capable of single or multiple exposures. All exposures were made at a nominal 10 ns. Intensification gain was selectable and was varied from 10,000 to less than 100, as required by each experiment.

La our first set of experiments, HMX was the explosive used. A sample containing a large fraction of coarse crystals was sieved to obtain a coarse cut of -20/+40 and a fine cut of -70(USA Standard Testing Sieve Designation). Samples were loaded by pouring into an aluminum shot assembly (Fig. 1), which contained a detonator at one end and could be sealed and evacuated. The density of the explosive was approximately 0.95 Mg/m³, or one-half of crystal density. All shots were e-vacuated to 1.3 Pa. Figure 2 is a photograph of the coarse cut of HMX detonating. The light from the detonation is very bright and is scattered forward through the translucent, undetonated crystal aggregate, confusing the interpretation of the photograph. This scattered light was largely eliminated by coating the HMX crystals with a black dye (Flo-master Transparent Black ink). Figure 3 is a photograph, of the blackened coarse HMX crystals with the same exposure as that of Fig. 2. There is little, if any, scattered light remaining, and the source of the light is a region with a thickness comparable to a few crystal dimensions. The experiment was repeated with the fine cut of HMX, also blackened. Exposure was the same as for Figs. 2 and 3. The result is shown in Fig. 4. The light is as bright as before, but the region emitting the light has a thickness proportional to the size of crystals and hence is narrower than that seen with the coarge cut.

Microscopic examination of the film negatives of the shots shown in Figs. 3 and 4 shows detail not visible in the print. Of particular interest are a number of small, very dense spots embedded in the image, indicating regions with a high radiation temperature surrounded by regions with a lower temperature.

An experiment was done to obtain an estimate of the radiation temperatures in these bright bands of light. Photographs were made of the two samples shown in Figs. 3 and 4; of a sample of coarse blackened NaCl crystals shocked by high-density HMX; of the sun and of argon shocked by Composition B. The photographs were taken on a single piece of film, and processed to ensure uniform treatment of all areas of the film. The image of the sun was barely detectable and much too faint co print. The remaining images were read on a microdensitometer. It was found that the shocked argon produced the greatest density on the negative (and hence was the brightest object photo-



Fig. 1. Shot assembly for observations of detonations in pour density HMX and PETN. PBX 9501 is the designation for 95% HMX bonded with 5% plastic and pressed to high density.



Fig. 2. Photograph of Detonating, coarse HMX.



Fig. 3. Photograph of detonating, coarse HMX blackened to suppress scattered light.



Fig. 4. Photograph of detonating, finegrain HMX blackened to suppress scattered light.

graphed). Both samples of HMX and shocked NaCl produced images indicating general brightness levels below that of argon but much greater than that of the sun. In all three samples, moreover, there were a few small areas indicating brightness levels only a little less than that of shocked argon. The sun has a blackbody temperature of approximately 5762 K (8) and argon shocked by Composition B has a blackbody temperature of approximately 18 000 K (9). The gamma of the film, especially in the higher density range, was not measured and an accurate estimate of the radiation temperature could, therefore, not be made. We can, however, make rough estimates based on consideration of the calculated response of the image-intensifier tube to blackbody radiation of various temperatures be-tween 6,000 K and 18,000 K. On this basis, the hottest spots seen in detonating HMX as well as shocked NaCl have radiation temperatures as high as 12,000 K to 14,000 K, while the remaining areas have lower temperatures in the 9,000 K to 12,000 K region.

These radiation temperatures correspond to real temperatures in the radiating medium only if it is a blackbody (i.e., if it is optically thick with a blackbody spectral distribution). The optical thickness was not measured, but a crude spectrum of the band of bright light was made. For our spectrograph we used the setup shown in Fig. 5. The shot assembly was loaded with fine (-70 sieve) blackened PETN and oriented so that the thin band of detonation light could act us its own slit. Because the exposure time was 10 ns, the image movement was less than 0.05 mm at the camera and produced no discernible blur. Lens No. 1 imageó the detonation on a combination clear glass plate and interference filter with band pass of ~10 nm, centered on the 546.1-nu line of Hg. This image in turn was reimaged on the I^2C by lans No. 2 after passing through the trans-mission grating. The grating used has 200 grooves/am, is blazed for the first order, and gives a spectral resolution some 10 times greater than the resolution of the camera (15 lp/mm). Figure 6 shows the spectrum of detonating PETN. The various horizontal streaks correspond to especially bright spots along the strip of detonation light. The small patch of light above the main spectrum near the center of the picture is the light transmitted by the interference filter at 546.1 nm. The spectrum extends from approximately 650 nm to 416 nm. It is continuous, and is much orighter in the blue than in the

red, even after allowing for photocathode response in the two regions. This observation supports the estimates of high temperatues.

In order to relate the position of the brilliant detonation light to the position of the traditional "detonation front" or pressure jump, some experiments were done with the shot setup shown in Fig. 7. A small argon flash illuminated the front of the e-



Fig. 5. Experimental setup to obtain a spectrum of detonation light.



Fig. 6. Spectrum of detonating PETN. Small blob above the spectrum is light transmitted by an interference filter with 10 nm band width centered on 546.1 nm. The blue end of the spectrum is to the right.

vacuated shot assembly filled with coarse HMX. Since the detonation light was almost as bright as the argon flash itself, it was attenuated to avoid severe overexposure of the photograph of the shot assembly. A Kodak gelatin neutral density 2 (ND-2) filter was found to be suitable. It was trimmed and oriented to just cover the image of the HMX. The photograph obtained is shown in Fig. 8. Shock wings can be seen in the polymethyl methacrylate (PMMA), on either side of the band of detonation light, making an angle of \sim 53° with the axis of detonation, and nriginating at the rear of the band of 1 ght. They show clearly that the detc.ation light in this explosive system is produced ahead of the pressure jump, as surmised by Seely (4).

A variation of this experiment was made by removing the ND-2 filter from the first image plane and placing it against the inside surface of the PMMA window, in contact with the HMX column. The result is shown in Fig. 9. Again, there is a band of light ahead of the pressure jump, as marked by the origin of the shock wings or either side of the detonating column. In this photograph there is a pattern of hot material behind the shock front, not seen in the previous photograph, which appears on the negative to be brighter than the band of detonation light. In other experiments, similar structure has been observed in the material behind the shock front when the band of detonation light was heavily overexposed. The radiation temperature of these glowing spots is estimated to be 2,500 X to 3,500 K. The passage of the shock front evidently destroyed the gelatin film ND-2 filter, thereby allowing 100 times as much light transmission as before, making it possible to record the brilliant detonation light and the much less brilliant light behind the shock front in a single exposure. The cause of the pattern of relatively cool spots is not known.

The observation of material preceding the shock front in the detonation, although not unexpected, stimulated experiments to characterize it. In the first experiment, a quartz gauge was placed at the end of a detonating column of blackened, evacuated, coarse HMX and the detonation was allowed to run into it as shown in Fig. 10. At the same time, three photographs were taken at 150-ns intervals. The photographs are shown in Fig. 11 and the gauge records are shown in Fig. 12.



Fig. 7. Shot setup for the simultaneous observation of detonation light in HMX and shock location in PMMA window. The ND-2 filter was cut to just cover the image of the HMX.



Fig. 8. Detonation light in coarse HMX photographed using the setup shown in Fig. 7. Note the shock wings in the PMMA window originating just behind the detonation light. The vertical lines on either side of the HMX column are scribed on the aluminum.

The pulse on the far left of the upper beam is the signal from an ionization pin placed 8 mm below the gauge. A pulse 1.6 µs later can be seen near the rise of the signal from the quartz gauge, corresponding to the first photograph in Fig. 11. The lower beam was delayed 1.0 µs with respect to the upper beam, wrote five times as



Fig. 9. Detonation light in coarse HMX photographed using the setup shown in Fig. 7 except that the ND-2 filter was placed in the shot assembly at the HMX-PMMA interface. Note the shock wings in the PMMA. The bright patches of light behind the shock are made visible by the destruction of the gelatin ND-2 filter by the shock. They are actually ~1/100 as brilliant as the brightest spots in the detonation light.

fast and had five times the voltage sensitivity. In the first photograph in Fig. 11, it appears that the band of detonation light has just contacted the gauge. From the gauge record, however, it is evident that the gauge begins to sense pressure approximately 0.3 μ s earlier, or more than half the width of the band of light. We surmise this is caused by high-velocity material, too tenuous to produce detectable light, but of very high velocity. Analysis of the gauge record is shown in Fig. 13. The stress on the gauge builds slowly for some 0.3 μ s, then more rapidly, reaches 2 GPa and runs off scale before the shock arrives. This observation must modify the traditional picture of a detonation front in porous explosives.

The derivation of an estimate of the areal mass accumulation from the gauge data follows a version of the "thick-plate technique" described by Asay (10) for investigating shockproduced ejecta from metal surfaces.



Fig. 10. Shot assembly for simultaneous photography of detonation in coarse, blackened evacuated HMX and measurement of stress on a guartz gauge.



Fig. 11. Three photographs of detonation light striking a quartz gauge. The first is on the left. The detonation is moving towards the right, and the band of light is just contacting the quartz gauge. The time interval between the photographs is 0.15 µs.



Fig. 12. Oscilloscope record of stress on a quartz gauge impacted by material causing detonation light in coarse HMX. Upper beam writing speed was at 0.5 µs/cm, sensitivity 1V/cm, 0.1 µs markers. Lower beam was delayed 1 µs from upper beam. Writing speed was 0.1 µs/cm, sensitivity 0.2 V/cm, 0.1 µs markers.



---- Areal Mass Accumulation x 10⁻³ (mg/mm²) ----- Areal Mass Accumulation x 10⁻² (mg/mm²) ----- Btress (GPa)

🖤 Phote 🖊 I 🛛 🔺 Phote 🕂 2

Fig. 13. Analysis of gauge record shown in Fig. 12.

In this method, the ejecta fly across a gap, impinge on a thick witness plate, and the stress history at the ejecta plate interface is determined. It is assumed that (a) the material is e-jected instantaneously from a free surface, (b) the lateral distribution of mass is uniform over the extent of the witness plate, and (c) the collision of ejecta with the witness plate is ine-lastic. Simple momentum conservation yields a relation between areal mass accumulation rate, dm/dt, and the stress at the face of the witness plate, σ , as

:

$$dm/dt = \frac{\sigma(t)}{V-tt}$$
(1)

where U(t) is the velocity of the interface and V(t) is the velocity of an increment of ejecta, dm, related to the gap thickness, d, and time, t, by:

$$v = \frac{d + b}{t} \frac{y \, dt}{z} \qquad (2)$$

In our experiment U is neglected because U << V. Furthermore, d, and hence V, are indeterminate. Also, assumption (b) is certainly not valid because the active gauge surface is 4 mm in diameter and the photographs of detonation light in the coarse HMX show considerable irregularities on a scale 1/10 of this dimension. If, however, we take V to be 5.3 mm/ μ s (the approximate detonation velocity of the explosive), values for dm/dt can be found from o(t), and graphically integrated to give an estimate of the areal mass accumulation as a function of time (also shown in Fig. 13). An estimate of the density of the material striking the gauge can be made by dividing the rate of mass accumulation, dm/dt, by the velocity, or

$$\rho(t) = \frac{dm/dt}{V} = \frac{\sigma}{\sqrt{2}} \qquad (3)$$

giving ~2 μ g/mm³ at 1.6 μ s and $\sqrt{70}$ μ g/mm³ at 1.75 μ s. These estimates are too high because the assumed velocity is only the detonation velocity and the eject necessarily move faster than this, but the estimates do show that the density of material in the ejecta stream is low (air at STP is 1.29 μ g/mm³).

A second experiment was done observing the ejects from a dense explosive traversing a 20-mm gap, a geometry more in keeping with the assumption stated above. Figure 14 shows a sketch of the shot assembly, and Fig. 15 shows the static photograph and two dynamic photographs. The scale in the static photograph, although distorted by the thick wall PMMA vacuum tube, gives a rough measure of the distance between the explosive free surface and the gauge (20.0 mm). The first dynamic photograph was taken just as the leading edge of the band of light on the wedge, made by the transparent ejecta, was even with the gauge surface. (Note that the black line on the rear surface of the PMMA tube remains visible until the usual detonation products, moving at ~ 8 mm/µs, cover it.) The last photograph was taken just before the cloud of opaque detonation products struck the gauge.

The gauge record analysis is shown in Fig. 16. It shows that some material has a velocity greater than 20 mm/µs, although the light detectable in the first photograph of Fig. 15 appears to come from slower material moving at ~20 mm/µs. From Eq. (3) we can estimate the density of the ejecta gas as a function of velocity. It varies from ~0.03 µg/mm³ for the material striking the gauge in the first photograph to ~0.9 µg/mm³ for corresponding material in the third photograph. If we assume that the highest-velocity material (>15 mm/µs) is hydrogen, corresponding to an areal mass accumulation of ~0.65 µg/mm², it is equivalent to all the hydrogen in a layer of explosive approximately 10-µm thick.



Fig. 14. Sketch of shot assembly for simultaneous photography of detonation ejecta from a PBX 9501 pellet scriking a PMMA wedge and quartz gauge measurements of the momentum of ct/. material.





Fig. 15. One static and two dynamic photographs showing ejecta from a PBX 9501 pellet striking a quartz gauge and a PMMA wedge.



- ---- Areal Mass Accumulation x 10-2 (mg/mm²)

Arrows Indicate Times of Photographs

Fig. 16. Gauge record analysis of ejects from a pellet of (BX 9501 detonated by a plane-wave lens. The arrows indicate the times of the photographs shown in Fig. 15. To characterize further the material producing detonation light, an experiment wis done to observe the spectrum produced by the detonation of a single crystal of PETN in a vacuum, in which the ejected material was allowed to strike a polished PMMA wedge. A crystal of very high quality, free of inclusions and measuring 9.5 x 37 x 21 mm, was glued to the face of a booster pellet of plastic-bonded HMX in a shot assembly sketched in Fig. 17.

The PMMA wedge was imaged on a vertical slit so that the end of the wedge face nearest the crystal was at one end of the slit and the end farthest from the crystal was at the other. The spectrum thus was resolved in time as well as wave length. The slit was imaged through the grating onto the photocathode of the camera.

The spectrum obtained is shown in Fig. 18. The H_{α} line is prominent and, in agreement with Hay, Peters, and Watson (6), is the first light seen. At first narrow, it broadens as pressure increases, finally merging into a continuum with a few lines superimposed. A very broad H_{β} line is faintly discernible and three other distinct lines can be seen. One of these lines appears to be the oxygen triplet at 615.7 nm. The other two lines at 493 nm and 455 nm have not been identified.

The properties of the ejected material, (i.e., high velocity, high stagnation temperature but low density) suggest that it might have some effect on the propagation of a detonation by preigniting the explosive grains ahead of the shock front. Clearly it cannot be necessary for the initiation (and therefore the continued propagation) of a detonation, because shock alone is known to be sufficient. In an attempt to detect this effect, a series of shots was fired in which a column of evacuated, blackened, coarse HMX, in-terrupted by a brass shim, was deto-nated at one end and the passage of the detonation front through the shim was observed. Three cameras were used and each camera was exposed three times. Figure 19 shows the results with a 0.42-mm-thick shim. At 0.67 µs the detonation light is being emitted from the bottom of the brass shim. By 1.07 us the light is gone and begins to reappear faintly at 1.47 µs, recovering essentially full brilliance by the last exposure $(2.27 \ \mu s)$. A number of such shots using brass shims of various thicknesses from 0.80 mm to 0.25 mm show the same behavior except that the thicker the shim, the longer it takes



Fig. 17. Sketch of shot assembly for observation of the spectrum produced by the ejecta from a detonating crystal of PETN striking a polished PMMA wedge.

PETN Spectrum



667.8 nm

. .

501.5 nm

Ha Spectrum

Fig. 18. Spectrum of detonation ejecta from the free surface of a single crystal of PETN stagnating against a PMMA wedge. The He spectrum was taken in a separate exposure with the spectrograph dimensions unchanged from those used for the crystal spectrum.



Fig. 19. Nine exposures showing the progress of a detonation wave in coarse HMX at bulk density through a brass shim, 0.42-mm thick. Times are from closure of an ionization pin.

C. 2.27

the detonation light to reappear. From these observations we cannot say that the band of detonation light has no effect on propagation, but any such effect is too small to be apparent.

III. SUMMARY

The very bright detonation light observed in porous explosives is caused, at least in part, by material ejected at high velocity from the reacting grains of explosive in the zone behind the shock colliding or stagnating against unreacted grains ahead of the shock. Radiation temperatures estimated to be as high as 14,000 K in small spots result from this stagnation. Such material is also ejected from a very smooth face of a crystal of PETN, and hence the mechanism of jetting by interaction of detonating grains, while not excluded as a contributing source of material, is not necessary to explain the detonation light. The stagnation light spectrum is a continuum, with H_{α} and other lines visible on the continuum when time and spectral resolution are sufficient. The ejected material is transparent, in contrast to the usual detonation products, and is very tenuous, having a density roughly comparable to air at STP. It can, however, produce stresses in the 1 to 2 GPa range by stagnating against a solid.

It is reasonable to surmise that such fast moving material, tenuous though it is, could ignite the surface of explosive grains and that these burning grains could burn fast enough in the region behind the pressure jump to support the detonation. A test of this surmise, obtained by interposing brass diaphragms in the path of detonation, indicates that detonation proceeds with only slight retardation even though the detonation light is extinguished for 0.4 μ s or more. While not ruling out the "hot spots" produced by stagnating ejecta as contributing factors, the test seems to call for other mechanisms to cause the continual initiation of the explosive and sustain the detonation.

ACKNOWLEDGMENTS

I greatly appreciate the many helpful suggestions concerning this work made by R. L. Rabie and W. C. Davis. Special thanks are owed to J. D. Wackerle for suggesting the gauge experiments and for helping with the analysis and interpretation. Thanks are also owed to H. H. Cady for growing the high-quality PETN crystals used in this work. The expert technical assistance of D. P. Childress in the performance of the experiments deserves the highest praise.

This work was performed under the auspices of the U.S. Department of Energy.

REFERENCES

- H. Muraour, Chimie et Industrie, Vol 47, p. 3 (1942).
- S. Paterson, "Source of the Light Recorded in Photographs of Detonating Explosives," Nature, Vol. 167, pp. 479-481, March 24, 1951.
- L. B. Seely and J. H. Blackburn, "Source of the Light Recorded in Photographs of Shocked Granular Pressings," Nature, Vol. 194, pp. 370-371, April 28, 1962.

- L. B. Seely, "A Proposed Mechanism for Shock Initiation of Low-density Granular Explosives," Proceedings of the Electric Initiation Symposium, The Franklin Institute, EIS-A2357, 27-1 to 27-50, October, 1963.
- 5. N. Lundborg, "Front and Mass Velocity at Detonation in Evacuated Chambers," Arkiv För Fysik, Band 25 nr 39, pp. 541-543, January 24, 1964.
- J. E. Hay, W. C. Peters, and R. W. Watson, "Observations of Detonation in a High Vacuum," Fifth Symposium (International) on Detonation, pp. 559-565, August 1970.
- O. G. Winslow, W. C. Davis, and W. C. Chiles, "Multiple-Exposure Image-Intensifier Camera," Sixth Symposium (International) on Detonation, pp 664-667, August 1976.
- J. A. Duffie and W. A. Beckman, Solar Energy Thermal Processes, p. 3, John Wiley and Sons, Inc., 1974.
- W. C. Davis, Los Alamos National Laboratory, private communication, July 1979.
- J. R. Asay, "Thick-plate Technique for Measuring Ejecta from Shocked Surfaces," J. Appl. Phys., 49 (12), pp. 6173-6175, December 1978.