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TITLE DECOMPOSITION OF ENERGETIC MATERIALS ON THE DROP-WEIGHT-IMPACT MACHINE

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DECOMPOSITION OF ENERGETIC MATERIALS ON THE DROP-WEIGHT-IMPACT MACHINE

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The drop-weight impact test is heavily relied upon for initial characterization of limited amounts of new energetic materials. The mechanisms for reaction of energetic materials on the drop-weight machine are largely unknown. Partly as a consequence of this lack of understanding, results obtained on the drop-weight machine are often misleading and inconsistent. We are investigating decomposition of explosives on the drop-weight machine using radiometric and spectroscopic methods. Initial radiometric results reveal consistent, sequential emissions for specific impacted explosives that may correspond to time-resolved chemical reactions. A method to quantify the extent of an impact-induced reaction using radiography is discussed. Limited results have also been obtained using time-resolved spectroscopy.

INTRODUCTION

The drop-weight impact test is one of the cornerstones of explosive sensitivity testing. Despite widespread usage, however, numerous problems detract from the reliability of the test. These problems have inspired work at this Laboratory directed towards better understanding the principles of the drop weight impact test as well as the decomposition chemistry of impacted explosives. Work in this vein is being conducted by radiometric and spectroscopic examination of impacted energetic materials.

The drop weight impact test is one of the methods most relied upon for initial characterization of high explosives (HE) sensitivity. This does not bespeak of superior results provided by the impact test, but rather a lack of better results obtained by other methods with limited amounts of materials and a limited investment of time and effort. Other preliminary sensitivity tests include the minimum priming, friction, spark, and small and large scale gap tests.¹ Subsequent large-scale tests include the Susan, splat, skid, bullet, high speed machining, and heavily confined heating tests.² While all of these tests provide valuable information regarding explosive sensitivity, all require far greater amounts of material and effort than does the impact test. Because of the pivotal role the drop weight impact test plays in explosive sensitivity testing, one might expect the test to be well characterized and defined. Unfortunately, this is not the case. For a given set of explosives, the ranking of sensitivity predicted by the impact test is often different from that predicted by other tests.¹ Indeed, even

correlation between different drop weight impact machines seems to be the exception rather than the rule. For example, Figure 1 lists H₅₀ values (that height from which a falling weight causes explosive reaction 50% of the time) for some common explosives tested at Lawrence Livermore National Laboratory (LLNL) and Los Alamos National Laboratory (LANL). The use of a 5 kg weight at LLNL compared to a 2.5 kg weight used at LANL should dictate drop height ratios of approximately 1:2. Obviously, this is not the case. The British impact sensitivity test, or Rotter Test, gives yet a different set of results.³ Problems of reproducibility are further aggravated by a lack of protocol for sample preparation at different laboratories. Particle size, percent voids, sample handling, etc. are all variables that profoundly affect explosive sensitivity yet are not addressed when reporting H₅₀ values. This degree of uncertainty in a test as crucial as the drop weight impact test is disturbing. Thus, efforts have been made in this Laboratory and others^{4,5,6,7} to better understand the chemistry occurring in the impact machine. While some progress has been made, the problem is far from resolved.

CURRENT RESEARCH

Efforts at Los Alamos have centered on utilizing light emission from impacted energetic materials in an attempt to better understand the decomposition chemistry occurring on the drop weight impact machine. These efforts can be divided into two parts: time-resolved radiometry and time-resolved emission spectroscopy.

	LLNL (5.0 kg)	LANL (2.5 kg)	Ratio
FEFO	28	60	2.1
TNT	80	148	1.9
PETN	11	14	1.3
Baxtol	95	110	1.2
PBX 9501	44	48	1.1
RDX	28	28	1.0
HMX	33	32	0.97
Octol	41	38	0.93
DIPAM	95	85	0.89

FIGURE 1. COMPARISON OF H_{50} VALUES (CM) PUBLISHED AT LANL AND LLNL.

RADIOMETRY

Equipment. In the course of radiometric work conducted with the drop weight impact machine, the relationship between drop height and temporally resolved light emission was examined. The experimental apparatus for this study (Figure 2) consists of a modified drop weight machine with a specially designed anvil (Figure 3) to allow light transmission via a fiber optic bundle to an RCA 8850 photomultiplier tube (PMT). The PMT signal is amplified by an Ortec 9401 fast pre-amplifier and an Ortec 9102 amplifier/discriminator. The amplified analog signal is stored and processed on a Nicolet 4562 oscilloscope. Signal plotting is accomplished with an Apple Macintosh computer, while integration is done with a Hewlett Packard 4190A recorder/integrator.

In a typical experiment, 30 mg samples of explosive powder are placed on the sapphire window of the anvil. The weight is dropped from varying heights and the resulting light signal recorded on the oscilloscope. The duration of light emission is typically between 70 and 90 microseconds. Representative signal traces are shown in Figure 4.

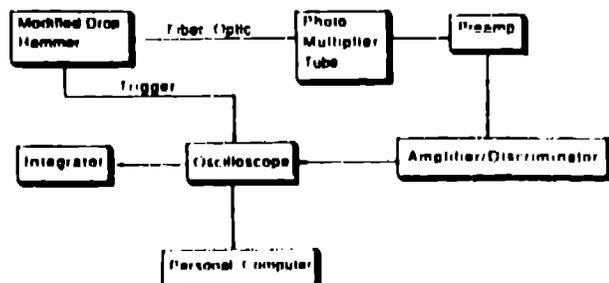


FIGURE 2. EXPERIMENTAL APPARATUS FOR RADIOMETRY.

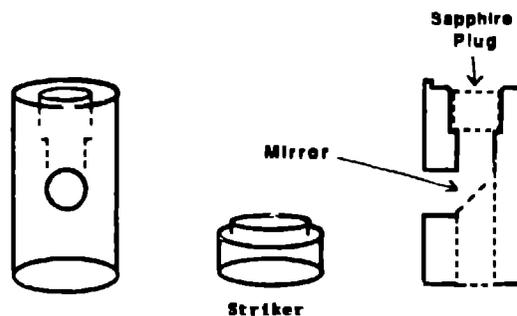


FIGURE 3. MODIFIED EXPERIMENTAL ANVIL.

Results. In the case of full "goes", a consistently shaped light trace was obtained for each explosive examined to date. The fact that light emissions from different explosives show characteristic and consistent features implies that unique, sequential chemical reactions are occurring and are being resolved temporally. Using filters, this light can be broken down into ultraviolet (UV), visible (VIS) and infrared (IR) components. UV light occurs early and is of short duration, while visible and IR components are present throughout the emission. Occasionally, duplicate, yet virtually identical traces are obtained, indicating perhaps that two separate hot spots are forming displaced in time and propagating independently. The consistency in emission characteristics falls when samples are only partially consumed and essentially no light is generated by "no goes".

Early radiometry results indicated the quantity of light produced by an impacted explosive varied as a function of impact height. Thus, total emission seems to be an intrinsic indicator of the extent of chemical decomposition. A set of explosives extensively used at Los Alamos was subjected to impact from a 4 kg weight on a modified drop weight impact machine and the total integrated light emission recorded. A plot of averaged total light emission for PETN, in arbitrary units, versus drop height is shown in Figure 5. At least five drops were made at each height. "No goes" were obtained along with "goes" at the lowest drop heights. In these cases, a value of zero light emission was averaged into the emission obtained from "goes". Apparently, drop heights above some threshold value provide sufficient energy for propagation of the decomposition reaction and are characterized by a mass of less consistent amount of emission. At lower drop heights, a break occurs where light produced by the explosive begins to drop off rapidly with decreasing drop height. This is probably indicative of insufficient impact energy to guarantee propagation of the decomposition

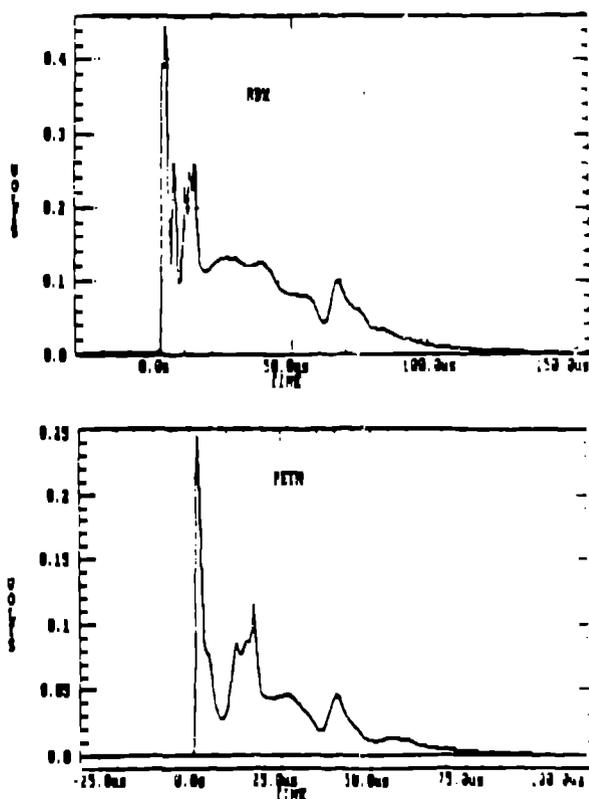


FIGURE 4. TIME RESOLVED EMISSION FROM IMPACTED EXPLOSIVES.

reaction to the point of sample consumption. Eventually a point is reached where no sample reaction occurs and no light signal is obtained within the number of experimental trials performed. Statistically, if the number of drops from very low heights were increased, a reaction with accompanying light would eventually be obtained, but experimental practicality limits the number of drops that are reasonable.

These results are consistent with high speed photography work done by J. E. Field where it is possible to see hot spots form and die out in impacted secondary explosives.⁴ At low drop heights, hot spots quickly die out, thus decreasing the amount of light signal detected.

Conclusion. We believe that light emission from energetic materials might offer an alternative method for quantifying sample decomposition on the drop weight machine. While this method is probably not practically suited to replace the electronic ear now used, it does offer supplemental information regarding explosive behavior on the drop weight machine and does not suffer from some of the limitations imposed by the current method. For example, this method could be far

less sensitive to operator and equipment-related inconsistencies associated with the electronic ear. In the radiometric method, long-term equipment fluctuations such as a loss of sensitivity in the PMT, would be characterized by shifts of the integrated light curve up and down without affecting the point where the curve breaks. Long-term fluctuations in the sensitivity of the electronic ear currently in use can allow misleading H_{50} values to be published. Furthermore, impact characterization of propel lants, which occasionally do not "pop" on the impact machine because of their slow rate of reaction, might be improved. Before conclusions can be drawn regarding the practicality of this method of characterization; however, more work must be done to assure test predictions are in agreement with historical knowledge regarding sensitivities of common energetic materials.

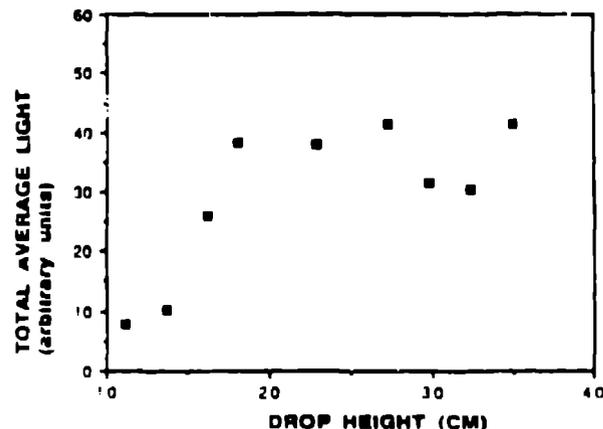


FIGURE 5. PLOT OF DROP HEIGHT VS. AVERAGE OF TOTAL INTEGRATED LIGHT FROM IMPACTED PETN.

SPECTROSCOPY

In addition to examining radiometry from impacted explosives, attempts were made to identify the molecular species responsible for emission. Previous work in this area involved examination of IR emission, but few questions were resolved.⁹ The mechanism whereby an impacted explosive goes from reactant to products is largely unknown. The light emitted during that process offers one of the few practical means of exploring this chemical discontinuity. Radiometry work has suggested that emissions from sequential chemical reactions occurring during explosive impact can be temporally resolved. This suggests that emission spectra obtained at different times after impact might permit identification of unique chemical species associated with different stages of decomposition. If time resolved

chemical emissions can be identified, some mechanistic insight regarding impact induced decomposition might be obtained.

Equipment. The experimental apparatus used in this study is described in Figure 6. A biturcated, fused-silica fiber-optic bundle is used to connect the drop-hammer apparatus with a 0.25 m Jarrel Ash spectrometer and a PMT. The PMT amplifier/discriminator section, already described, is used to provide a trigger for a Tracor Northern TN6500 optical multichannel analyzer (OMA) and TN6143 vidicon-array detector. By utilizing a TN6130-1 pulse amplifier/driver, it is possible to gate the detector on and off to obtain spectra down to 150 nanoseconds pulse width. By varying the delay between first emission and fast spectrum acquisition, it is possible to obtain time-resolved emission spectra.

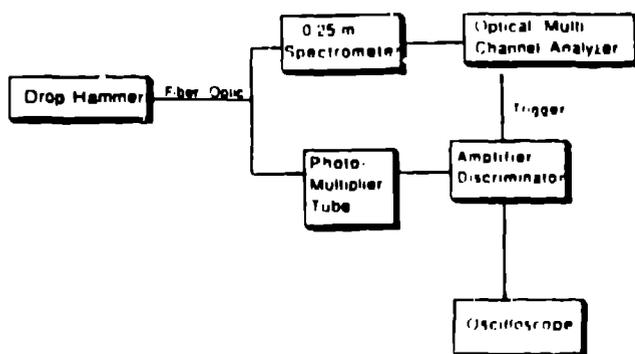


FIGURE 6. EXPERIMENTAL APPARATUS FOR SPECTROSCOPY.

A large variety of explosives was examined on the drop hammer, including RMX, RDX, CAPB, NTO, TNT, tetryl, HNS, 4 nitro-1 picryl-1,2,3 triazole, sulfur nitride, and PETN. Less sensitive explosives that would not react on the experimental drop hammer under normal conditions were initiated using a slapper accessory in conjunction with the modified anvil already described. This slapper accessory consists of a hardened steel piece that attaches to the anvil and locates a 2 mm hole directly over the sapphire window. A flyer from a Kapton sheet placed over the hole is driven into the explosive under investigation by the primary explosive sulfur nitride that is resting on top of the Kapton and initiated by impact. Barrel length is approximately 5 mm.

Results. Attempts to obtain emission spectra from impacted explosives have met with limited success. The light obtained from all the explosives examined is largely black body in nature with a few areas of exception.

Most of the explosives investigated exhibit a strong emission in the vicinity of 5890Å

(Figure 7). Sodium emission has been identified frequently by other workers examining light from detonating explosives, and consequently, this feature was initially attributed to the strong sodium lines occurring at 5890Å and 5896Å.⁸ Further experimentation, however, indicated sodium an unlikely source for this emission feature.

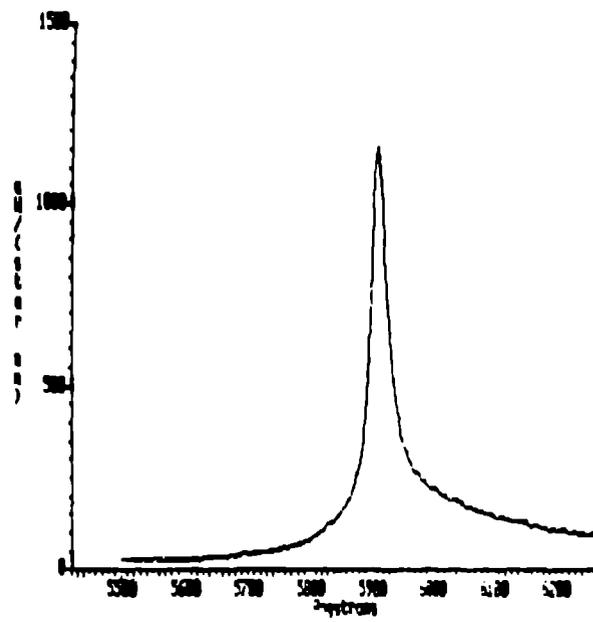


FIGURE 7. EMISSION FROM IMPACTED PETN.

Spectra obtained from a sodium source show a sharp, unresolved doublet with a maximum consistently at 5890 Å. The emission from impacted explosive samples maximized between 5896Å and 5920Å although 5899Å was typical. The shape and intensity of this peak proved to be a function of time after impact. For example, impacted PETN samples first exhibit the 5899Å band 40 microseconds after first light, with maximum intensity occurring after 70 microseconds. All the samples exhibiting a 5899Å emission did so at late time relative to first light. When the samples were slapper initiated, however, prompt initiation eradicated time dependence of the 5899Å emission.

Only carbon-containing explosives show emission at 5899Å, although not every carbon-containing explosive emits in this region (for example, 4 nitro-1 picryl-1,2,3 triazole). The non-carbon-containing explosive, sulfur nitride, does not emit in this region even when loaded with sodium salt. Furthermore, in late work of strong oxidizers such as $KMnO_4$ or CrO_3 (Aldrich) drastically reduce or eliminate the

peak in question. NTO emits strongly at 5899Å, yet the sodium and potassium salts of NTO show no emission above black body in this area.

Thus the source of emission in the 5899Å region in impacted explosives is probably not sodium. Positive identification of the emission source is impossible. The peak position does correspond closely with a high pressure C_2^* (6,8) emission attributed to carbon radical recombination, e.g., $[C(^3P) + C(^3P) \rightarrow C_2^*(d^3\pi_g)]$. If the emission source were C_2^* , this would help explain the effect of strong oxidizers on the peak intensity. Other investigators have identified C_2^* emission from detonating explosives, but have identified numerous peaks in a band system rather than a single emission.^{9,10} Why, in this instance, only the C_2^* (6,8) emission would occur is not known, but others have observed this phenomenon.¹¹

No other molecular emissions were consistently identified. Nitramines occasionally showed a strong OH band at 3064Å, but no pattern for appearance was discerned. TNT exhibited a shoulder at 4315Å above black body radiation. A strong OH system begins at 4315Å, but no other peaks in the system were observed, thus definitive assignment is impossible.

Emission for all the explosives examined consists of UV, VIS and IR components. UV light is of short duration, occurring early, while VIS and IR components persist throughout the emission. In PETN, for example, UV light is detectable for the first 40 microseconds of emission, while VIS and IR light linger the entire 90 microseconds of emission.

Limited results using powdered and pelletized explosive samples prompted usage of very thin, optically transparent explosive samples. Unfortunately, this did not alter the experimental results in any positive way. It was concluded, therefore, that definitive identification of discrete molecular emissions from impacted explosives was not practically feasible.

CONCLUSION

A thorough understanding of the chemistry occurring on the drop weight impact machine remains elusive. Results from this work imply reproducible sequential chemical reactions unique to a given explosive are probably initiated by impact. Furthermore, the amount of light emitted by an impacted explosive may be used as an intrinsic indicator for extent of impact induced reaction. Identification of specific molecular species along the reaction pathway for impact induced explosive decomposition has not proven possible by this method. Further mechanistic work in this area is certainly

warranted. At this Laboratory, research examining emission from explosives subjected to laser induced decomposition is underway in conjunction with other projects.

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