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PROGRESS REPORT

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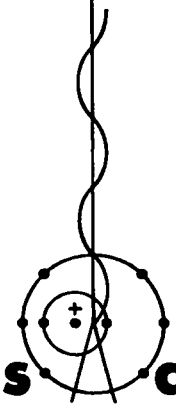
Joint Services Explosive Program

Defense Advanced Research Projects Agency

DARPA Order No. 2502



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UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

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LA-5402-PR
Progress Report
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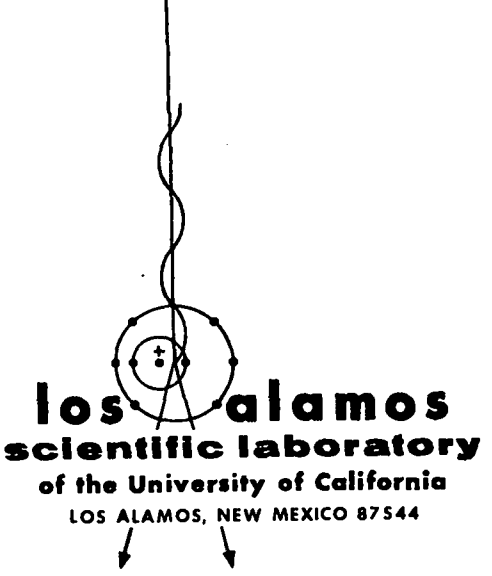

Joint Services Explosive Program

Quarterly Progress Report

June 15 through August 15, 1973

Compiled by

A. Popolato

This work was performed with funds provided by the Defense Advanced Research Projects Agency under DARPA Order No. 2502.

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JOINT SERVICES EXPLOSIVE PROGRAM

QUARTERLY PROGRESS REPORT

FOR THE PERIOD JUNE 15 THROUGH AUGUST 15, 1973

compiled by

A. Popolato

PREFACE

This is the first of a series of progress reports describing the status of four tasks undertaken by the Los Alamos Scientific Laboratory (LASL) for the Defense Advanced Research Projects Agency. The intent of LASL is to issue these reports on a quarterly basis. Since the tasks undertaken by LASL form an integral part of the Joint Services Explosive Program, copies of this report are being distributed to the agencies of the services participating in the program. Separate final reports will be issued for each of the tasks at the termination of each task.

ABSTRACT

The results obtained, for the period from June 15 through August 15, 1973, on the Joint Services Explosive Program are presented. The tasks undertaken by the Los Alamos Scientific Laboratory include: (1) Physical and Processing Characteristics of Nonideal Explosives - Special Emphasis on Amatex, (2) Analysis of Prematures, (3) Synthesis of HMX, and (4) Initiation and Sensitivity.

I. SUMMARY REPORT

A. PHYSICAL AND PROCESSING CHARACTERISTICS OF NONIDEAL EXPLOSIVES - SPECIAL EMPHASIS ON AMATEX (TASK A)

1. Introduction

In the context of this work, nonideal explosives are materials in which oxidizers such as ammonium nitrate (AN) and energy contributing fuels such as TNT and RDX are separated into discrete phases. The emphasis of the Los Alamos Scientific Laboratory (LASL) investigation is being devoted to a systematic investigation of the processing properties of Amatex, mixtures of RDX, TNT, and AN. The work is being coordinated with work being performed by the Army at Picatinny Arsenal (PA) and by Lawrence Livermore Laboratory (LLL).

The initial phase of the LASL work is to develop a base line of engineering information that represents the minimum preparation and casting costs. For this phase, the Amatex formulations are based on the use of raw materials such as AN, and Composition B, as received and without further processing by the ammunition loading plant.

The characteristics of interest are the melt and casting properties; the initiation and performance

properties of the cast material: and finally, the handling and storage characteristics. These characteristics will be determined in subsequent studies as a function of AN source, AN particle size, AN content, and finally, with phase stabilized AN. Work performed by other organizations will not be duplicated unless we feel that some duplication is required as tie points.

2. Results to Date

A literature review is nearing completion. A series of Amatex melts has been prepared with high density fertilizer grade AN prills.

In a melt prepared under atmospheric conditions, the prills have a tendency to float in the melt. During the casting or solidification procedure, prills segregate to the top of the casting. In melts prepared under vacuum, the hollow prills are filled with TNT and the tendency to segregate is eliminated both in the melt and in the casting. As the quantity of RDX is increased at the expense of AN, with the TNT content held constant, there is little difference in the melt and casting properties.

3. Future Work

Work with the prills will continue in an effort to determine if, as a result of using prills, there is a significant degradation in energy, detonation failure dimensions, and initiation properties. After this phase, studies will be started with fine particle AN, in the region finer than is presently being investigated by PA.

B. ANALYSIS OF PREMATURES (TASK B)

1. Introduction

The long history of experience with Army and Navy large caliber guns indicates that prematures occur at a frequency varying between one in a hundred thousand to much less than one in a million. The frequency seems to be related to the setback pressures imparted to the projectile as it is accelerated through the gun tube. Although a wide variety of defects in the projectile have been suggested as possible causes, this study only considers the effects of defects in the explosive fill and the type of explosive used in the fill.

The objective of this study is to develop a number of analytical models in which thermal energy is generated and transported into the explosive. The generation of thermal energy will be determined as a function of the type of defects in the explosive, the type of explosive, and the force fields experienced by the explosive in the gun tube. The ultimate goal is to identify the critical parameters leading to initiation and to verify these parameters experimentally.

2. Progress to Date

To date, two one-dimensional models have been considered and developed. The first considers the time-dependent closure of an air filled void or gap between the base of the projectile and the high explosive fill. With this model, the initial source of thermal energy is the compressed gas. This energy is allowed to flow into the metal base and the high explosive. As the surface temperature of the explosive is increased, a second source of energy becomes available as the result of the exothermic decomposition of the explosive. An ignition occurs if the energy generated by the gas compression and the subsequent surface decomposition of the explosive exceeds the energy lost to the surroundings. For a given explosive with a given set of thermophysical properties, one can calculate a set of critical conditions relating the rate of closure or the rate of pressure increase to the initial gap size. A critical curve has been calculated for an RDX-containing explosive.

The second model considers the time-dependent closure of a spherical void located within the high explosive. With this model, the initial source of thermal energy is the plastic deformation occurring at the void-explosive interface. The plastically deformed region increases radially as the pressure in the explosive is increased. The second source of energy and the energy transport are identical with that of the gap closure model.

Expressions defining the regions of plastic deformation have been developed and have been coupled with heat conduction equations containing the HE decomposition energy generation term.

3. Future Work

The activity during the next quarter will be devoted to the design and development of experimental techniques that can be used to test the validity of the two models discussed. Since it is recognized that the models described may be an oversimplification of the early stages of ignition, it is important to start on a parallel experimental program.

C. SYNTHESIS OF HMX (TASK C)

1. Introduction

As the result of research conducted by PA and the University of Idaho, under contract to PA, a new synthetic procedure for the conversion of hexamethylenetetramine to pure HMX has been developed. This procedure has the potential of making HMX competitive in cost with RDX.

Laboratory work performed at PA indicated that two problems existed in the preparation of the intermediate. The first was the excessive liberation of energy (exotherm) occurring during the nitrolysis, and the second was the inability to separate this intermediate from the reacting media.

2. Results to Date

Results of preliminary work performed at LASL indicate that both problems can be solved by substituting AN for nitric acid in the nitrolysis. This change to AN has another important advantage in that the requirement for concentrated nitric acid is eliminated.

3. Future Work

During the next report period a parametric study of the preparation of the intermediate, diacetyl-pentamethylenetetramine (DAPT), used in the preparation of HMX will be conducted.

D. INITIATION AND SENSITIVITY (TASK D)

1. Introduction

The ultimate objective of this investigation is to obtain a quantitative understanding of the mechanisms leading to the initiation of a violent reaction in the high explosive. One approach to developing a quantitative model of the initiation is to study the response of an explosive subjected to a single shock of known amplitude and duration in a one-dimensional system. This requires the development of a plane wave shock generator and instrumentation capable of providing the data that can be used to define the state of the shocked explosive as a function of time and initial conditions.

2. Results to Date

Computer programs, developed at LASL, have been used to help design a plane wave, low amplitude, short time-duration shock generating system. Several prototype shock wave generators have been fabricated and tested. The desired time simultaneity and pressure amplitudes of the plane shock wave have not been achieved.

There is also an indication that the time duration of the pressure pulse is significantly shorter than predicted with either a hydrodynamic computer program (PAD) that ignores elastic-plastic effects or with a computer program (SIN) that takes these properties into account. Since it has been found that, for this problem, the effect of including the elastic-plastic properties of the metals is negligible, all design computations have been conducted with PAD.

3. Future Work

Work will continue on the design of plane wave generators and the instrumentation required to determine the amplitude and duration of the shock pulse.

II. PROGRESS REPORT

A. PHYSICAL AND PROCESSING CHARACTERISTICS OF NONIDEAL EXPLOSIVES - SPECIAL EMPHASIS ON AMATEX (TASK A), P. G. Salgado, H. R. Lewis, A. Popolato

1. Introduction

In the context of this work, nonideal explosives are materials in which oxidizers such as AN, and energy contributing fuels such as TNT and RDX are separated into discrete phases. Some of these materials are being used and others are being considered as alternate or emergency fills for high use munitions. The use of AN has the potential of substantially reducing the costs required to modernize, and maintain the plants used for the production of high explosives.[1]

The emphasis of the investigation to be performed by LASL is being placed on a systematic characterization of the processing properties of Amatex, mixtures of RDX, TNT, and AN. This work is part of the long-range Joint Services Explosive Fill Program with the overall objective of optimizing the utilization of explosives in munitions in terms of effectiveness, system design, cost, and material availability. To meet these objectives, one must have basic information. Since a significant effort is being applied to the study of specific formulations for use in artillery projectiles, bombs, and some varieties of specialized ammunition such as shaped charges, the work performed at LASL will be coordinated with the appropriate military services in an effort to avoid duplication. This work will also take advantage of, and be coordinated with, the work being performed by LLL and PA on the more fundamental properties of nonideal explosives relative to the chemical reactivity of particulate oxidizers and their contribution to the energy liberated in the detonation process, including the expansion of the detonation products.

An important aspect of the concept of an alternate or emergency fill is the ability to change from the preferred fill to the alternate fill with a minimum perturbation on the manufacturer of the raw materials and the ammunition loading plant. Other practical considerations are possible changes required in the fuse and booster system and the storage and handling procedures. Alternate fill formulations requiring significant changes in boosting, storage, and handling requirements should, if possible, be avoided. For these reasons, it seems appropriate to obtain a base of engineering information that can be used in a study of munition cost effectiveness.

The least costly process for preparing and loading Amatex formulations or any alternate fill should be one in which equipment existing at the loading plant can be used without modifications or additions. For Amatex, this implies that the AN be used as manufactured for use in the fertilizer industry. Any departure from this would represent added manufacturing cost. In order to determine if these additional costs are warranted, one must have a basis for comparison. For example, Finger[2] has shown that fine (< 5 micron diameter) particulate oxidizers such as perchlorates when mixed with contributing explosives participate in the detonation reaction in times that could affect the performance of fragmenting munitions. However, the problems associated with the preparation of fine particle AN, and the problems associated with casting these, add to the manufacturing costs.

If the added performance justifies the cost, fine particles of AN should be seriously considered. There are other examples of a balance between added performance or better storage properties and added cost. One in particular has to do with the problems relative to the solid-solid phase transitions in AN and the possible elimination of storage problems with the added cost of phase stabilized AN.

Our intent is to investigate the above factors systematically.

2. Results to Date

a. Literature Review

A review of the published literature dealing with AN and mixtures of AN with explosives has been started and is nearing completion. The literature dealing with Amatex formulations is limited. A paper by Cook[3] reviews work on amatols (mixtures of AN and TNT) and presents some experimental data on the detonation velocity of an Amatex consisting of 70.7 wt% Comp. B/29.3 wt% AN as a function of charge diameter. The AN used was fertilizer grade, ground to an average particle size of ~700 μ.

Cook's detonation velocity diameter data are shown in Fig. 1. Also shown in Fig. 1 are data on Comp. B obtained by LASL.[4] Note the difference in the effect of diameter on the detonation velocity.

The results of an evaluation of the performance characteristics of Amatex/40 in a standard 3.2-in. shaped charge have been described by Simon and DiPersio.[5] An evaluation of Amatex/20 in the 105-mm M-1 projectile is described by Forster.[6] Results in the 105-mm application look more interesting than those of the shaped charge.

Only one paper has been found describing the casting properties of these mixtures. Ziegler[4] recommended that more effort be placed on improving the casting properties of Amatex melts. Our limited experience indicates that he was right.

The problems related to handling and storing AN have been well described.[7,8,9,10] The polymorphism of AN has been partially characterized.[11,12,13,14] However, from the standpoint of the application of AN to military explosives, the Naval Ordnance Laboratory (NOL) work[15] is most applicable. They found that irreversible growth due to cycling could be avoided with a mixture of AN containing 10 wt% KNO_3. These results could have a major impact on the use of AN in military applications.

b. Experimental Program

The initial phase of the experimental program has just been started. This phase is devoted to the preparation of formulations with the AN available from the fertilizer industry. It is representative of the minimum cost system. The independent variables are the type of AN used, the composition, and the melt preparation procedure. The dependent variables are the melt viscosity, casting properties, relative energy, shock sensitivity, and storage properties.

A schematic of the test arrangement is shown in Table I.

TABLE I

MELT MATRIX

	Composition (wt%)			Type AN: High Density Prills, Type Melt		Type AN: High Density Grains[a], Type Melt	
	RDX	TNT	AN	Vac.	Open	Vac.	Open
Amatol	0	40	60		X		
Amatex	20	40	40	X	X	X	X
Amatex	40	40	20	X	X	X	X
Comp. B	60	40	0		X		

[a]Prepared from anhydrous melts.

These compositions represent an approximately constant oxygen balance for the products, assuming that the AN reacts to give one-half mole of oxygen for each mole of AN.

Melts have been made with commercial prills meeting the requirements specified in Mil-A-50460, August 12, 1970. The results are presented in Table II.

These results indicate that castable Amatex/40 can be prepared from prills and that vacuum melt techniques are required to prepare Amatex/20. The Amatol/60 prepared with prills is not castable with either open or vacuum melt techniques.

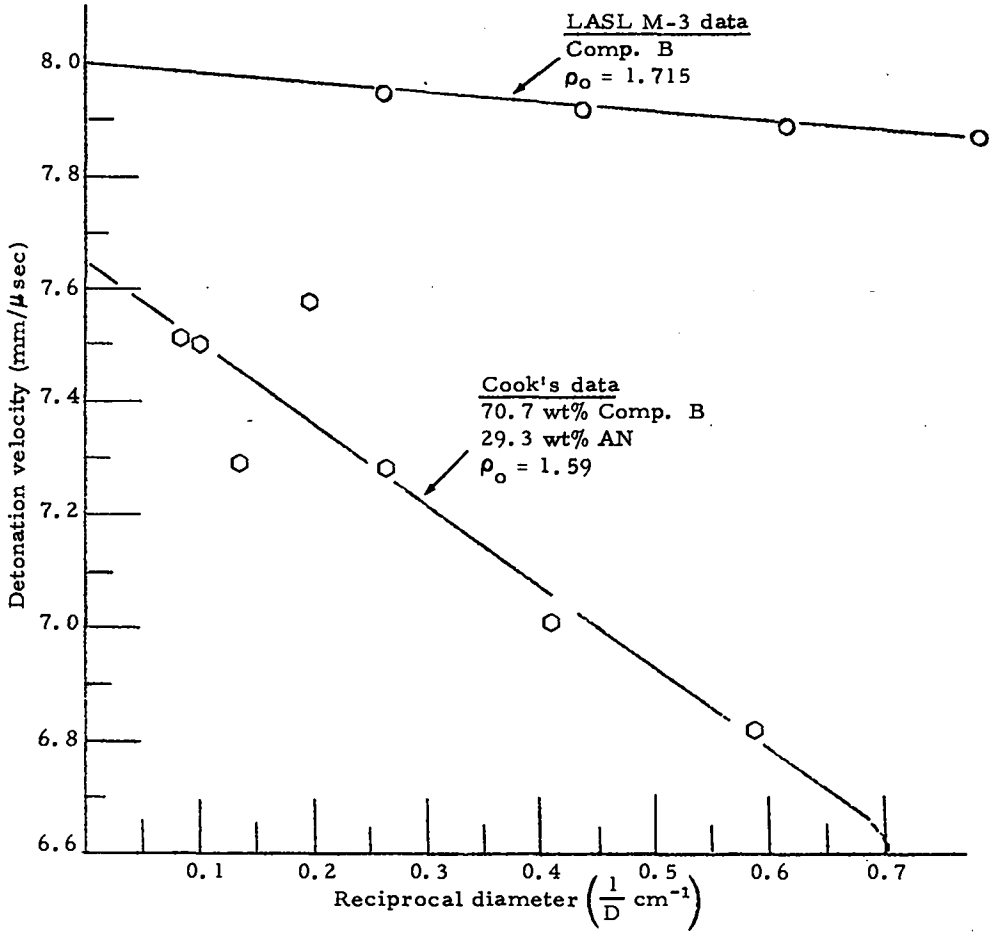


Fig. 1. Detonation velocity as a function of charge diameter.

TABLE II

FLOW AND CASTING PROPERTIES OF AMATEX AND AMATOL

Composition (wt%)					Melt Viscosity Efflux	
RDX	TNT	AN	Type AN	Type Melt	(sec)	Remarks
0	40	60	Prills	Open	a	Melt not pourable; AN would not disperse properly.
0	40	60	"	Vacuum	a	TNT forced into prill cavity and melt too viscous to pour.
20	40	40	"	Open	10 ± 5	Prills settled up or floated.
20	40	40	"	Vacuum	4 ± 1	Prills segregated down.
40	40	20	"	Open	3 ± 1	No differences observed between
40	40	20	"	Vacuum	3 ± 1	open and vacuum melt on castability.

[a]Efflux viscosity could not be determined.

3. Future Work

PA has prepared charges and evaluated the performance characteristics of Amatex/20 and Amatex/40 with three grinds of AN. LASL will prepare charges to test the energy and initiability of the Amatex/20 and /40 prepared with unground prills. These data should connect with the PA data to provide a curve of castability, energy, and initiability as a function of composition and AN particle size. It should start to answer the question relative to the contribution of energy as the particle size is reduced.

Attempts will be made to prepare melts with the AN particle size below the limits achieved by PA.

B. ANALYSIS OF PREMATURES (TASK B), C. A. Anderson

1. Introduction

The long history of experience of the Army and Navy with large-caliber guns indicates that when an explosively filled shell is accelerated during firing, there is a small probability that the fill will explode prematurely. Such firing mishaps are referred to as prematures, and they can occur either in the gun tube (in-bore) or after the projectile leaves the gun tube (out-of-bore). The results of an in-bore premature range from a slight swelling of the gun tube up to complete rupture with possible loss of life among the gun crew. Frequencies of prematures of guns in field service generally vary from one in a hundred thousand to much less than one in a million.[16] The frequency appears to be related to the setback pressures imparted to the projectile as it is accelerated through the gun tube.

Prematures are usually associated with defects in the fuse, the projectile case, or the explosive fill. Typical of the first two defects is a faulty fuse leading to premature initiation or a faulty base plate closure which leads to ignition of the explosive by the hot propellant gases. However, the defect in the explosive fill itself is to date the least understood - at least in comparison with fuse or base plate defects - and which is the subject of this investigation. The puzzling aspects of prematures caused by explosive defects have resulted in the comprehensive proof testing of gun/shell components, and the development of experimental premature simulators.[17,18,19]

The accidental initiation of high explosives has been the subject of considerable study for many years. Calculational models which rely on heating caused by shock compression of the explosive have been formulated and have been found applicable to a wide range of initiation phenomena.[20] In general, shock pressures in the order of tens to hundreds of kilobars are required to initiate detonation in solid explosives and the initiation times are on the order of microseconds. Even so, some investigators feel that a low amplitude shock (at the kilobar level) can be built up to an initiating level by the method of shock interactions.

Studies based on a thermal theory of ignition have indicated that not only can violent reactions be initiated by heating of an explosive,[21,22] but that the thermal mechanism may apply to initiation by shock or even relatively low-level stressing (1-5 kbar) of an explosive charge.[23] In fact, there is considerable experimental evidence that violent reactions can be achieved at relatively low stress levels in the explosive although the exact mechanism of initiation (i.e., conversion of a burn to detonation) is sometimes not well understood. Quantitative models based on the conversion of mechanical energy to thermal energy, with subsequent thermal ignition of the explosive depending on the reactive heating and thermal properties of the explosive, have been formulated at LASL[24,25] and elsewhere.[25] Such thermomechanical models have been used to confirm experimental observations on the initiation of explosions in specific configurations with well defined boundary conditions. Generally, ignition times on the order of a millisecond are observed.

2. Objective

The main purpose of this investigation will be to study situations of confinement and relatively low stressing of high explosives, typical of those levels provided during a projectile launching, as far as their potential for thermal ignition of the explosive is concerned. The ultimate goal will be to identify critical explosive and loading parameters, and to correlate calculational models to controlled experiments and observed data from large caliber gun prematures.

3. Progress to Date

Roughly speaking, thermal ignition of an explosive occurs whenever the heat conduction process cannot carry away the heat generated by reaction. The reaction heat is in turn governed by the reaction kinetics, thermal properties, and the local temperature of the explosive. The local temperature, for example, can be raised by the deposition of mechanical energy from inelastic deformation of the explosive, frictional effects, or the compression of an included gas bubble. One-dimensional ignition models incorporating these mechanisms will permit direct estimation of the mechanical energy dissipated in terms of dimensionless loading, geometrical, and mechanical parameters. Ignition of the explosive will be determined by numerical solution of the reactive heat equation. Two of these models are discussed below.

a. Gap Ignition Studies

A thermal mechanism that has been advanced to explain in-bore premature initiation of HE-loaded artillery projectiles is HE heating due to compression of an air gap between the pressed or cast explosive and the shell base plate. If nonresistive movement of the HE charge occurs, the air gap will compress at the shell base plate setback pressure. Conceivably, the surface of the HE could be heated to ignition by the compressed gas, and this heating could possibly result in explosion or detonation of the charge.

The proposed thermal ignition model was investigated in detail with numerical thermal transport calculations. The model studied involved numerical solution of the reactive transient heat transfer equation in one-dimensional planar coordinates for the composite case/gap/HE system. Continuity of thermal flux and temperature across each interface was assumed. Temperature and pressure-dependent transport properties were used in the calculations.

The following mathematical equations define the model that was studied. Subscripts 1, 2, and 3 refer to case, air gap, and HE, respectively. Fig. 2 illustrates the coordinate system used and the basic concept of the gap model.

Case or base plate

$$\rho_1 c_1 \frac{\partial T}{\partial t} = \frac{\partial}{\partial X_1}\left[k_1 \frac{\partial T}{\partial X_1}\right] \quad , \quad -\infty < X_1 \leq 0 \qquad (1)$$

Air Gap

$$\rho_2 c_2 \frac{DT}{Dt} = \frac{\partial}{\partial X_2}\left[k_2 \frac{\partial T}{\partial X_2}\right] + \frac{Dp}{Dt} \quad ,$$

$$0 \leq X_2 \leq \delta(t) \qquad (2)$$

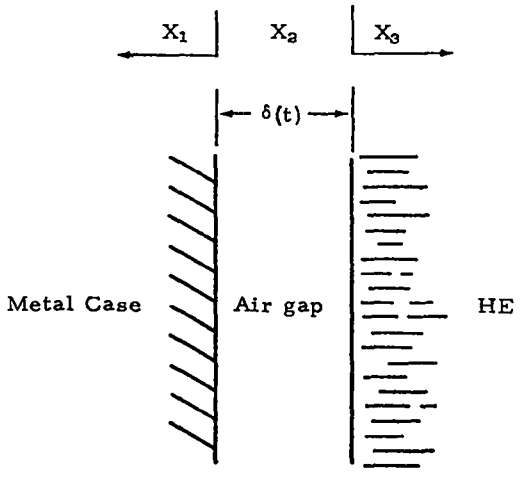
HE

$$\rho_3 c_3 \frac{\partial T}{\partial t} = \frac{\partial}{\partial X_3}\left[k_3 \frac{\partial T}{\partial X_3}\right] + \rho_3 Q z \exp\left[\frac{-E}{RT}\right] \quad ,$$

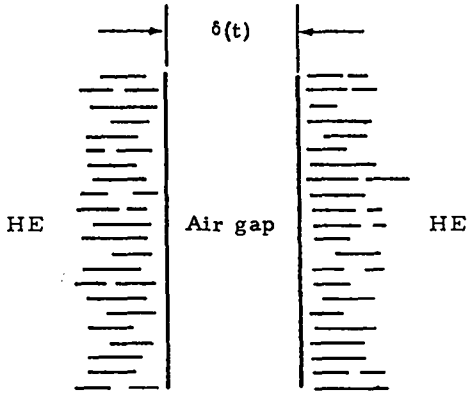
$$\delta(t) \leq X_3 < \infty \qquad (3)$$

Here ρ_i, c_i, k_i, $i = 1, 2, 3$, represent thermal properties - density, heat capacity, and conductivity - which depend on temperature; Q, z, and E are the kinetic properties of the explosive for zero order reaction; and Dp/Dt represents the rate of work done in compressing the gas in the gap. D/Dt is a material derivative. These equations were made dimensionless and programmed for digital solution on LASL's computers using an explicit finite difference scheme.

Preliminary results of the thermal calculations indicate that the gap compression mechanism



Metal-air-HE



HE-air-HE

Gap pressure is assumed equal to setback pressure.

Fig. 2. Gap initiation model.

could indeed result in HE ignition over realistic ranges of setback pressure, rate of increase of setback pressure, gap thickness, and system thermokinetic properties. Thus, although several other thermal mechanisms might also cause ignition (e.g., plastic flow heating due to strain of the HE around voids), compression ignition due to a gap in the shell is a likely source of such in-bore initiations.

We should stress that these thermal calculations merely calculate the point of surface ignition as given by a runaway temperature due to the accelerated pyrolysis reaction in the HE. Whether such ignitions are extinguished in the shell or lead to explosion or detonation is a complex function of shell geometry and HE loading and is beyond the scope of the present calculations. We feel, however, that HE ignition provides a reasonable and conservative criterion of shell initiation sensitivity to setback pressure.

Fig. 3 plots the results of our preliminary calculations expressed as a go/no-go curve in the dp/dt vs δ_o plane. As would be expected, compression ignition is much more difficult to achieve with small gaps.

b. Cavity Model

Another defect in the explosive fill which we are treating is that of a spherical cavity at the base of the fill. Under the setback load, the stress field at the base of the fill is likely to be hydrostatic in nature caused by the confinement of the exterior shell. Thus, we have hypothesized that the stress field away from the defect (three or four cavity radii is sufficient) is hydrostatic and that this time-dependent pressure is the setback pressure. As the setback pressure is increased, a point will be reached where yielding at the inner surface of the cavity takes place. As the setback pressure is further increased, a spherically symmetric plastic zone will extend further into the explosive as shown in Fig. 4. Energy is dissipated in the explosive by the product of the deviatoric stress and deformation rate tensors. Conditions for ignition are then determined by numerical solution of the reactive heat equation with this dissipation term. In material coordinates X_i, this expression is

$$\rho c \frac{\partial T}{\partial t} = \frac{\partial}{\partial X_i}\left(k \frac{\partial T}{\partial X_i}\right) + S_{ij} \frac{\partial v_i}{\partial X_j} + \rho Q z \exp\left[\frac{-E}{RT}\right] , \quad (4)$$

where the summation convention of tensor analysis ($a_i b_i \equiv a_1 b_1 + a_2 b_2 + a_3 b_3$) has been employed and

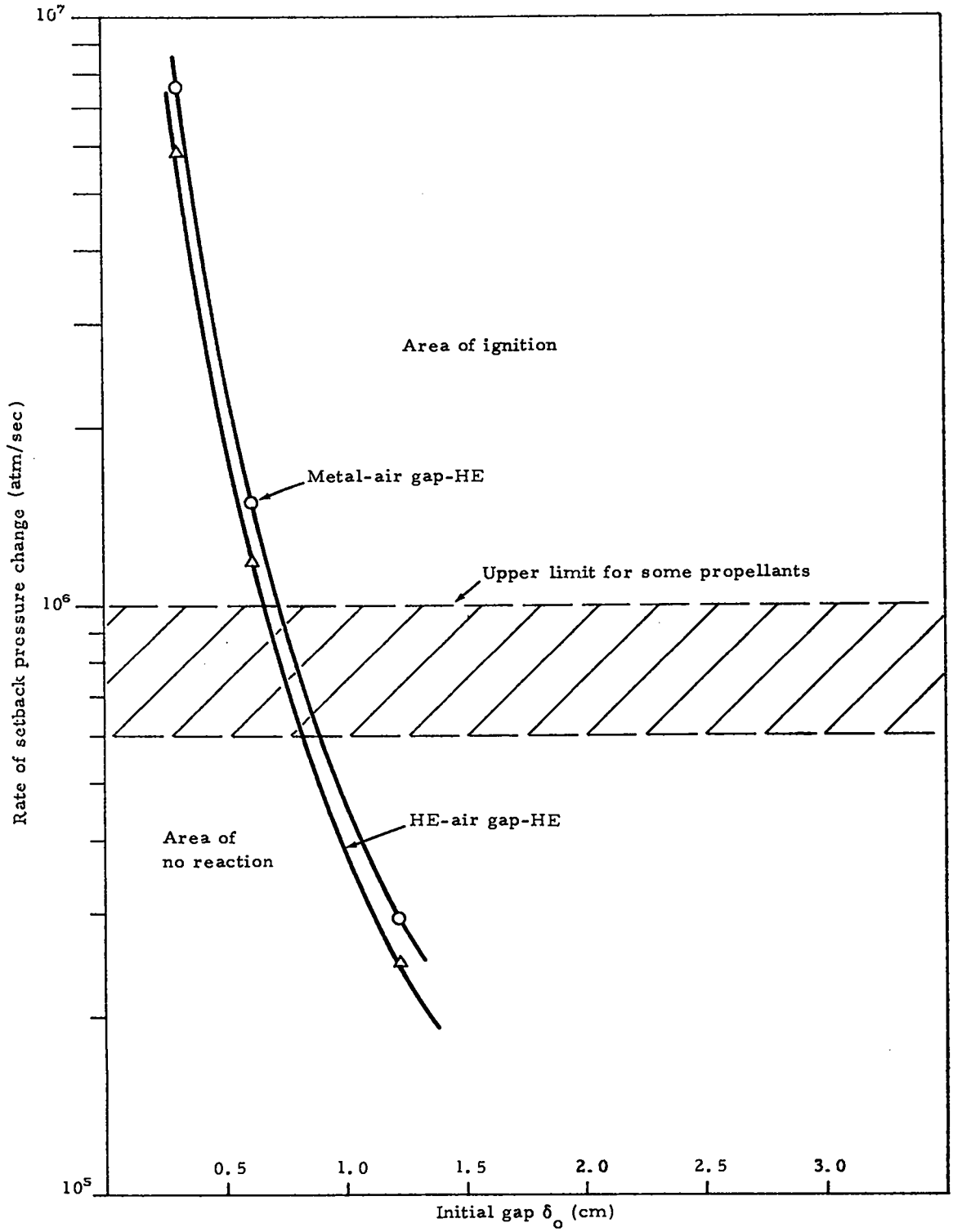


Fig. 3. Critical ignition curve, RDX explosive kinetics.

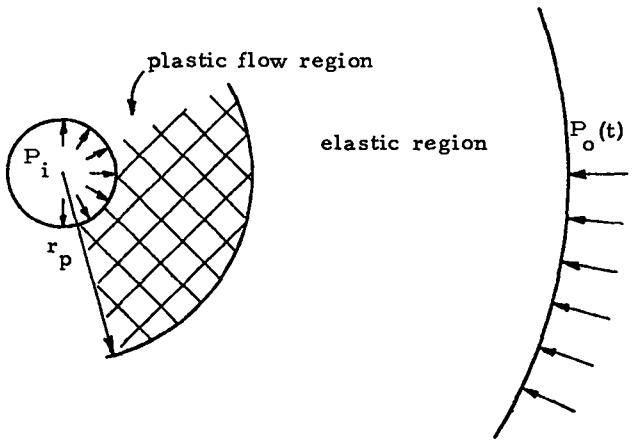


Fig. 4. Thermomechanical model for included gas cavity.

where the thermokinetic parameters are defined as in part a, above. In the above equation S_{ij} and $\partial v_i/\partial X_j$ represent the deviatoric stress and deformation rate tensors, respectively.

Expressions for the energy dissipation term have been obtained for large elastic-plastic deformations around a spherical cavity. We are now in the process of obtaining numerical solutions of the above transport equation. These numerical calculations will also include[27] thermal energy available from gas compression in the collapsed cavity.

The approximations involved in this model are:

(1) The process of cavity collapse is quasi-static in nature and wave propagation effects are unimportant. For typical acceleration time profiles of conventional HE rounds, this is not a bad approximation.

(2) The explosive itself obeys an elastic-plastic constitutive law. However, for setback pressure fields of interest, one would suspect that even an ordinarily brittle material such as an explosive would possess considerable plasticity.

(3) The defect is spherical and the resulting deformation field is spherically symmetric.

Finally, we point out that this model incorporates explosive mechanical parameters (such as the yield strength as a function of temperature, elastic modulus, etc.) which will relate to ignition of the explosive given the setback pressure-time profile and the size of the defect. These mechanical parameters can be varied, in many cases, by changing the binder, the explosive particle size, or by adding waxes or plasticizers to the explosive.

4. Future Work

The main activity on the premature program during the next quarter will be setting up an experimental program to verify the analytical models, particularly the gap compression model. After having considered the available HE sensitivity tests, as far as their capability to apply a time-dependent stress to an explosive charge that realistically simulates that of large caliber guns, the Picatinny Activator or setback simulator (with additional instrumentation) offers the best tool to study the gap compression model. In addition, we are considering the use of sensitive explosives, possibly at elevated temperature with a sensitizer added, in order to reduce the number of tests for model verification.

We will also be continuing the calculational work on the cavity model and the gap compression model. One hypothesis which has been advanced for initiation of explosives under short duration shocks of amplitude p and duration τ is the requirement that $p^2\tau$ = constant for a given explosive.[24,28] Using the two models, we will examine the $p^2\tau$

requirement for longer duration pressure profiles typical of large caliber guns.

Other calculational work is in progress which treats both hydrodynamic and thermal behavior for a gap compression model.

C. SYNTHESIS OF HMX (TASK C), M. D. Coburn, T. M. Benziger

1. Introduction

Unpublished research conducted at PA and the University of Idaho has led to a new process for converting hexamethylenetetramine to pure HMX.

This process has the potential for producing HMX at a cost that is comparable to the present cost of RDX. Treatment of a slurry of hexamethylenetetramine and ammonium acetate (NH_4OAc) in water with acetic anhydride (Ac_2O) gives a solution containing a near quantitative yield of DAPT. The DAPT can be converted to 1,5-diacetyl-3,7-dinitrotetraazacyclooctane (DADN) in high yields when the crude solution is added to a mixture of nitric and sulfuric acids. Subsequent nitrolysis of DADN should give HMX. These reactions are illustrated in Fig. 5.

Working closely with PA, LASL has undertaken a development program designed to optimize the preparation of DADN and to determine its properties. The nitrolysis of DADN to HMX is being investigated by PA on a laboratory scale.

Results of laboratory scale investigations conducted by PA on the preparation of DADN indicate that two problems must be solved before the reactions can be scaled to larger equipment. The first is the sudden uncontrolled exotherm that occurs during the nitrolysis of DAPT to DADN. A large excess of nitric acid has been used by PA to control the exotherm. The second problem is the separation of DADN from the reaction medium. Separation has been achieved on a laboratory scale at PA with the addition of approximately 10 volumes of a mixture of ice and water. This quantity of water would be prohibitive in large-scale production.

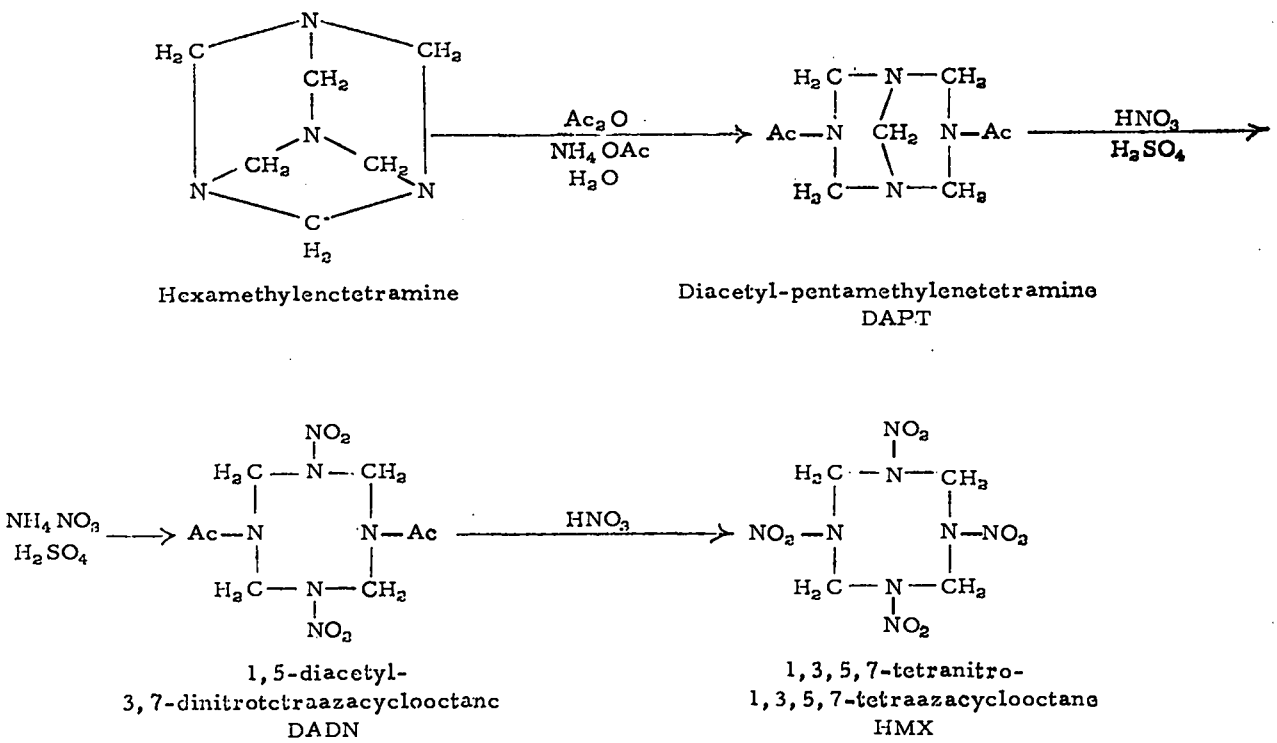


Fig. 5. Synthesis of HMX.

2. Activities to Date

During this report period we found that DADN is virtually insoluble in 40-50% sulfuric acid and we have concluded that DADN should separate from solution when the nitrolysis mixture is diluted with about two volumes of water if the amount of excess nitric acid is minimized. The PA results indicate that when near stoichiometric amounts of nitric acid are used in the nitrolysis of DAPT to DADN, the undesirable exotherm occurs. Since the source of the exotherm may be the nitric acid oxidation of the formaldehyde that is formed in this reaction, we decided to try a mixture of AN and sufuric acid as the nitrolysis medium with the hope that the resulting ammonium bisulfate would react with the formaldehyde to give the salt of methyleneimine. This salt is known to be stable in acidic media. An alternate consideration behind this approach is that AN may be reversibly dehydrated by concentrated sulfuric acid to nitramide; thus, nitric acid may be available only on demand in such a system and an excess of nitric acid may never exist. In these experiments with the nitrolysis of 0.1 mole of DAPT with AN in sulfuric acid, we found that no exotherm occurred if at least 2.25 moles of free sulfuric acid was present. Thus, we had to employ an additional amount of sulfuric acid above 2.25 moles equivalent to the AN used in order to avoid the exotherm (Table III). We were also pleased to find that the highest yields of crude DADN obtained by this method resulted when nearly stoichiometric quantities of AN were used. In these experiments we found that quenching the reaction with approximately two volumes of water was sufficient to cause the DADN to crystallize from the solution.

Since the exotherm occurs after a reaction time of 95 min when 0.1 mole of DAPT is nitrolyzed with 0.33 moles of 98% nitric acid in 2.25 moles of sulfuric acid (PA results), we made an identical run with a reduced reaction time that gave a reasonably good yield of DADN with no exotherm. Other runs were made with commercial 90% nitric acid in sulfuric acid with oleum added such that the water was removed and approximately 2.25 moles of sulfuric acid was present in the resulting mixture. The results of these experiments are given in Table III. The purity of the products could not be determined because our nmr spectrometer is in need of repair; however, the PA data indicate that products melting above 250° are better than 90% pure.

On the basis of very limited data that has not been verified with duplicate runs, it appears as if the problem of isolating the DADN from the nitric acid-sulphuric acid reaction medium can be solved. There is also the possibility of reducing the exotherm with increased reaction times. Since neither the isolation nor the exotherm problem appears to exist with AN in sulfuric acid, we feel that this system should be evaluated further.

3. Future Work

During the next report period, we will begin a parametric study of the DAPT preparation.

D. INITIATION AND SENSITIVITY (TASK D), B. G. Craig

1. Introduction

The objective of this task is to demonstrate the feasibility of studying the response of Comp. B to relatively short duration, plane shock waves. This objective is part of a long-range objective which is to develop and calibrate a quantitative model for predicting the response of a variety of common explosives to a broad range of stimuli, or to obtain a more quantitative understanding of initiation process. Emphasis is to be placed on defining those conditions that result in the violent decomposition type behavior, i.e., deflagration, or low order detonation.

Specific tasks are to develop systems for generating plane shock waves of appropriate peak pressures, peak-pressure durations, and release wave patterns; to apply these systems to an explosive of interest to the military (Comp B); to develop techniques for obtaining quantitative data

TABLE III

NITROLYSIS OF DAPT SOLUTION[a] (0.1 mole) AT 18 ± 1°C

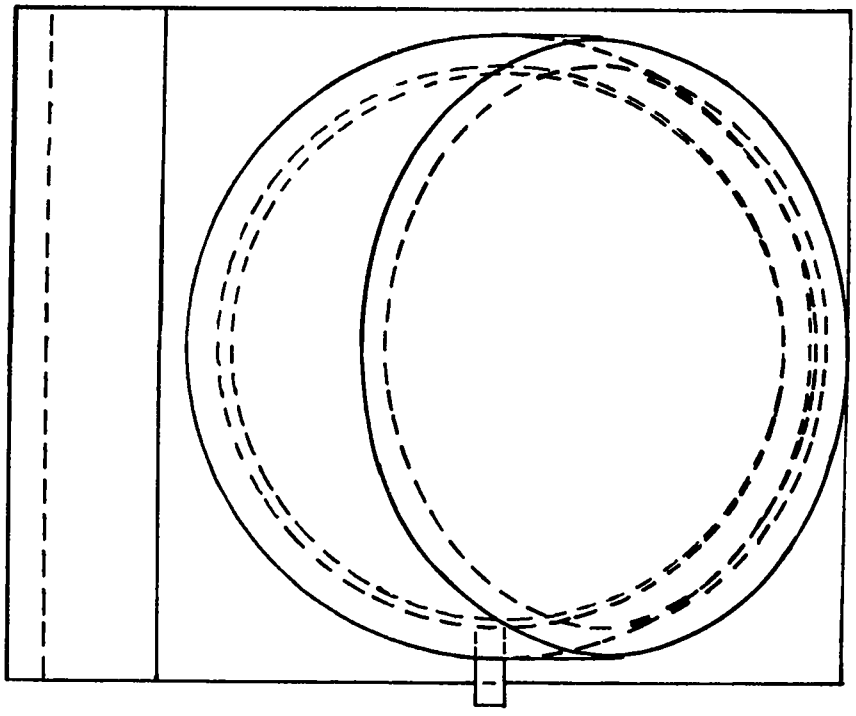
Nitrolysis Medium	Time (min)[b] Addition	Time (min)[b] Total	Exotherm	DADN Crude Yield	Melting Point (°C)
98% HNO_3 (1.0 mole) 98% H_2SO_4 (2.25 mole)	80	100	No	26.4 g (91%)	264.5
98% HNO_3 (0.33 mole) 98% H_2SO_4 (2.25 mole)	70	90	No	25.3 g (87%)	253.1
90% HNO_3 (1.0 mole) 98% H_2SO_4 (1.12 mole) 30% Oleum (60 ml)	75	95	No	26.9 g (93%)	259.5
90% HNO_3 (0.5 mole) 98% H_2SO_4 (1.68 mole) 30% Oleum (30 ml)	77	97	No	26.1 g (90%)	262.4
90% HNO_3 (0.33 mole) 98% H_2SO_4 (2.25 mole)	50	100	Yes	24.4 g (84%)	260.4
NH_4NO_3 (0.66 mole) 98% H_2SO_4 (2.25 mole)	70	90	Yes	20.6 g (71%)	252.6
NH_4NO_3 (0.66 mole) 98% H_2SO_4 (2.91 mole)	80	100	No	22.6 g (78%)	255.5
NH_4NO_3 (0.33 mole) 98% H_2SO_4 (2.25 mole)	80	100	Yes	23.6 g (81%)	252.0
NH_4NO_3 (0.33 mole) 98% H_2SO_4 (2.58 mole)	75	95	No	26.2 g (90%)	251.3
NH_4NO_3 (0.25 mole) 98% H_2SO_4 (2.50 mole)	70	90	No	25.9 g (89.5%)	250.1
NH_4NO_3 (0.25 mole) 98% H_2SO_4 (2.50 mole)	50	70	No	23.0 g (79%)	249.6

[a] Procedure - Acetic anhydride (0.3 mole) was added dropwise to a rapidly stirred slurry of hexamethylenetetramine (0.1 mole) and ammonium acetate (0.082 mole) in water (7 ml) at 5-10°C. The resulting solution was stirred for 30 min, then it was added to the nitrolysis medium as indicated above.

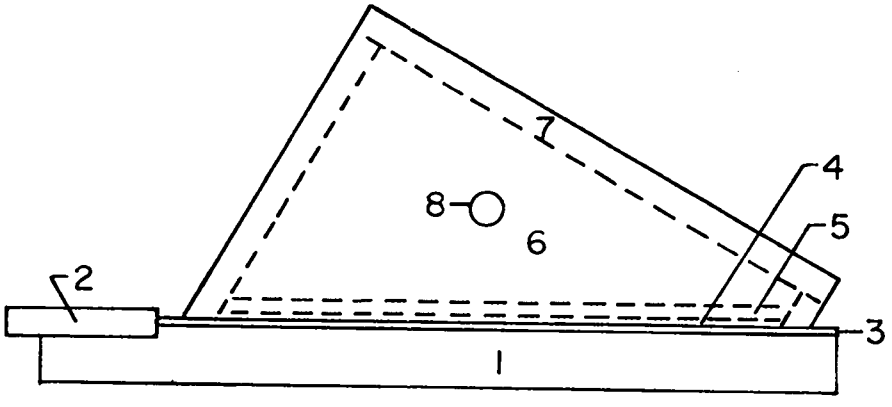
[b] The total time represents the time from the start of the addition of the solution to the nitrolysis medium to the quench of the reaction. The addition time represents the time required to add the solution, so the difference between these times is an age time.

relating to the reaction of the test explosive as a function of time; and to deduce the range of conditions which result in violent decomposition of the test explosive.

Short duration shock waves are readily generated by guns; however, violent decomposition of large explosive charges is not compatible with experiments done with guns. We propose to attempt to develop three explosive systems which will drive a flyer plate across a relatively large gap (a vacuum) and impact a buffer plate which is in contact with the explosive sample to be studied. A schematic of the plane wave shock wave generator, sometimes called a "mousetrap", is shown in Fig. 6. The flyer plate thickness will be fixed for each system; duration of the peak pressure induced into the explosive sample will be varied by varying the thickness of the buffer plate. Current specifications for the shock wave generators call for systems which will induce peak pressures near 25, 45, and 70 kbar into Comp. B. Precision line wave generators and precision sheet



Top view



Side view

Key

1.	Base plate	5.	Flyer plate
2.	Line wave generator	6.	Vacuum
3.	Sheet explosive	7.	Target (buffer plate)
4.	Gap (optional)	8.	Vacuum connection

Fig. 6. Essential features of a plane wave shock wave generator.

explosive have been fabricated at LASL in an effort to make better shock wave generators than can be made with commercially available materials. Current specifications call for the shock waves to be simultaneous within 0.35 μsec over the central 15-cm diam. This will allow study of Comp. B samples up to 7.5-cm thick.

In the event suitable plane wave generators of this type cannot be developed, alternative techniques are available. A gun with significantly smaller charges than those desired could be used. These experiments would be supplemented by experiments with large charges impacted by flyer plates thrown by conventional high explosive systems. The appropriate early shock conditions could be attained in the latter experiments but the following waves would be more complicated than desired.

2. Results to Date

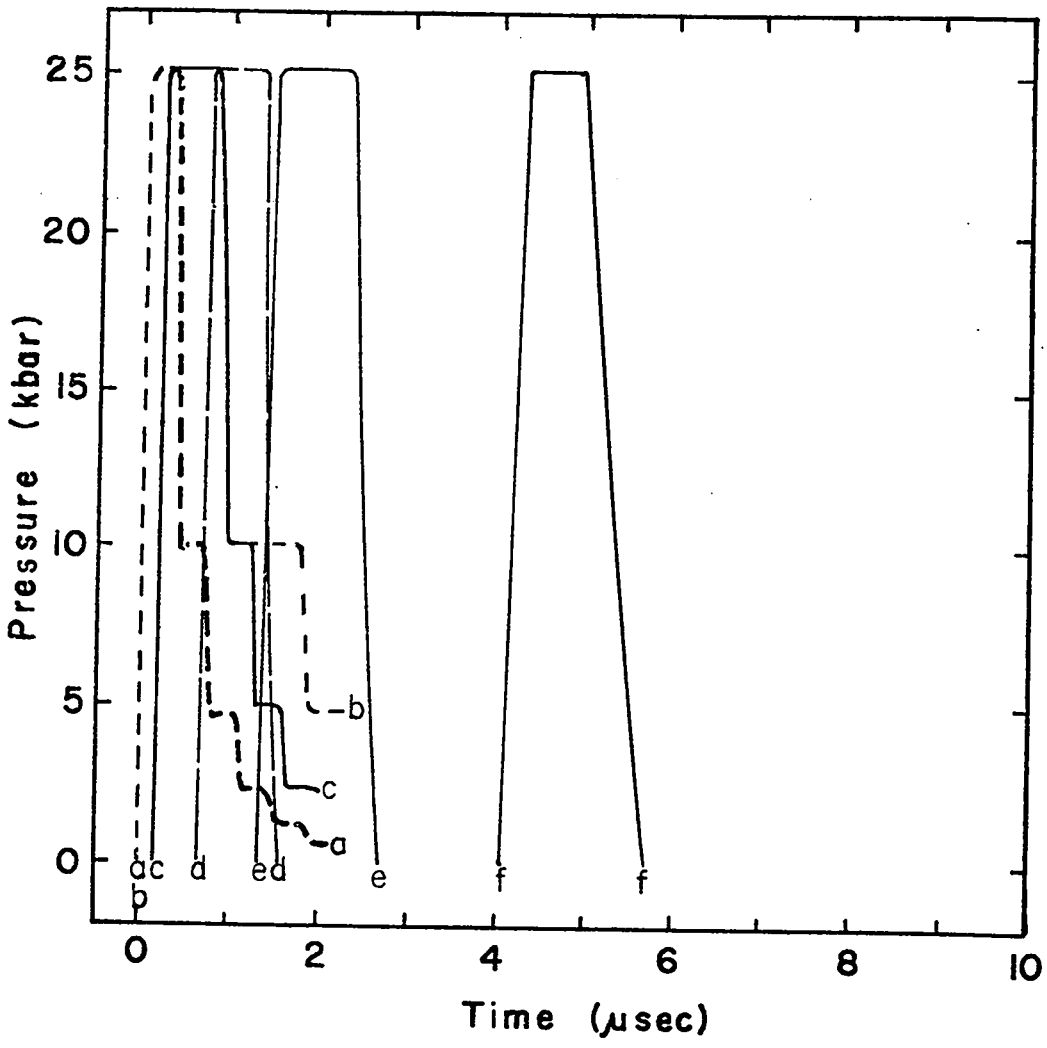
The literature has been researched and durations and peak pressures of interest have been estimated. The best expressions in the literature of durations at given peak pressures are in terms of dural flyer plate thicknesses.

Computer programs have been used to transform from dural plate thickness to time duration of the peak pressure in the Comp. B at or near the collision boundary. In one program, PAD, elastic-plastic effects were ignored; in the other program, SIN, elastic-plastic effects (at a greater cost of computer time) were included. Selected identical problems were run with both programs. The results implied that elastic-plastic effects are relatively small for the conditions of interest; consequently all subsequent calculations have been made with PAD.

Subsequent calculations include a parameter study to determine the desirable materials to be used as flyer and buffer plates in the design of plane wave generators. Some of the results obtained are plotted in Figs. 7, 8, and 9. The materials considered in this study were dural, magnesium, beryllium, nickel, copper, polyethylene, and Plexiglas. Dural was among the poorest materials in terms of the change in the peak pressure duration as a function of thickness of the buffer plate. Beryllium was unique in that a given flyer plate thickness produced the shortest duration peak pressure. The relatively slow motion required for nickel or copper resulted in unusually tight dimensional tolerances in order to achieve the desired simultaneity of arrival. Furthermore, the use of materials with a shock impedance greater than that of unreacted Comp. B resulted in undesirable relief wave patterns (stepped) or in a succession of complicated shock and relief waves. Only those materials with a shock impedance lower than that of unreacted Comp. B, for example, polyethylene and Plexiglas, resulted in the desired consistently smooth relief wave and freedom from successive shocks during the possible time of interest. Also, the higher shock impedance materials did not result in as much change in the duration of the induced peak pressure with change in buffer plate thickness as desired.

On the basis of the parameter study, Plexiglas was selected as the most desirable flyer and buffer plate material assuming it could be subsequently demonstrated that the Plexiglas flyer plates do not spall in an undesirable fashion. The flyer plate thickness was chosen to be 3 mm as a compromise between practical tolerances, peak pressure duration, and gradient of the release wave.

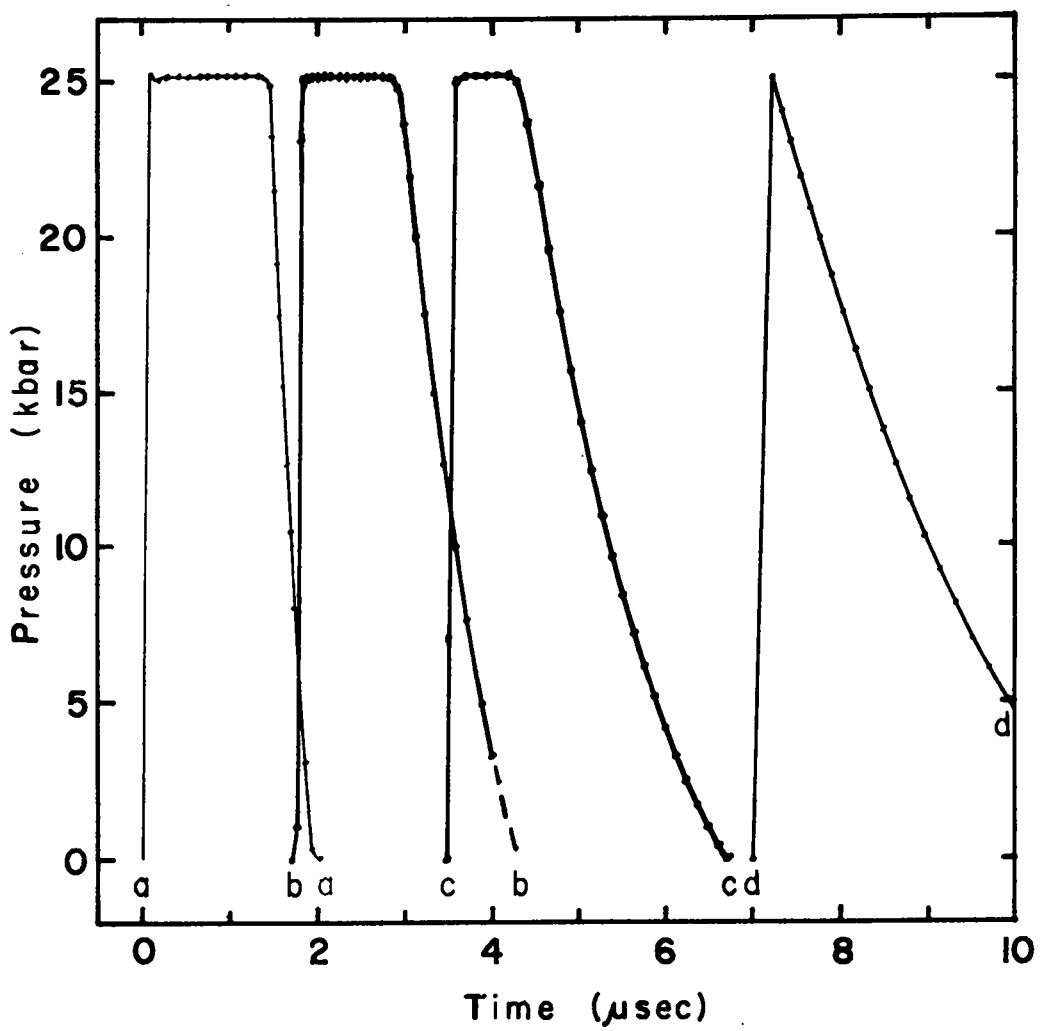
Several prototype shock wave generators were designed and constructed in half scale. The use of half scale reduced the cost for "rough" experiments. The simultaneity of arrival of the shock wave induced into the buffer plate at the 1-cm level by the flyer plate was measured and most of the Plexiglas parts were recovered in fragments. A number of the fragments were positively identified as being from the flyer plate; a layer about $\frac{1}{2}$ mm thick had disappeared from the side originally adjacent to the sheet explosive. We believe that



Key		
Curve	Flyer Thickness (mm)	Buffer Thickness (mm)
a-a	1.0	0
b-b[a]	2.5	0
c-c	2.0	1
d-d	2.0	4
e-e	3.0	8
f-f	3.0	24

[a]Maximum duration of interest for this pressure.

Fig. 7. Calculated shock pressure induced into the first cell of unreacted Comp. B as a function of time for various thicknesses of dural flyer and dural buffer plates. Zero time is the time when the flyer plate contacted its target (buffer plate or Comp. B). The flyer-plate velocity was 0.59 mm/μsec for all cases and induced a peak pressure of 25 kbar into unreacted Comp. B. Elastic-plastic and spalling effects were neglected in these calculations.

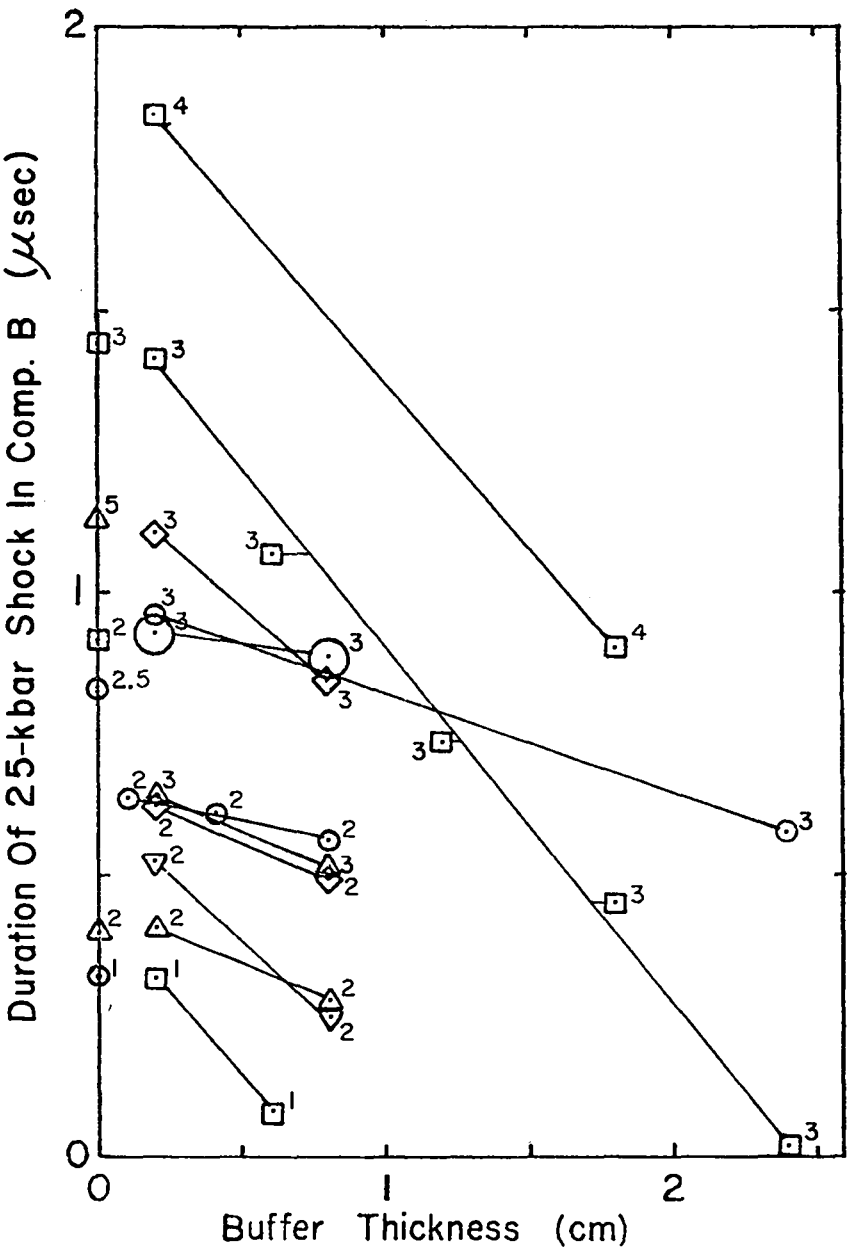


Key

Curve	Buffer Thickness (mm)
a-a	0
b-b	6
c-c	12
d-d[a]	24

[a]Compare with curve f-f of Fig. 7.

Fig. 8. Calculated shock pressure induced into the first cell of unreacted Comp. B as a function of time by a 3-mm-thick flyer plate of Plexiglas and various thicknesses of Plexiglas buffer plates. Zero time is the time when the flyer plate contacted the buffer plate. The flyer plate velocity was 1.03 mm/μsec for all cases. Elastic-plastic and spalling effects were neglected in these calculations.



Key

Symbol	Flyer	Buffer	Symbol	Flyer	Buffer
⊙	Dural	Dural of Comp. B	⊡	Plexiglas	Plexiglas or Comp. B
◉	Dural	Copper	◇	Plexiglas	Copper
△	Beryllium	Copper or Comp. B	▽	Magnesium	Plexiglas

Fig. 9. Representative results from the parameter study showing the effect of flyer and buffer plate materials and thicknesses on the duration of the peak pressure induced into Comp. B by the buffer plate. The numbers beside each symbol are the flyer plate thickness in mm. All data plotted were obtained with the PAD computer program. The flyer plate velocities were adjusted to result in a 25-kbar shock being induced into Comp. B regardless of the type of plate material. Similar plots were obtained for peak pressures of 45 and 70 kbar.

either the layer was eroded by action of detonation products or the layer spalled after the flyer impacted the buffer. In the latter case it would not affect the experiment. In the former case we can compensate by either increasing the flyer plate thickness, by decreasing the buffer plate thickness, or possibly by reducing the erosion by introducing a small gap between the sheet explosive and the flyer plate.

The angle defined by the original positions of the flyer plate and the buffer plate has been adjusted in successive experiments. Currently we have two half-scale generators of different pressures which produce shocks at the 1-cm level in the buffer which are simultaneous within 0.33 μsec over the central 7.6-cm diam. A third system within tolerance is expected soon. The peak pressure of these systems has not been accurately measured; the duration of the peak pressure and the gradient of the following wave were measured in one shot. From the geometries which give the most nearly simultaneous arrival traces, the peak pressures appear lower than expected for the 70-kbar system, about right for the 45-kbar system, and higher than expected for the 25-kbar system. The one available measure of peak-pressure duration suggests that the duration is significantly shorter than predicted by the PAD calculations; the gradient of the relief wave is approximately as predicted by the PAD calculations. It will be relatively easy to lengthen the duration of the peak pressure if the above results prove valid.

A concurrent effort is under way to develop manganin-gauge assemblies of high sensitivity and lifetime under shock loading. This work has progressed to the point where the gauge typically remains functional for at least 15 μsec after the initial shock in the absence of a Comp. B sample. The one measure of peak-pressure duration and relief-wave gradient was obtained with a manganin-gauge assembly. The gauge has not been calibrated so the peak pressure cannot be deduced accurately from the experiment.

3. Future Work

Parts for three full-scale shock wave generators are being fabricated. These will be tested as soon as practical. Current plans call for manganin gauges to be used to measure peak-pressure durations and relief-wave gradients in these experiments. These experiments will be followed with similar tests of designs modified in accordance with the outcome of the previous designs until adequate simultaneity, peak pressure, peak-pressure duration, relief-wave gradients, and reproducibility are obtained.

As soon as it has been demonstrated that a particular manganin-gauge configuration will function satisfactorily in the full-scale system, that design will be frozen and the gauge will be calibrated.

Following the development and proofing of both the shock wave generators and the manganin gauge, a series of tests designed to survey the response of Comp. B to selected shock waves will be commenced. Reproducibility will be checked early in this series. Completion of the survey will be followed by a report with recommendations for additional studies as may appear justified.

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