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Deflagration-to-Detonation Transition in Granular HMX*

A. W. Campbell

Introduction

For the past three decades the study of the deflagration-to-detonation transition (DDT) has been strongly influenced by the views of G. B. Kistiakowsky.¹ Very briefly stated, he postulated the origin of this phenomenon as localized burning in a mass of subdivided explosive, with the possibility of confinement either by a strong container or by the inertia of the explosive mass. Resistance to the flow of gases through the interstices of the explosive mass causes the pressure at the site of burning to rise, and the rate of burning increases with the pressure. If "conditions are favorable, ... shock waves will be formed within the body of the explosive ... and ... with their discontinuous rise in pressure and the accompanying, more-than-isentropic rise in temperature, will start rapid deflagration of the explosive layers through which they pass... The energy thus released reinforces the shocks. Eventually they reach such intensity that the entire explosive is consumed in their passage and therefore the entire available energy may be utilized for their propagation."

Attempts to elaborate this description into a detailed view of all the steps in the process have occupied many workers. The question is how shock waves are formed by the burning explosive. Mack² and Price and Wilson³ showed that burning, initiated by a hot wire, in cast DINA and Pentolite would transit to detonation if the explosive were heavily confined. Mack hypothesized that pressure increases from the burning propagate as a wave, that this wave steepens into a shock at some distance from the burning zone, and that this shock wave continues to grow by receiving energy from the accelerating

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burning. Finally, the amplitude of the shock wave is large enough to initiate detonation.⁴ Jacobs has shown that Macek's treatment of shock formation is inadequate.

Another hypothesis, widely investigated, is that convective combustion plays a central role in bridging the gap between conductive burning and the transition to detonation.⁵ Driven by the pressure gradient generated by burning, hot combustion products spread burning at an ever faster rate. Ultimately, under favorable conditions of inertial or other confinement, the rate of energy release is sufficient to cause shock formation and the initiation of detonation. For this process the permeability of the explosive to gases is a factor of prime importance. Several workers have shown that indeed the distance to the onset of detonation depends strongly on the permeability. Griffiths and Groocock⁶ studied the distance to detonation when burning was initiated in granular tetryl, RDX, HMX, and TNT under heavy confinement. Some of their data for HMX are shown in Fig. 1, where distance to transition is plotted as a function of the permeability, which was changed by varying the particle size. Price, Bernecker, and their coworkers have explored DDT in a variety of explosives and propellants over a wide range of densities and have published a series of reports describing their work. In agreement with the results shown in Fig. 1, they also observed a minimum in the transition distance in granular tetryl⁷ as a function of the density or permeability. Much additional work has been done. Convective combustion has been a key concept in studies of the DDT process. But attempts to model DDT about convective combustion have been beset with formidable difficulties, prominent among which have been very rapid surface ignition, bed collapse, and choked flow of the combustion products.

It is difficult to reconcile the convective combustion picture with some experimental observations. Brandon⁸ observed in work with granulated

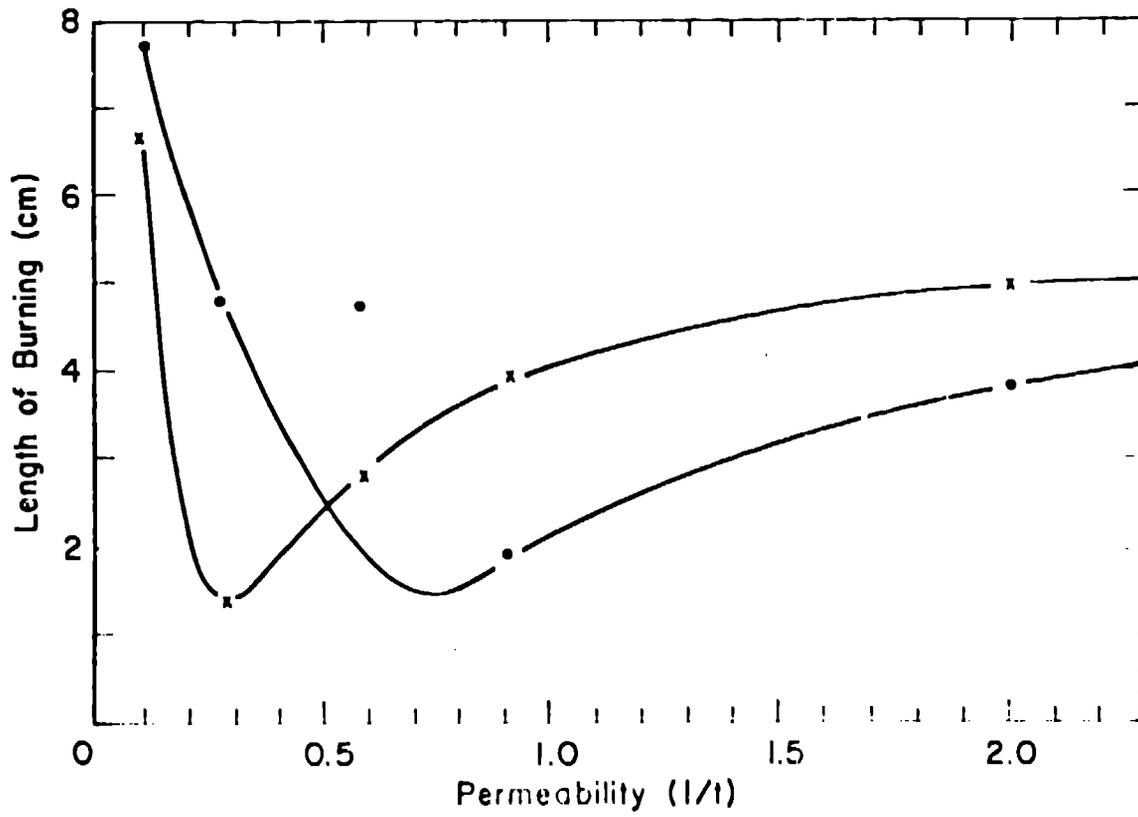


Fig. 1. DDI distance in HMX. Permeability varied by change in HMX particle size. Fine HMX at the left, coarse at the right. Permeability in arbitrary units.

propellant that the onset of detonation could occur well beyond the burning front, separated from it by a region that emitted no light, and that sometimes detonation ran backward from the detonation to the burning zone. This "dark zone" and the occurrence of detonation were also reported by Griffiths and Grocock. Price, Bernecker, and coworkers observed that their ionization pins were triggered by a rapid wave which swept through much of the explosive. They also found, from records of strain gauges, that a second wave, stronger and more significant for leading to detonation, originated in the combustion zone and followed the first wave.

In this paper the following hypothesis is presented. When burning starts, the hot combustion products flow away through the porous explosive, heating and igniting additional explosive and being cooled in the process. As the burning increases, drag forces on the combustion products cause the pressure to rise in the burning zone, and the pressure rise causes the rate of combustion to increase. As the pressure rises further, the bed begins to compact and the permeability starts to decrease and to further impede the flow of gases. After a time, the explosive beyond the combustion zone is compacted to such a degree that it becomes effectively impermeable to combustion products and forms a plug, driven by the high-pressure gas behind it. The compaction wave at the head of the plug steepens as the higher pressure waves from the combustion zone overtake it. In the initial stages, the compaction is so slow that shear forces do not ignite reaction. At later stages, compaction becomes more rapid and ignition by shear becomes general. From this point on, the initiation of detonation follows the usual course for initiation of a heterogeneous explosive. Experiments to support this hypothesis are presented below.

Experimental Apparatus

The experimental method was similar to that of Griffiths and Grocock. Granular explosive was packed in a heavy-walled steel tube closed at both ends

by plugs sealed with epoxy. Steel masses of several kilograms were placed over the ends of the tube for inertial containment of the plugs. Burning was ignited at one end of the column of explosive, and the course of events was evaluated from measurement of the changes in the internal diameter of the steel tube or in the wall thickness of the tube fragments. DDT was assumed to be complete when the internal diameter reached a maximum, or the tube wall thickness reached a minimum.

Igniter

In most of the experiments, 200 mg of a mixture of finely divided titanium powder and amorphous boron, ignited with an exploding bridge-wire, formed the igniter. This mixture was tamped in place to a density of 1.3 g/cm^3 and formed a plug 9.6 mm in diameter, tapering to 6.3 mm in diameter, and having a thickness of 3.8 mm. From data in the literature⁹ this igniter released about 220 calories, but when one considers the slow rate of fusion (ca. 3cm/s), the measured release of six cubic centimeters of permanent gas (probably mostly hydrogen), and the rate of heat loss (adiabatic temperature ca. 3770°C), it seems probable that only a small part of the igniter heat release was involved in any experiment.

Explosive

Three lots of HMX of differing degrees of fineness (Table I) were used in these experiments. If it were not for the press of time, it would have been desirable to have removed some of the finer and coarser portions of the Class A and Class C samples by sieving. By this action the danger of segregation during the handling of small quantities of the powders could have been reduced.

In the following discussion of experiments, they will be referred to by number as listed in Table II.

TABLE I
HMX PARTICLE-SIZE DATA

Sieve Opening (μm)	F (HOL-934-3) (%)	Class A (HOL-920-32) (%)	Class C (HOL-703-9) (%)
1400			0.3
1000			2.8
710			8.6
500		1.3	17.9
350		4.0	19.6
250		15.6	18.2
177		18.2	9.3
125	1	27.8	8.0
98		11.8	5.4
62		12.1	
44	7	4.9	
SS	92	4.3	9.9
Specific Surface area (cm^2/g)			

TABLE II
SHOT SUMMARY

Expt. No.	Shot No.	HMx	(mm)	Disks (No.)	Booster (mg)	HMx ^d DDT (mm)
1	C-5009	F	0			72
2	C-5004	F	1.8	15 (27) ^b	220	83
3	C-4983	A	0			45
4	B-8500	A	0.8	6 (4.8)	685	46
5	C-5002	C/A ^a	1.8	1 (1.8)	220	99
6	C-4998	C	0			58
7	C-4958	C	0.8	13 (10.4)	220	67
8	C-4997	C	1.9	12 (22.8)	220	67
9	C-5013	C	1.8	8 (14.4)	440	52
10	C-5000	C	1.8	18 (32.4)	220 ^c	106 ^e
11	C-5014	C/C ^a	1.8	1 (1.8)	220	48
12	C-5015	A/C ^a	1.8	1 (1.8)	220	43
13	C-5005	F/C ^a	1.8	1 (1.8)	220	32

^aThe left symbol denotes the HMx type used for the 220-mg combustion increment; the second symbol identifies the HMx type filling in remainder of tube.

^bNumbers in parentheses are the total thickness of disks traversed before DDT occurred and are obtained as the product of disk thickness and the number of disks traversed.

^cLoaded at 0.6 g/cc.

^dEntries are the net length of HMx column traversed from igniter to the point at which detonation occurred.

^eMeasured from disk at top of brass tube.

Experimental Results

Experiments 3,4. Experiment (4) is diagramed in Fig. 2. A series of diaphragms cut from neoprene sheet 0.8 mm thick was arranged throughout a column of HMX-A, and burning was ignited at one end. The diaphragms in this experiment and those in (7) were cut with a cork borer and were somewhat ragged; however, it appears that when all of the results are considered together, these disks were adequate for the purpose.

The purpose of (4) was to interfere as much as possible with convective combustion and then to observe whether DDT still occurred. It did occur and at a distance in HMX-A of 46 mm. (See footnote to Table II.) This result was encouraging in that the distance to DDT was in the range found by Griffiths and Grocock (Fig. 1).

For comparison, experiment (3) was fired (Fig. 3). With no diaphragms present, the distance to DDT was found to be indistinguishable from that of (4). (Fragments of the steel tube are shown in Fig. 4.) This agreement was puzzling at the time, and proved later to depend partly on the size of the initial HMX increment in (4).

Experiments 1,2 and 6,7. In these experiments the effect of diaphragms on the DDT distance was explored in HMX-F and HMX-C. A standard initial increment size of 220 mg of HMX was adopted and, where the diaphragm thickness of 1.8 or 1.9 mm was used, the disks were machined on a lathe to a precision of 0.02 mm so as to show a slight interference fit when tamped in place.

In experiment (1) (Fig. 5) the steel tube was increased in diameter to 76.2 mm. At this diameter the tube did not burst during firing and a more desirable record was obtainable (Fig. 6); however, because of the long wait required for the machine work this size tube was not used in other experiments.

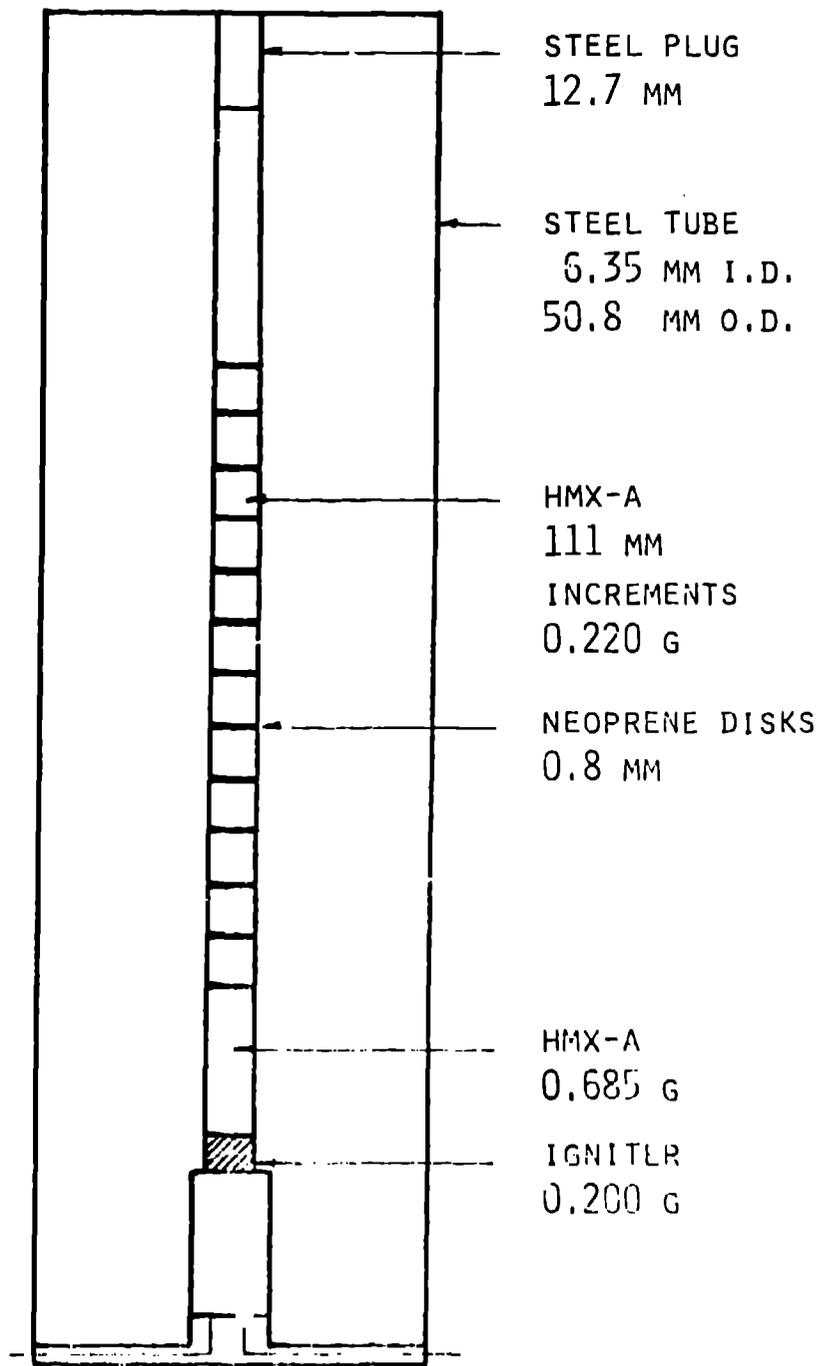


Fig. 2. Experiment 4.

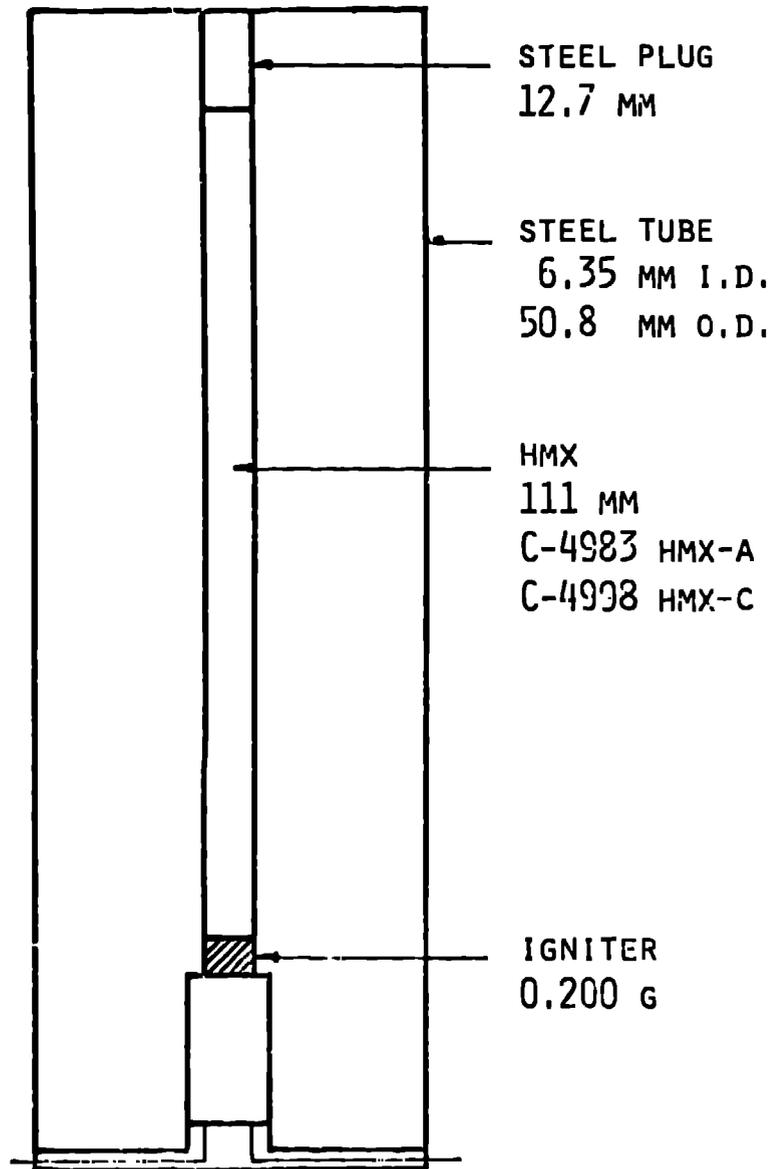
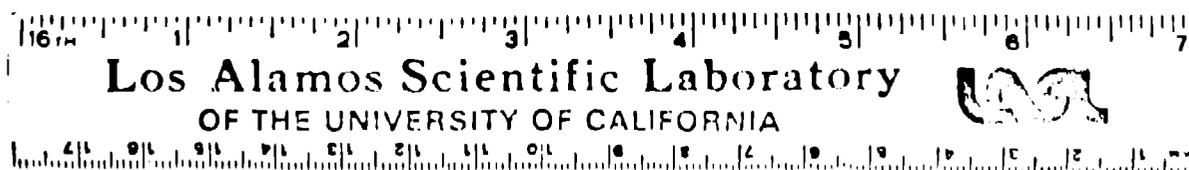
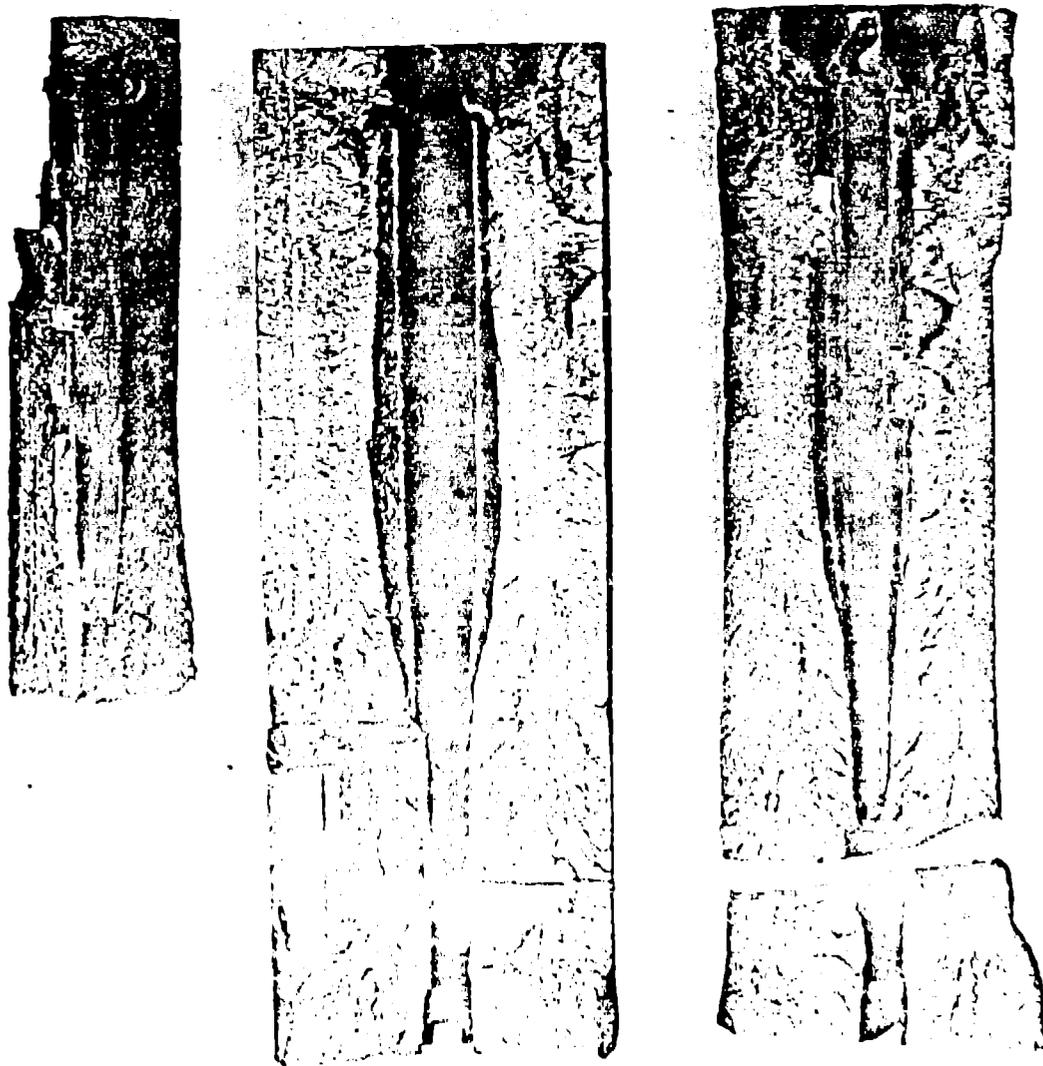


Fig. 3. Experiments 3 and 6.



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Fig. 4

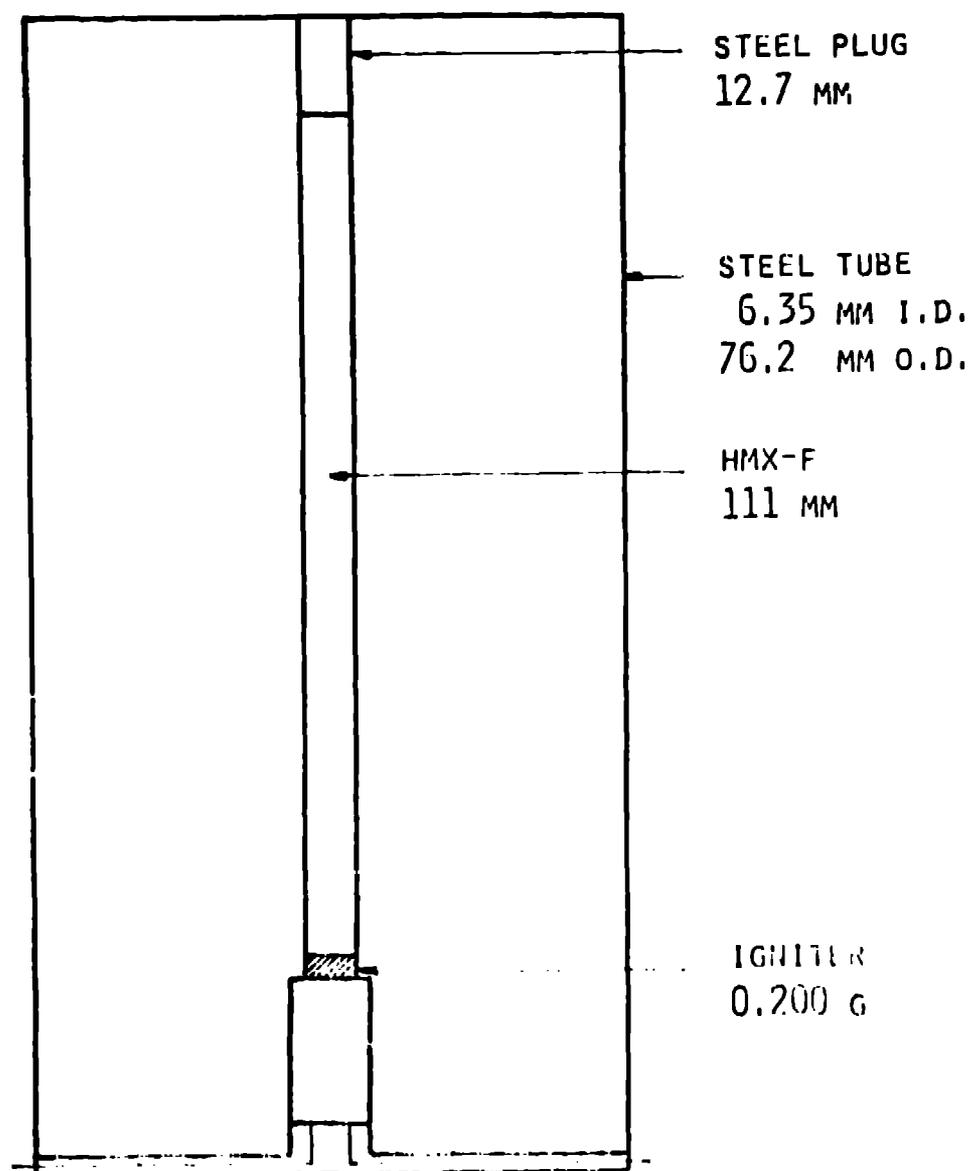
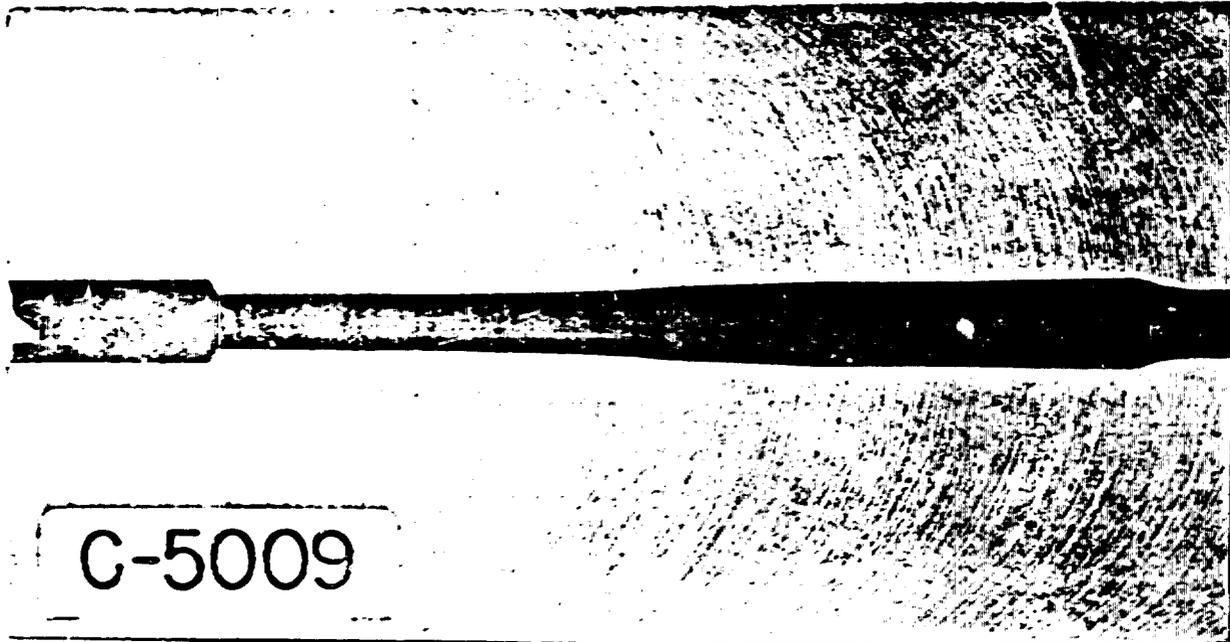


Fig. 5. Experiment 1.



16 15 14 13 12 11 10 9 8 7 6 5 4 3 2 1

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Experiment (6) was as diagramed in Fig. 3. From the results of experiments (1), (3), and (6) it is evident that there is a minimum in the DDT distance as a function of the average particle size, of specific surface area, or presumably, of permeability as found by Griffiths and Grocock.

The effect of diaphragms in HMX-C was tested in experiment (7) (Fig. 7) and in HMX-F in experiment (2) (Fig. 8). When the results of these experiments are compared with those from (1) and (6), it is seen that the DDT distances were resolvable longer when diaphragms were present. If significant burning were confined to the initial increment of HMX, then some of this increase in distance to detonation might be due to the small size of the initial increment (220 mg). Recall that the results of (3) and (4) were almost identical. Obviously, if the initial increment were reduced to near zero, a very long DDT distance might be expected to result; on the other hand, if the first diaphragm were removed to a distance of, say, 100 mm from the igniter, it might have no perceptible effect on DDT.

Experiments 8,9. Experiment (9) was a test of the effect of doubling the size of the initial increment, but since the preferred diaphragms were now 1.8 mm thick, it appeared necessary to repeat the reference experiment (7) with the thicker disks. Experiment (8) (Fig. 8) employed the thicker disks and gave a DDT distance (67 mm) in HMX identical to that for (7). Since a greater mass of neoprene was traversed in (8) than in (7) before DDT occurred, and thus the DDT distance might have been expected to increase, the agreement points to some error in reproducibility of DDT distance, which is estimated to be 2 or 3 mm.

Experiment (9) (Fig. 9), in which the initial increment was doubled to 440 mg HMX, gave a shorter run to DDT -- 52 mm; however, this run was also

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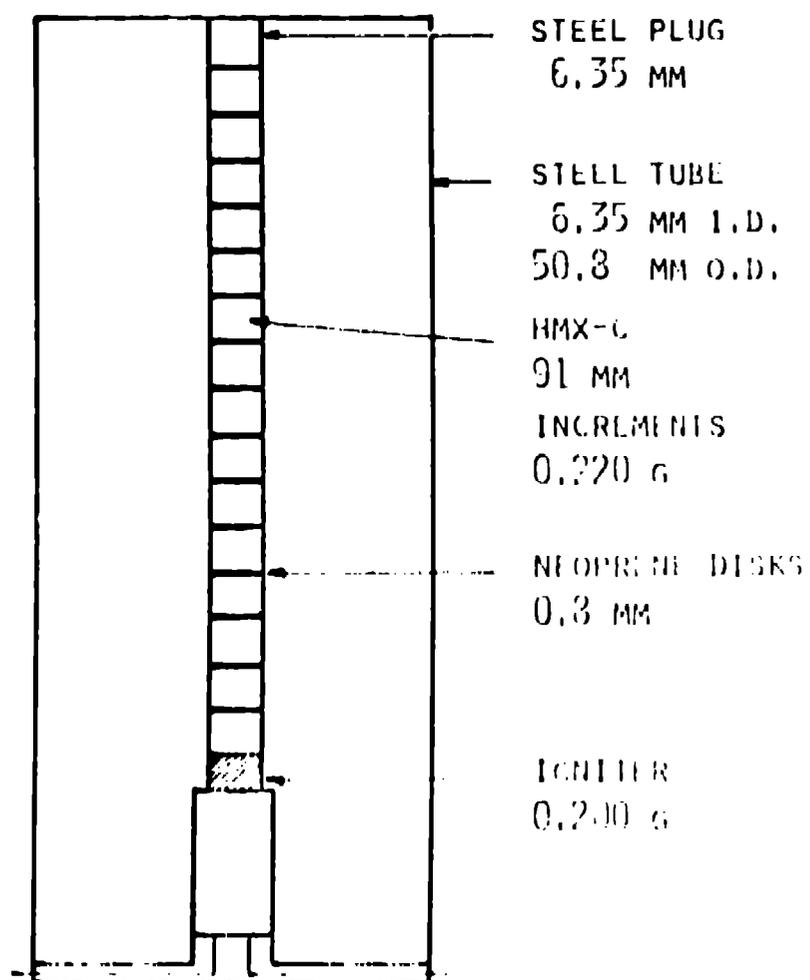


Fig. 7. Experiment 7.

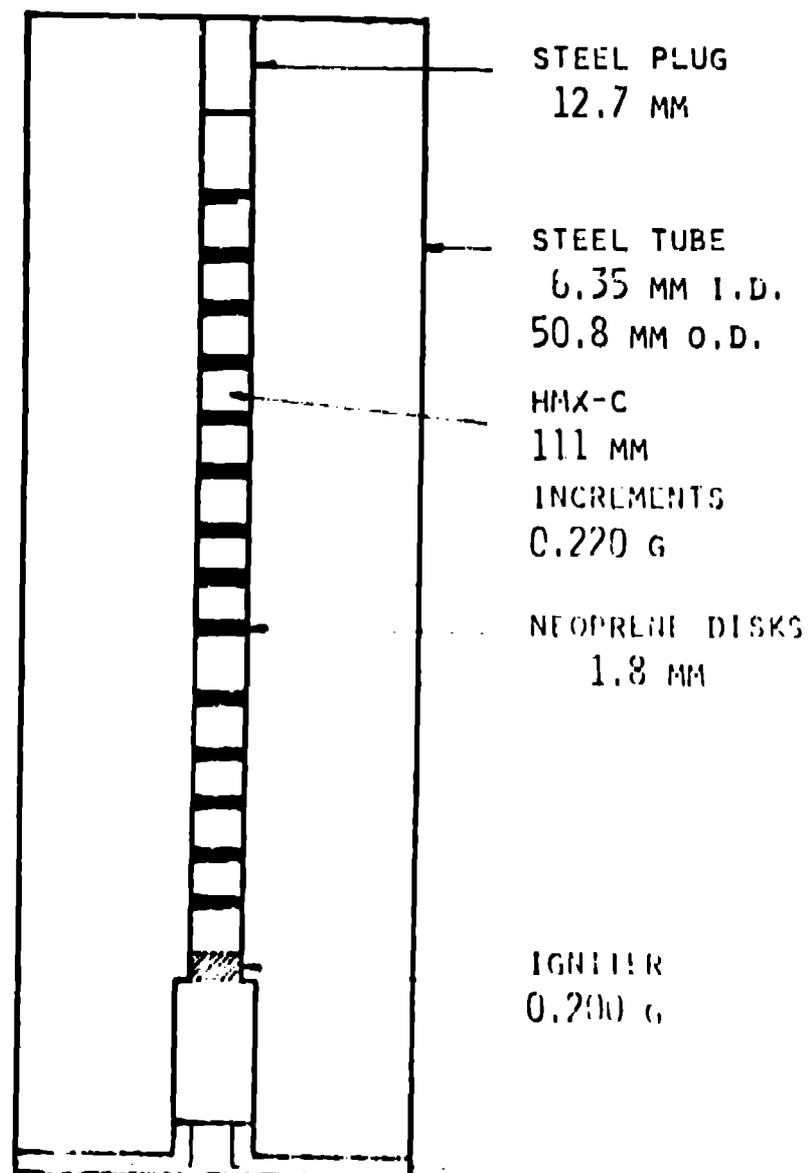


Fig. 8. Experiment 2: HMX-E; Experiment 3: HMX-C.

The irregularities in increments as drawn reflect the irregularities observed during loading. It is believed that when tamping was completed, the increments were more uniform.

6-5015

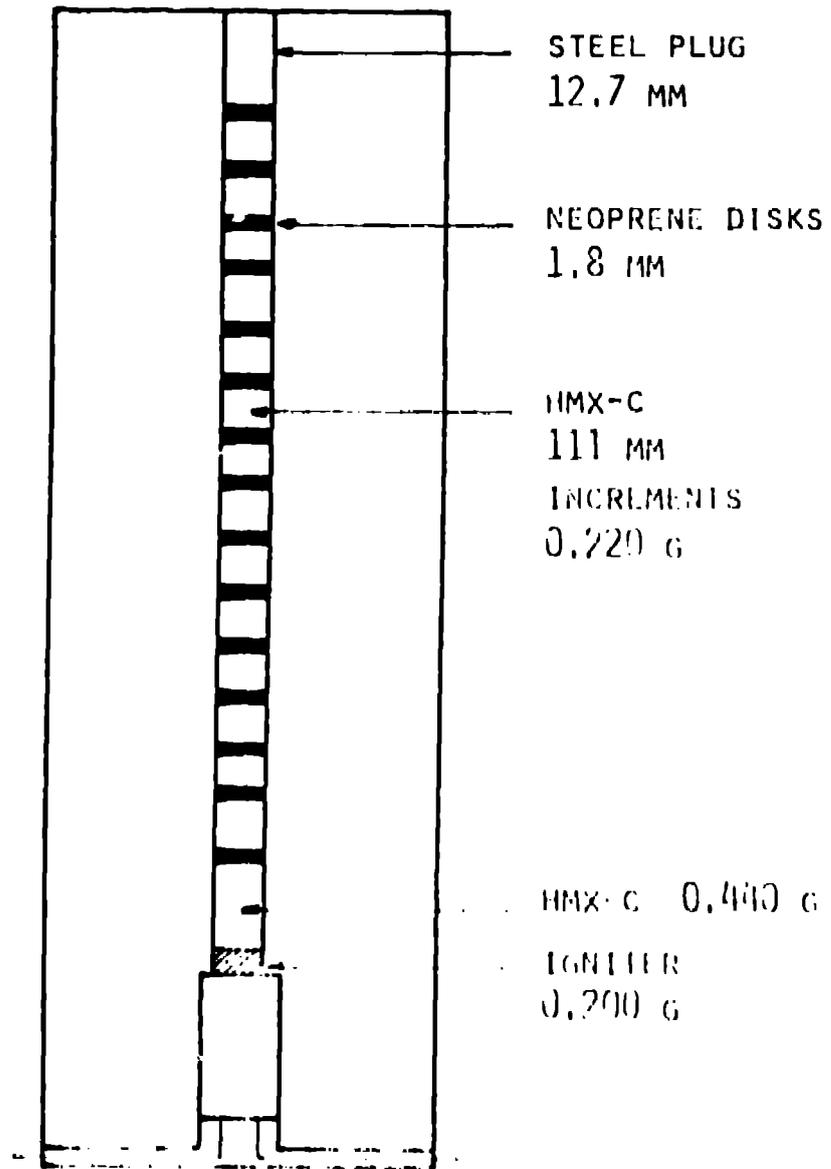


Fig. 9. Experiment 9.

shorter than the result for (6)(58 mm), where no diaphragms were employed. Comparing this result with the finding of Griffith and Grocock that venting of their tubes caused an increase in the DDT distance, one is led to the conjecture that the first diaphragm prevents the escape of combustion products from the combustion zone and thus increases the rate of pressure rise and its attendant effect on the burning rate in the combustion zone.

Experiment 10. An illustration of the effect of reduced rate of pressure rise in the combustion zone is offered in this experiment (Fig. 10). A small brass tube was inserted into the steel tube and was loaded with 220 mg of HMX at a loading density of half that of bulk density -- 0.6 g/cm^3 . The DDT distance, measured from the end of the combustion chamber, was increased to 106 mm. See Fig. 11.

Experiments 5, 11, 12, 13. In these four experiments the effect of varying the specific surface of the HMX in the combustion zone was explored (Fig. 12). In (11), (12), and (13) HMX-C, HMX-A, and HMX-F were used as combustion increments to drive DDT in HMX-C. The results show that as the specific surface area of the burning HMX was increased, and, presumably, the rate of pressure rise was increased, the DDT distance was shortened (Fig. 13). Experiment (11) corresponds to (6) with one added diaphragm. Even though the added diaphragm in (11) may limit the burning mass of HMX to less than, or no more than, the mass involved in burning in (6), the DDT distance is decreased. This result again supports the conclusion that escape of combustion products from the combustion zone is an important factor.

In experiment (5), 220 mg of HMX C were burned to drive a column of HMX-A. In spite of the beneficial effect of the diaphragm (just discussed) in accelerating combustion, the DDT distance relative to that of (3) was more than

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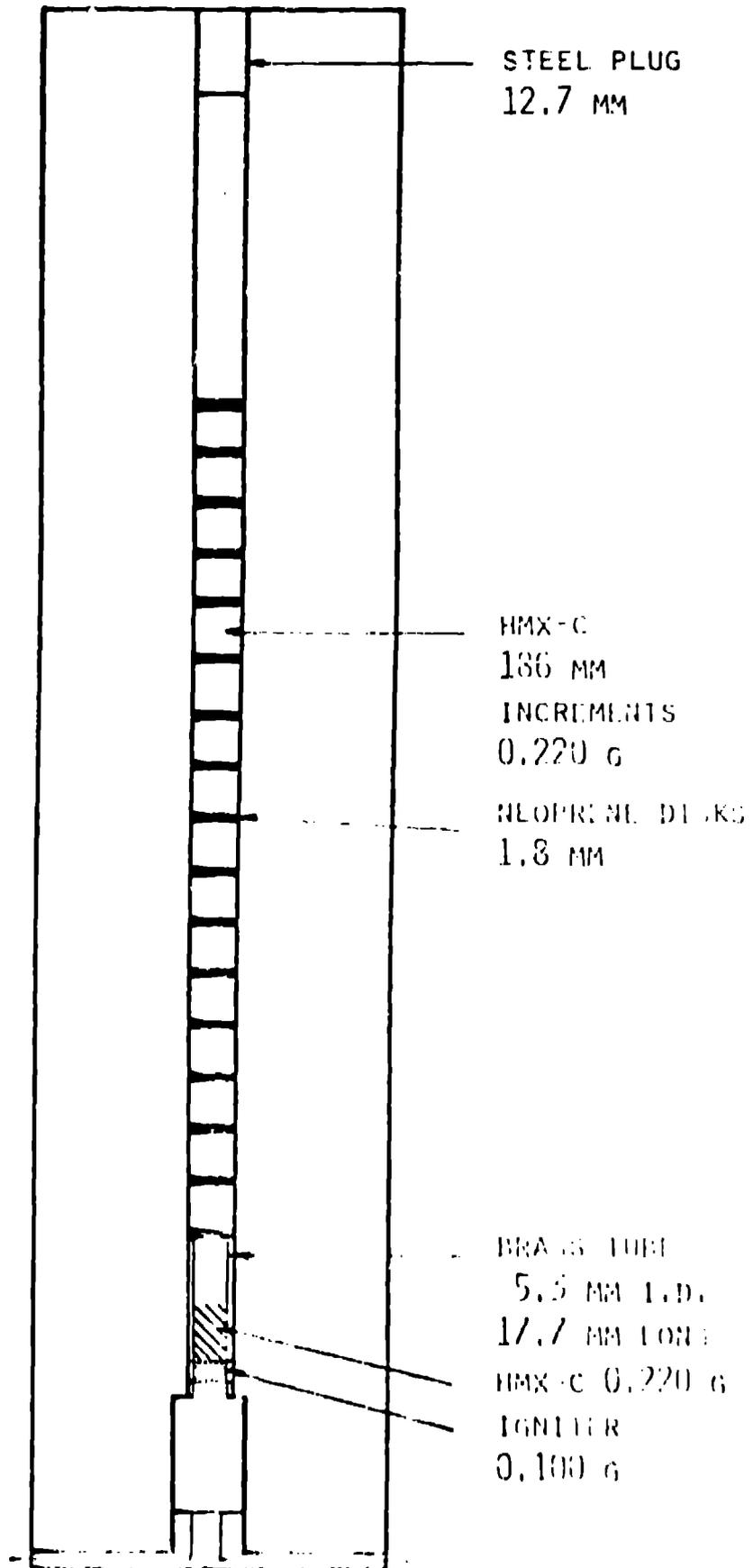
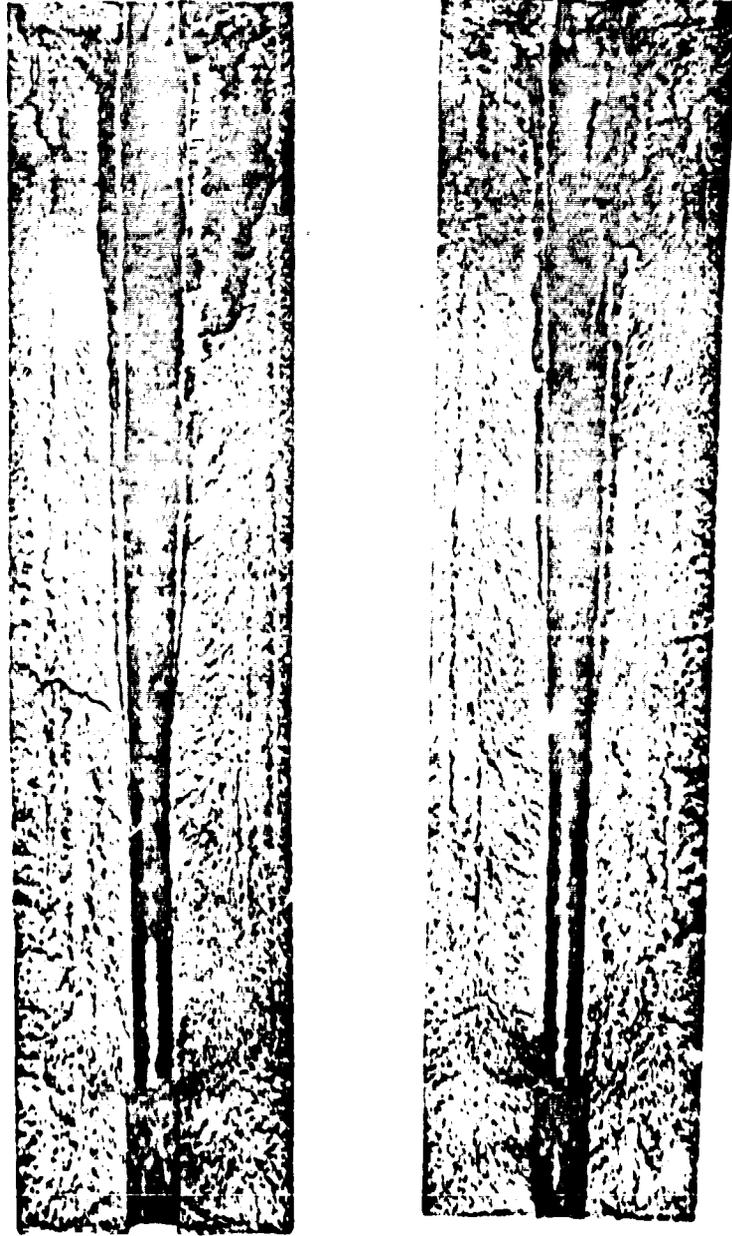


Fig. 10. Experiment 10.



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Fig. 11

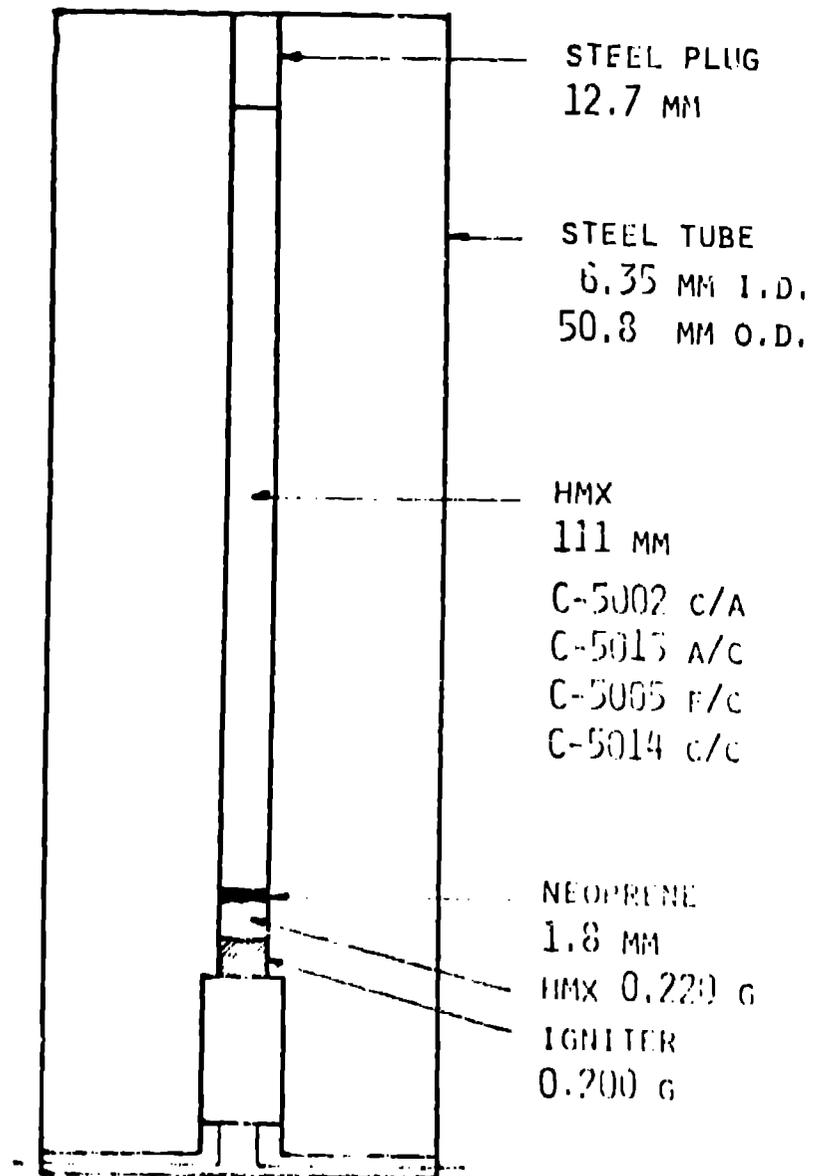
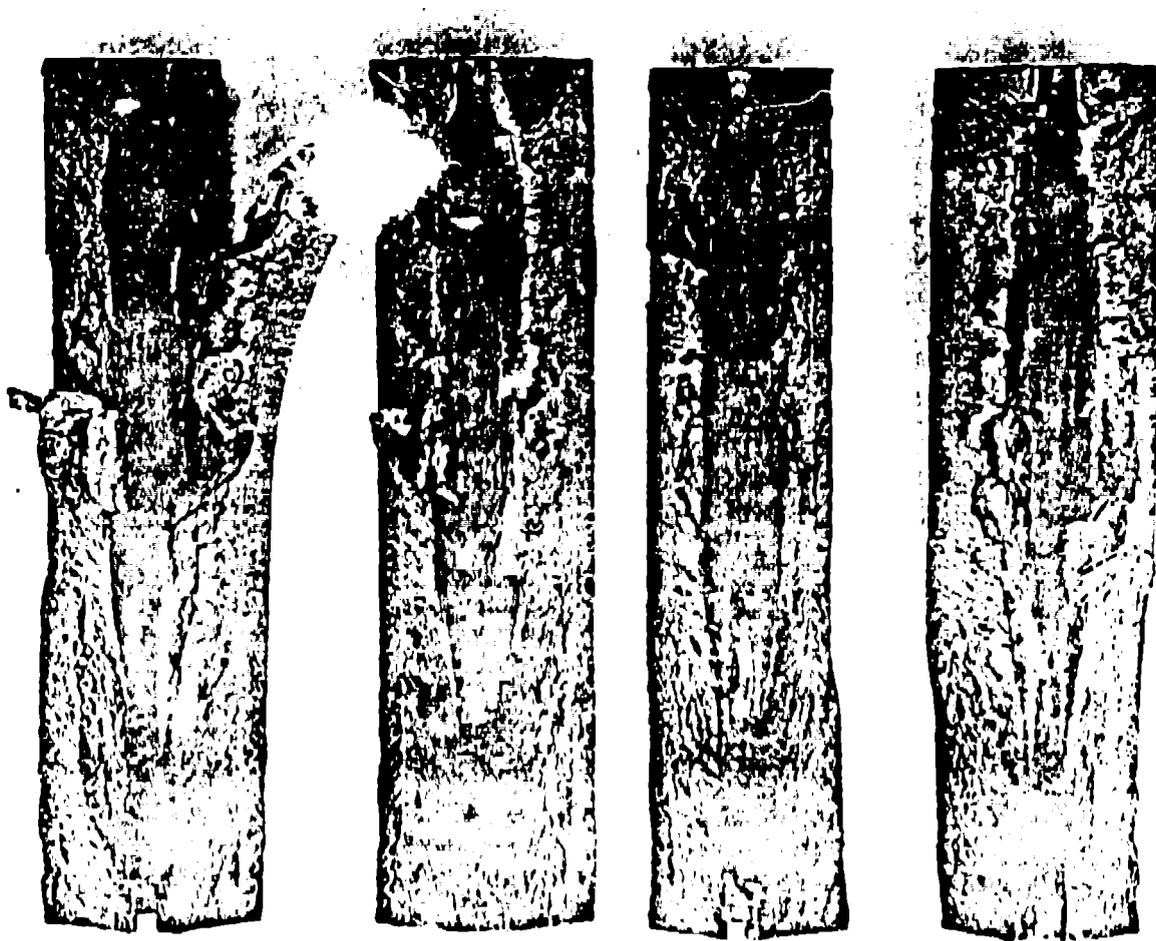


Fig. 12. Experiments 5, 11, 12, 13.



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C-5005

Fig. 13

doubled. This result is attributed to the decreased specific surface area of HMX-C relative to that of HMX-A and the slower pressure rise during combustion.

Discussion

The Combustion Zone

Combustion begins as a conductive process. As the pressure rises it passes into convective combustion, which more rapidly ignites additional explosive. Experiments (5), (11), (12), and (13) show that the rate of pressure rise is important; any leakage of combustion products from the combustion zone slows the rate of pressure rise and increases the distance to detonation. The diaphragms keep the products in place and thus cause the distance to detonation to decrease. Griffiths and Grocock also found that venting of their tubes resulted in longer runs to detonation than when the tubes were closed.

Convective combustion of explosives over long distances is not necessary or important for DDT, as is shown by the experiments with diaphragms positioned every 7 mm. Gases could not pass the diaphragms to propagate convective combustion beyond them.

The rapid wave observed by Price and Bernecker may originate in the combustion zone. It might be driven by a gassy igniter, or it might be due to reaction of pyrolysis products.⁹ The origin of the wave has not been determined, in part because ionization signals from any waves in the initial part of the DDT process have been very difficult to obtain, and when obtained, have been difficult to interpret, as evidenced in the work of Price and Bernecker, Griffiths and Grocock, and the present author.

Fine explosive, having greater surface area, burns more rapidly than does coarse explosive and is more effective when it is used in the combustion zone. This behavior leads to the decrease in transition distance as one proceeds

from the right-hand edge of Fig. 1 toward the minimum. The upturn at the left is caused by behavior in the build-up zone to be discussed below.

Dark Zone

The combustion zone is often terminated by a region which emits little or no light. Brandon, in early work with M-7 propellant, and Griffiths and Grocock with high explosive, have observed the dark region. Sometimes retonation has been seen to propagate backward through it while the detonation goes forward. In M-7 propellant, retonation without simultaneous forward-going detonation has also been observed.

Figure 14 is an example of the occurrence of the dark zone in M-7 propellant. Viewing the record with time horizontal, one sees the record of light from the burning starting at early time at the lower left and progressing slowly upward. The light has a fuzzy outline because the propellant is granulated. Abruptly, the light terminates at its leading (upper) edge and a dark region spreads both forward and backward. After a lapse of time a luminous wave arises at some distance from the beginning of the dark region and proceeds with an initial velocity much higher than that of the burning wave before the onset of the dark zone. The new luminous wave accelerates and becomes a detonation wave.

This dark zone is interpreted as a region of bed compaction. As the pressure rises in the combustion zone, a critical pressure is reached at which the granular bed of explosive begins to collapse and to restrict the further flow of cooled combustion products preceeding the flame front. At first the mass velocity and rate of compaction are so small that no burning is ignited by the compaction, and a plug of unreacting explosive is formed. As burning continues in the combustion zone the rising pressure accelerates the plug and the plug

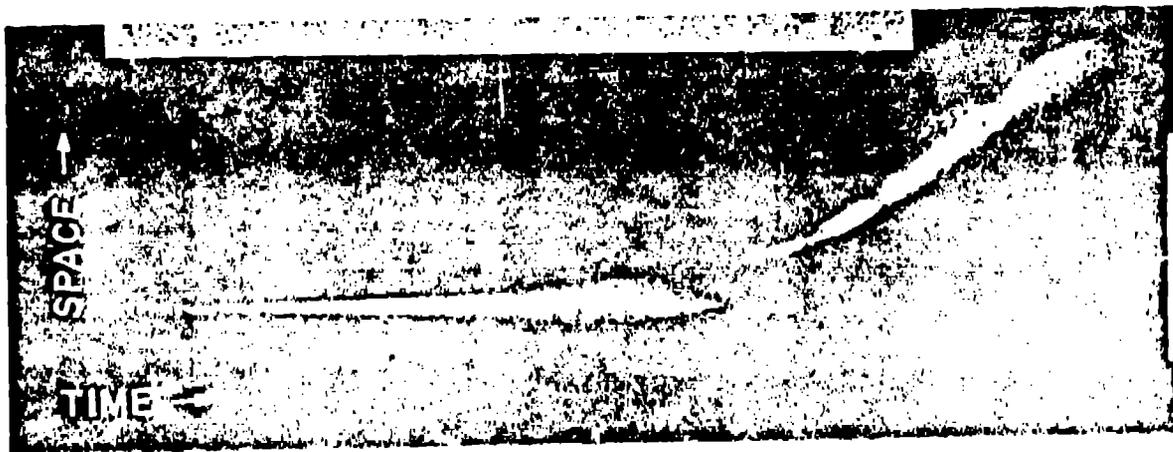


Fig. 14. Streak trace showing dark zone in deflagration-to-detonation transition in M-7 in a 5/16-in. glass tube. Sample contained 10% NaCl. Vertical distance ca. in. Horizontal space ca. 110 μ s. (Ref. 8.),

grows at the front at an increasing rate. When compaction of the explosive at the front of the plug becomes sufficiently rapid, friction between the grains and adiabatic shear within the grains reignite burning at the front of the plug. This burning develops into a detonation.

Burning may also be ignited by friction between the plug and the confining wall. When this happens, photographic observation of the dark zone is impossible. This burning may produce random ionization signals, but however confusing to observation, it is inconsequential for the events leading to detonation, because of the small surface area involved.

In experiment (10), Fig. 11, and experiment (1), Fig. 13, long regions of little or no expansion of the internal bore of the steel tubes indicate the occurrence of plugs. If the pressure in the combustion zone rises rapidly enough, the plug may be very short or may not occur at all. It is not evident in experiment (13), Fig. 13.

The Buildup to Detonation

Reignition of burning beyond the dark zone occurs when the rate of compaction becomes high enough. Intercrystalline friction and adiabatic shear are two heating mechanisms which are important in this region. Adiabatic shear has long been important in the study of the behavior of metals¹⁰ and has become of interest for explosives in recent years.^{11,12} In this form of shear, occurring within the crystallites, the rate of energy deposition in the shear zone greatly exceeds the energy loss by conduction. Thermal softening displaces work hardening as the dominant change in local strength. Continued stress causes continued slip with further local energy deposition and attendant temperature rise. Thus, the process of local heating continues and ignition may ensue. Adiabatic shear is of particular interest because it increases the burning surface and thus is a mechanism for increasing the mass burning rate.

There is as yet little evidence for the effect of adiabatic shear in the explosive literature, but an implication of its effect occurs in the work of the Cavendish Laboratory.¹³ Lead azide was initiated by aluminum spheres $200 \pm 30 \mu\text{m}$ in diameter impacting at a velocity of 200 m/s. Because the energy delivered in the impact was insufficient to cause significant bulk heating of the lead azide, persuasive arguments point to adiabatic shear, occurring during rapid plastic deformation of the explosive at the point of impact, as the most plausible cause. This result is the more dramatic when, in a drop-weight impact test where the energy delivered to a sample of lead azide was orders of magnitude greater, the lead azide failed to react. Because energy was delivered slowly by the drop weight, apparently adiabatic shear was absent.

The occurrence of adiabatic shear in the buildup process offers an explanation of the minimum in the DDT distance in Fig. 1. Two opposing processes are active in determining the DDT distance. One is the rate of pressure rise in the combustion zone, and this is aided by large specific surface or small average particle size; the second process is the ignition in the buildup region by mechanical action, and this is favored by large particle size. The greater sensitivity of coarse-grained explosive to marginal mechanical stimulus has been pointed out by Scott¹⁴ and Dinegar et al.¹⁵ This increase in sensitivity with grain size must be due to the longer shear path in larger crystallites.

When the buildup reaction begins, it may be a plastic wave moving at less than local sound velocity. Gipson and Macok¹⁶ observed reactive waves in cast DINA moving at less than sound velocity and Price and Weber¹⁷ reported similar phenomena in cast pentolite. Obmenin et al.¹⁸ studied low velocity "detonation" waves in pressed PUIN at 92% of theoretically maximum density. When reaction was initiated by flame, it soon propagated stably at velocities

as low as 1.3 mm/ μ s in steel tubes. Stable velocities both below and above that of sound in either the explosive or the steel confinement were observed. The value of the velocity obtained depended on the degree of confinement; heavy confinement always resulted in high-order detonation. Pressures were measured with quartz gauges in the wall of the confining tube, and it appeared that stable low-velocity detonation could be obtained below about 10 kbar. Ermolaev et al. and Sulimov et al.¹⁹ have attempted theoretical treatments of these results.

Thus, although the buildup process may start as a subsonic, plastic wave with sufficient confinement, energy loss is reduced so that pressure in the reaction region rises to some critical value, as observed by Obmenin et al., and the continued pressure rise is then rapid until the DDT process is complete.

Conclusion

The picture of the DDT process as presented here results from an attempt to incorporate common experimental observations which have heretofore been puzzling. It differs from that presented by Kistiakowsky in that the role of convective combustion is terminated and mechanical processes are postulated as the means of continuing the reaction buildup until shock waves are formed. In order to validate this picture it will be necessary both to review the experimental literature for observations which may not be reconcilable with it, and to subject each step in the proposed DDT process to detailed scrutiny.

Acknowledgement

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