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P. E. Luebcke, P. M. Dickson and J. E. Field

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An experimental study of the deflagration-to-detonation transition in granular secondary explosives

By P. E. Luebcke, P. M. Dickson, and J. E. Field

Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, U.K.

The deflagration-to-detonation transition (DDT) has been studied in prepared columns of granular secondary explosive. The secondary explosives 2-(5-cyanotetrazolato) pentammine cobalt (III) perchlorate (CP) and pentaerythritol tetranitrate (PETN) were chosen for the study due to their known propensity to undergo DDT within a few millimetres of ignition. Confinement of CP columns within polycarbonate and PETN within metallic confinement fitted with slit windows allowed direct high-speed streak photography of the events. Deflagration and detonation velocities and the run-to-detonation lengths were measured as a function of charge pressed density. Ignition of the explosive column was attained thermally through a copper barrier with a gasless pyrotechnic. Deflagration and detonation velocities were seen to depend strongly upon pressed density with both explosives. There appeared to be a maximum density conducive to DDT with both explosives but no minimum with CP.

Studies of DDT continue to have interest for the safe storage and use of reactive materials, and for the development of a detonator based on a secondary explosive.

1. Introduction

The mechanism by which a deflagration makes the transition to a detonation is not yet fully understood. The salient features observed in the DDT of a granular secondary explosive are an accelerating combustion wave which moves through the unreacted charge, followed by the onset of detonation at some point downstream from the combustion wave. A retonation wave can sometimes be seen to run upstream from the point of initiation. Central to this process is the formation of the shock waves which ultimately lead to detonation. This final shock-to-detonation transition is fairly well understood; it is the mechanism by which sufficiently strong shock waves are first formed which is less clear.

The phenomenon of DDT has been investigated for many decades since Dixon (1903) and Bone *et al.* (1936) examined the transition to detonation in explosive gas mixtures. Pioneering work by Kistiakowsky (1948) resulted in the hypothesis that upon ignition of an explosive, convective burning would govern the combustion process until initiation of detonation. Convective burning is the process whereby flame spread results from the convective heat transfer by hot reaction gases which flow into the porous unreacted charge ahead of the ignition front. The

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velocity of convective combustion is typically two to three orders of magnitude greater than layer-by-layer conductive burning. Griffiths & Groocock (1960) suggested that detonation was brought about by an initial conductive burn followed by an accelerating convective front producing the precursor shock necessary for initiation of detonation. Gipson & Macek (1962) also postulated that compression waves of increasing strength were produced emanating from the burning zone. These waves coalesce at a point downstream, forming a shock which generates a rapid increase in temperature, leading to chemical reaction and the initiation of detonation. Results produced by Bernecker & Price (1974) from studies of the DDT in RDX/wax compositions, indicate that the compressive waves do not originate from the convective front but are caused by rapid pressure build-up from a region near the point of ignition.

Campbell (1980) tested the importance of convective burning in the run-up to detonation in studies of granular HMX, in which gas-impermeable disks were placed in the column of explosive to prevent hot gaseous products moving ahead of the combustion wave. By this means he showed that convective combustion was only important during the initial stages of deflagration. As the pressure rises, intrusion of hot gases into the porous bed becomes increasingly impeded due to compaction of the material. Eventually the bed compacts to such an extent that an impermeable plug is formed. This plug is propelled downstream by the gaseous reaction products forming a compaction wave at the leading edge of the plug. The compaction wave increases in strength as higher pressure stress waves originating from the accelerating combustion wave overtake it.

This experimental work was continued by McAfee *et al.* (1993) who ignited beds of HMX using a gasless pyrotechnic. This led to the formulation of a more detailed description of the DDT process which also seems consistent with previous observations. A representation of this model can be seen in figure 1. In this model, the pressure rise at the initial deflagration front sends a compaction wave into the unreacted material initiating reaction by friction and shear between explosive grains. The combustion products do not readily permeate the compacted material and so a well-defined deflagration wave follows the compaction wave, accelerating as the pressure rises with the extent of reaction. Pressurization by reaction product gases along with the compaction of the bed leads to the formation of the plug. The downstream boundary of this fully compacted region is a shock wave supported by the upstream boundary which behaves like a piston. This shock wave accelerates slowly until the deflagration wave overtakes the upstream boundary, which then accelerates the piston causing more rapid acceleration of the shock wave until it exceeds the critical shock initiation pressure of the compacted explosive. This model is consistent with the observed data and differs from most previous models in that convective combustion plays little part in the process, other than affecting the rate of reaction and hence the pressure rise in the initial stages.

Much work has been carried out on the possibility of replacing primary explosives in detonators by CP (Leslie *et al.* 1976; Ewick 1982; Lieberman 1984; Lieberman *et al.* 1984, 1986; Kopczewski & Weinmaster 1976). Leslie *et al.* (1976) found that CP will undergo DDT following low-voltage hot-wire ignition, and that in terms of safety it could replace materials such as lead azide. The recognition of CP as a potential replacement for primary explosives has led to the investigation of the burning characteristics of related cobalt coordination compounds. Lieber-

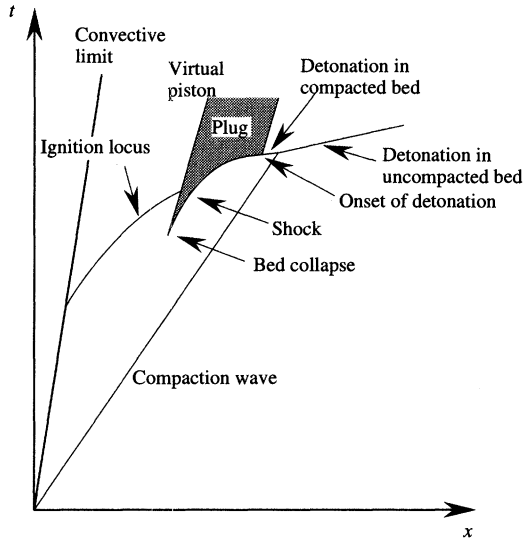

Figure 1. Graphical representation of the DDT model proposed by McAfee *et al.* (1993).

man & Fleming (1986) compared the performances of 15 complexes, mainly amine ligand coordinated with perchlorate anions, by loading them into a standard detonator and measuring the projection velocity of the end plate. The velocities were measured by using the velocity interferometer system for any reflector (see Barker & Hollenbach 1965), these data being supplemented with data from X-ray radiographs of the fired charge holders. The results indicated that although a good oxygen balance in the complex was important to achieve the velocity jumps associated with detonation, it was the presence of a trigger group such as an azido or tetrazolato ligand which was the greatest determining factor affecting the detonability of the complexes. Unfortunately, due to the toxicity of CP, its production has been discontinued.

PETN has also been shown to be relatively shock sensitive (Griffiths & Groocock 1960; Dinegar 1978, 1979, 1981, 1983) with run-to-detonation lengths less than the experimental dimensions used. Due to its lower sensitivity, stronger confinement is required than for CP.

The objective of this experimental investigation is to conduct laboratory-scale DDT tests on these two relatively shock sensitive explosives in order to compare the observed mechanisms with current models such as that of McAfee *et al.* (1993). DDT mechanisms which have been experimentally elucidated can then be used in theoretical modelling.

2. Experimental

The granulated explosives chosen for this study were 2-(5-cyanotetrazolato) pentammine cobalt (III) perchlorate (CP) and pentaerythritol tetranitrate (PETN). CP, a yellow free-flowing crystalline material with crystal density 1.97 g cm^{-3}, has been shown by Baer *et al.* (1986) & Stanton *et al.* (1981) to be highly reactive with growth to detonation occurring after only a few millimetres from the point

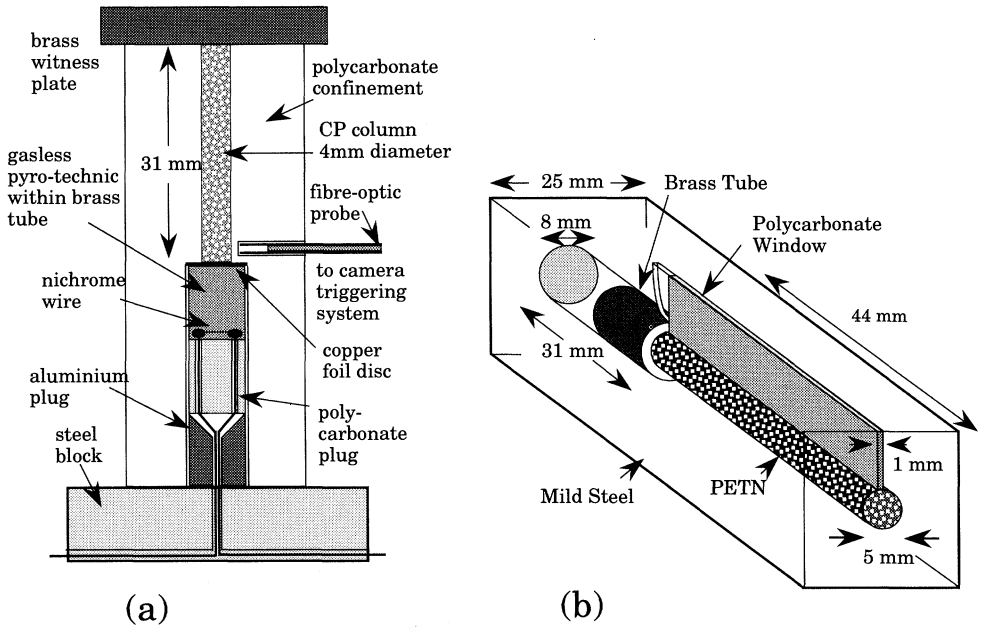

Figure 2. (*a*) CP and (*b*) PETN confining arrangements.

of ignition. This may be due to it having a low shock initiation pressure. CP is thus ideally suited to small-scale laboratory investigation of DDT. Baer *et al.* (1986) reported the average particle diameter of their batch of CP, the same used in our study, to be of the order of 140 μm, but that under the action of pressing the particles easily fragment, resulting in an average particle size of 10–15 μm. The chemical structure of CP is more closely related to that of many inorganic primary explosives, such as mercuric-5-nitrotetrazole, than to most secondary explosives which are generally organic CHNO compounds.

High-speed photography was used to record the reaction of thermally ignited columns of CP and PETN. Thick walled metal confinement is normally used for DDT studies, which does not permit direct photographic study, although some workers (Demissy & Michot 1988; Dickson *et al.* 1991) have used fibre-optic probes to follow the reaction zone in beds of energetic material. The confining material chosen for CP was polycarbonate, which, although having a lower yield strength than metals, was found to be strong enough to permit DDT to occur in CP and thus gave the advantage of allowing direct visual observation of the reaction along the entire column length. However, polycarbonate does not provide sufficient confinement for PETN to undergo DDT. Therefore a steel confinement system fitted with a polycarbonate slit window was developed for PETN charges, which does allow direct and continuous high-speed streak photography.

Figure 2 shows the construction of the test-pieces. A fibre-optic probe was positioned at the ignition end of the explosive column so that light emitted from the initial stages of deflagration could be used to trigger the camera. The acceptance angle of the fibre-optic was reduced (to prevent premature triggering) by lengthening the fibre sleeving at the end of the fibre.

The charge was prepared by pressing the explosive incrementally, the height of

each increment being less than the diameter of the explosive channel in order to ensure a consistent pressed density. Stepped density columns were also prepared in order to investigate the effect on run-to-detonation lengths when the deflagration moves from a higher density to a lower density pressing. It was thought that the production of a shock wave at the density discontinuity might result in a reduction of the run-to-detonation length of the lower density section.

Ignition of the explosive column was attained thermally with an electrically ignited, virtually gasless pyrotechnic mixture of boron and potassium dichromate in a finely divided state. This minimized any pre-pressurization of the explosive column which would complicate the interpretation of the initial explosive deflagration. Emission of light from the pyrotechnic, which might prematurely trigger the camera in the polycarbonate-only test-pieces, was prevented by pressing the pyrotechnic into a brass sleeve and separating it from the explosive by a 25 μm thick copper disk. At the end of the explosive column, a brass witness plate was positioned to give an indication of the occurrence of detonation. The test-piece was clamped between two heavy steel blocks by using connecting bolts to prevent rear-venting. The test-piece was then placed inside a vented firing-box fitted with polycarbonate windows to allow photography whilst protecting the camera and surroundings from fragments. The reaction in the explosive was recorded using a Hadland Image Converter camera (Imacon 790) operating in streak mode.

3. Results

The CP was examined by scanning electron microscopy to determine its particle size distribution. The particles, which ranged in size from 20 to 200 μm, appeared to consist of loosely bonded clusters of smaller octahedral crystals in the range 5–20 μm. Examination of pressed samples showed that these clusters usually disintegrate during the pressing process.

The high-speed streak photographs of the progress of the reaction front through CP and PETN were scanned and digitized to facilitate accurate measurement of deflagration and detonation velocities and run-to-detonation lengths. The digitized records of the position of the reaction front with time were also differentiated to give velocity-time profiles.

The deflagration velocity just before DDT, or in the case of those samples which did not undergo DDT, the velocity at the end of the column, generally increases with pressed density. The detonation velocities are in broad agreement with values found by earlier workers (Stanton *et al.* 1981; Baer *et al.* 1986) show a linear relationship with pressed density. The detonation velocities above about 75% theoretical maximum density (TMD) were obtained by allowing DDT in a lower density column and then letting the detonation run into a higher density pressing.

Figure 3 is a plot of run-to-detonation length against %TMD, which indicates that this rises with pressed density up to the point at which it exceeds the length of the columns used in these tests. The errors in the density and run-to-detonation length were estimated to be 6% and 1 mm respectively. It differs from the commonly observed 'U-shaped' plots of many other secondary explosives in that there is no evidence of the run-to-detonation length increasing at lower densities. Run-to-detonation lengths for the stepped density columns were not significantly dif-

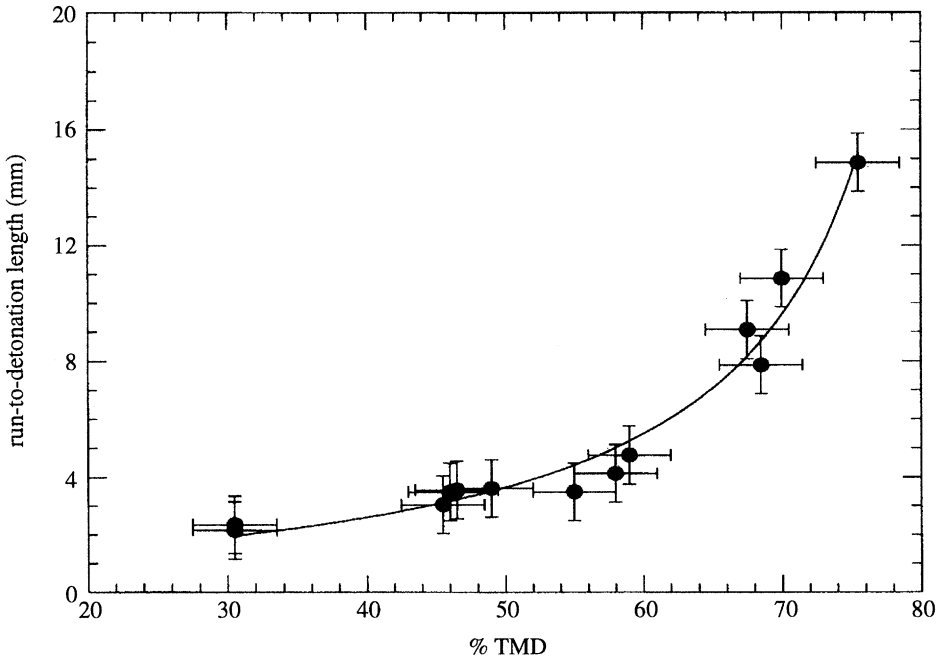

Figure 3. Run-to-detonation length versus %TMD for CP.

ferent to those for uniform pressings, indicating that shock wave formation at density discontinuities have little effect on the DDT process in this situation.

Figure 4 shows DDT in a column pressed to 67% TMD which exhibits a run-to-detonation length of 12 mm and an accelerating deflagration which increases to 1.5 mm μs^{-1} immediately before DDT. In a column pressed to 58% TMD there is a 6 mm run-to-detonation length and an apparently constant deflagration velocity of 1.1 mm μs^{-1} which increases to 1.5 mm μs^{-1} immediately before DDT.

At DDT, the change in velocity appears to be discontinuous, with an initial detonation velocity of 6.0 to 6.5 mm μs^{-1} which is maintained for 2 to 3 mm, before decreasing to a stable value of 5.5 to 6.0 mm μs^{-1}. A low velocity shock wave is visible moving upstream from the point of DDT at a velocity of about 1.7 mm μs^{-1}. This velocity does not appear to depend on either the velocity of the preceding deflagration or the subsequent detonation.

Occasionally a near-vertical line appears on the streak record (labelled in figure 4) at the time when the detonation reaches the witness plate. This is thought to be due to adiabatic compression of the small air-space at the end of the column causing light emission which is then scattered by the decomposition products throughout the column.

At higher densities, columns of CP frequently exhibited long induction times from ignition of the end of the column to propagation of a deflagration wave. In many of the streak records of reaction in high density CP columns, an interesting observation was that two light-emitting waves are present, the second wave initially travelling 0.3 mm μs^{-1} slower than the first. Figure 5 is an example of a streak record exhibiting this phenomenon. Note the first wave is usually fainter

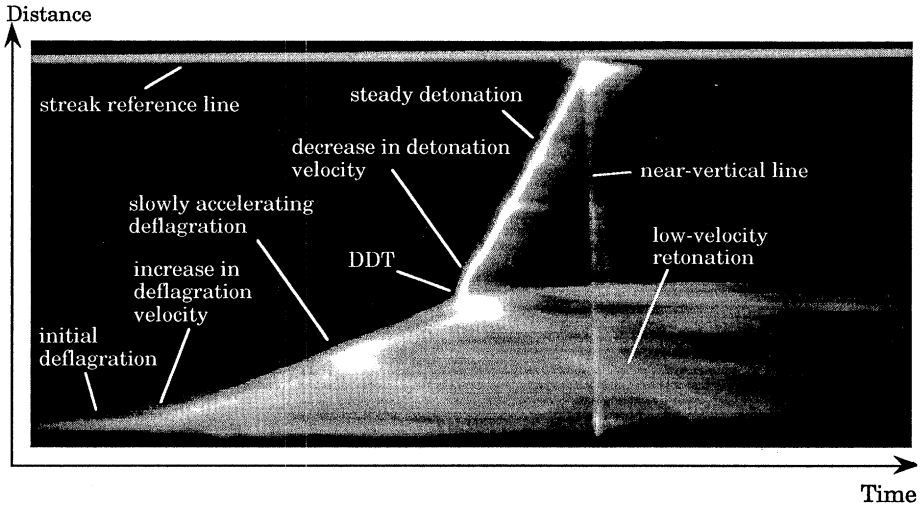

Figure 4. Streak record of DDT in 67% TMD CP.

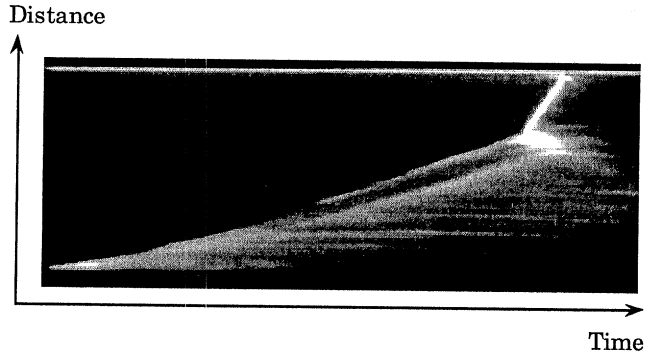

Figure 5. Streak record of DDT in 72.5% TMD CP exhibiting 'double-wave' deflagration.

and that no horizontal striations occur in the dark space between the waves. Striated light emission which is particularly clear in figure 4, occurs after the second wave. In the deflagration regime there are many striations whereas in the detonation regime the striations are less frequent, more regular, sharper and have shorter duration.

Some streak records, particularly of samples pressed to higher densities, show an increase in light output just before DDT, continuing for 2–3 μs until the transition. In columns where no DDT occurs, the reaction wave accelerates down the column without this emission of light.

Preliminary results obtained from PETN columns within steel confinements also show accelerating deflagrations, but with a notable absence of a second light-emitting wave as seen with CP. Some streak records also exhibit what appears to be a break-out of detonation at a point downstream from the combustion front. The streak records of CP undergoing DDT did not show this phenomenon, with the point of DDT coinciding with the front of the deflagration wave.

Retonation waves are clearly seen emanating from the point of DDT. The initial retonation velocity is faster than its subsequent velocity as the wave starts in

compacted and unreacted explosive before passing into a region of incompletely reacted explosive and product gases. Detonation velocities and run-to-detonation lengths, as with CP, are seen to depend upon the pressed density of the charge.

4. Discussion

CP will undergo DDT using the confinement dimensions used in this work only up to *ca.* 75% TMD. At densities greater than this, it is possible that DDT may occur in longer columns. However, the relatively slow attainment of the critical shock initiation pressure could allow the polycarbonate confinement to fail, making DDT unobtainable in columns over a certain density.

Three burning stages, distinguishable by their velocities, can be observed in the streak photographs. A slow initial burning stage is seen, especially at high densities (where long induction times are exhibited) but is seemingly absent in columns of CP pressed to less than about 55% TMD. This could be due to the increased importance of a conductive burning stage at higher densities, where slow, layer by layer burning precedes the faster convective stage caused by intrusion of hot gaseous reaction products.

An increase in burning velocity to 0.6–0.8 mm μs^{-1}, which continues for only a few microseconds, could be attributed to a convective burning stage.

The deflagration wave then increases in velocity to 1.0–1.2 mm μs^{-1} which may be due to compressive burning. In low density pressings, the run-to-detonation length is relatively short and the deflagration velocity appears to be constant. In higher density columns the deflagration accelerates to the point of DDT, or to the end of the charge in those columns not undergoing DDT. There is a pronounced double-wave structure in the streak records of this higher velocity mode of burning. The first wave is followed by a 2–3 mm long region of comparatively lower radiance.

The first of the two deflagration waves could be evidence for a leading compaction front as suggested by Baer *et al.* (1986), which causes adiabatic compression of air-spaces yielding light emission. Alternatively, Ermolaev *et al.* (1988) suggested that it could be a leading combustion wave which results in only partial decomposition of the CP. The striations as described in the results section give clues about possible mechanisms. In the detonation regime the striations appear to be associated with the boundaries between the pressed layers, with higher light output from highly compacted material. The striations following deflagration are on a much finer scale and may be associated with the reaction of individual larger grains. The absence of striations after the first deflagration wave in a two-wave stucture suggests that the first, fainter wave does not involve reaction but only light output from gas-space compression. This would support the Baer hypothesis rather than that of Ermolaev. Further research is needed to clarify the precise mechanisms of the two-stage structure.

The initial detonation velocity observed in the streak photographs is typically 0.5 to 1.0 mm μs^{-1} faster than the subsequent velocity. This supports the model proposed by McAfee *et al.* (1993) in which the detonation wave starts in a precompacted region of the explosive and then slows down when it overtakes the compression wave.

The absence of long run-to-detonation lengths at relatively low densities, which contrasts with the results found by Baer *et al.* (1986) could be due to the dif-

ference in the ignition method. They used a hot-wire igniter whereas the use of a pyrotechnic igniter through a barrier in our study, although virtually gasless, could produce a degree of compaction in the low density pressings.

There is extensive literature on deflagrations and DDT in gases (see Chue *et al.* (1993), and references therein, for recent publications). There is not room to discuss this literature in detail here but in future research we plan to explore the similarities between these two areas.

5. Conclusions

We have successfully attained continuous records of the DDT in laboratory-scale, low-confinement tests. Previous DDT work has typically involved highly confined secondary explosive columns of considerably greater diameter and up to a magnitude greater in length.

The granular explosive CP exhibited DDT in prepared columns confined within polycarbonate. The run-to-detonation lengths were seen to increase appreciably with pressed density. Above 75% TMD, no transitions occurred for the column dimensions used in this work. There appears to be no minimum run-to-detonation length for CP, with loose powder columns undergoing DDT after only a few millimetres. In this respect, CP is unlike many other secondary explosives, where run-to-detonation lengths increase at low, as well as high, densities.

The streak photographs clearly show the presence of two light-emitting waves. Observed features on the streak records provide evidence which suggests that the leading wave is compressive in nature and that little or no decomposition occurs at this stage. The point of DDT coincides with this leading wave and not the second wave which suggests that the strength of the compressive wave increases as it accelerates down the column until the critical shock initiation pressure has been attained.

PETN undergoes DDT in the higher confinement offered by metal test-pieces, having polycarbonate slit windows to allow streak photography. The apparent difference in build-up to DDT could be either due to a different mechanism or to the difference in confining conditions. This is a topic of ongoing research.

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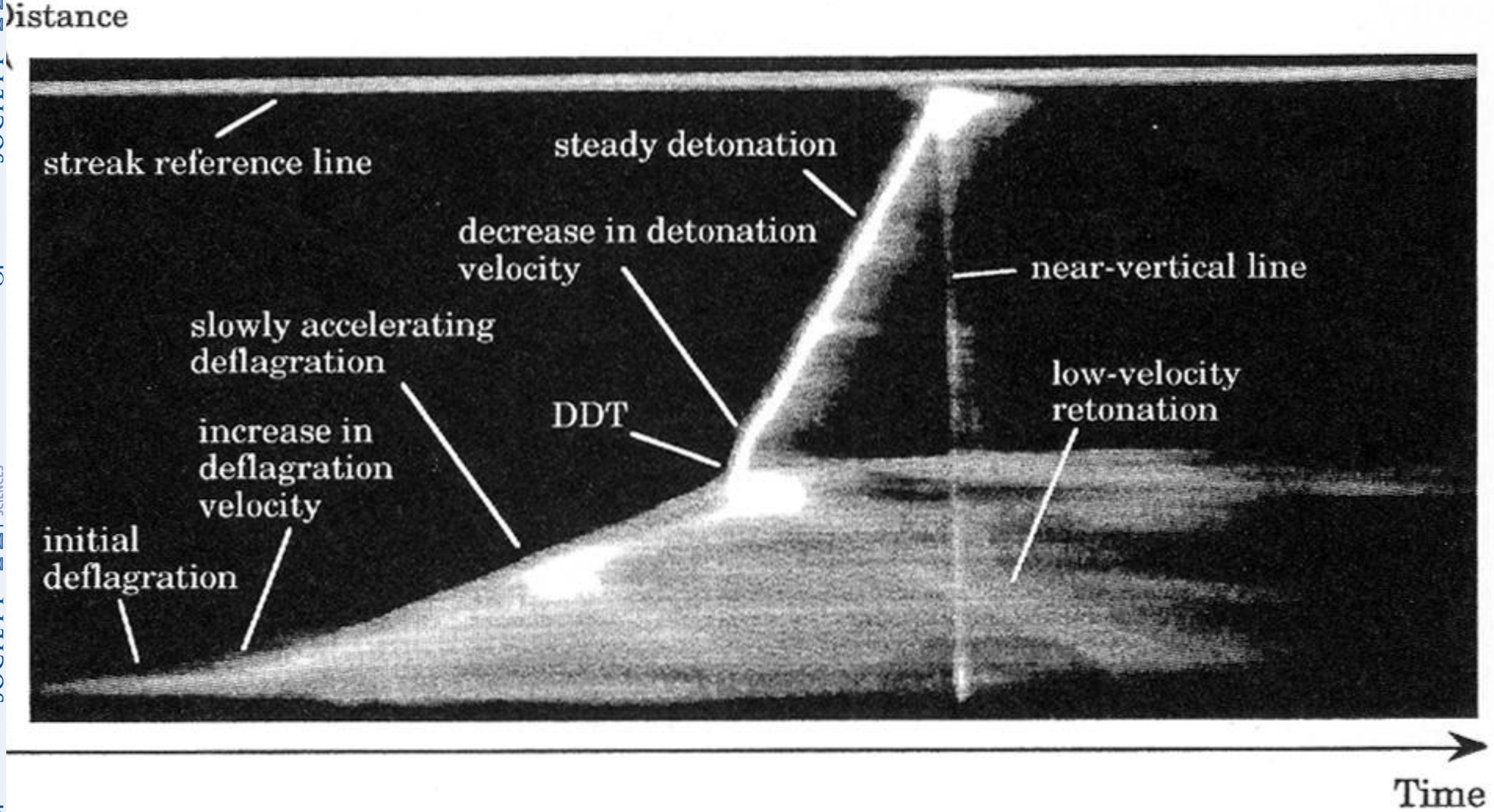
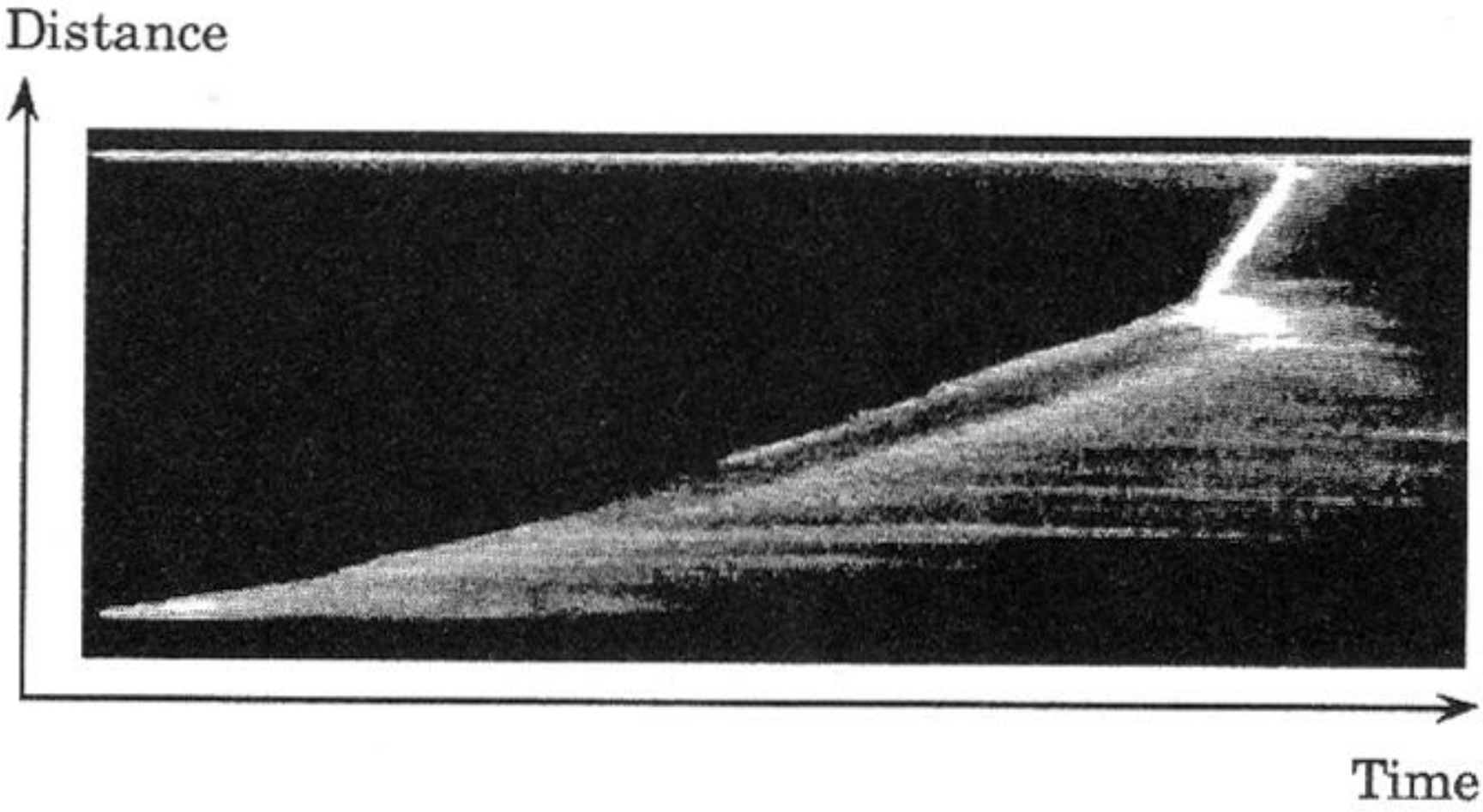


Figure 4. Streak record of DDT in 67% TMD CP.



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