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Title: On The Deflagration to Detonation Transition in 1.65 g cm-3 Pentaerythritol Tetranitrate

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On The Deflagration to Detonation Transition in 1.65 g cm^{-3} Pentaerythritol Tetranitrate

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INTRODUCTION

The LANL Weapons Response Group is charged with making determinations for a wide range of scenarios regarding the probability that inadvertent ignition of the high explosive (HE) components might cause nuclear yield, or dispersal of special nuclear material, during assembly/disassembly operations at the Pantex Plant. These scenarios are categorized based on the nature of the ignition causing stimulus, *i.e.* impact, electrostatic discharge, pressure-shear, etc. However, for all of these scenarios, regardless of how ignition is achieved, there must also be a credible path for the burning HE to transition to detonation for yield or dispersal to occur. If it can be determined that there is no feasible path for deflagration-to-detonation transition (DDT), the scenario space that requires mitigative action is greatly reduced. The strategy to support Weapons Response determinations is to use experimentation and observation to evaluate HE response in severe "overtest" configurations. To increase with confidence in the margins of HE response, these overtests are designed, by controlling parameters known to enhance HE response violence, to be more severe than what is ever encountered in the actual operations environment at the Plant.

This study is focused on DDT in pentaerythritol tetranitrate (PETN), a high explosive that is used in LANL's detonators. PETN is known to be able to DDT at certain densities[1,2], and this represents a safety concern. The PETN in LANL's detonators is commonly a density of 1.65 g cm^{-3}. However, DDT has never been observed at this density in PETN. In this work, we studied the DDT reaction in 1.65 g cm^{-3} PETN to determine whether DDT is possible at this density, and if so, what length of PETN is required for the transition to occur. In any of the tests conducted, we did not observe DDT in 1.65 g cm^{-3} PETN. It is possible that the DDT length for 1.65 g cm^{-3} PETN is greater than 6" (the available run length in the experiments), but this length is simply not available in a detonator, and is therefore not a concern.

BACKGROUND

DDT occurs as a result of mounting pressure produced by evolving product gases from burning explosives that drive compaction waves into the unreacted material[3]. The ability of an explosive to undergo DDT is contingent upon the confining environment being able to contain rising pressures for long enough that compaction waves eventually form a shock discontinuity and shock initiate the HE some distance from the onset of the burn. Therefore, the available length of HE is also a constraining variable for the ability of a particular explosive sample to undergo DDT.

The DDT process in PETN has been studied previously, most notably by Luebcke *et al*[1,2]. They present the characteristic run length to detonation versus initial density for densities ranging from 60% of the theoretical maximum density (TMD, equivalently crystal density, which for PETN is 1.778 g cm^{-3}) up to 88% TMD. The run length curve is reproduced in figure 1.

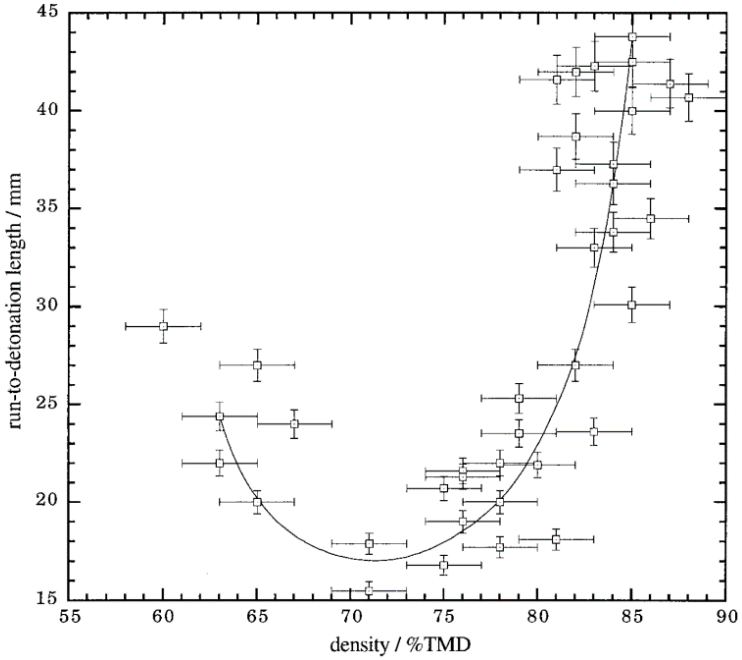

Figure 1. Run length to DDT versus PETN percent theoretical maximum density from Luebcke et al[2]. 73% TMD PETN will undergo DDT in the shortest run length, while 1.65 g cm^{-3} PETN (93% TMD) lies off of the plot.

There is a minimum in the curve described by run-length versus density, and this occurs at roughly 73% TMD, or powder pressed to 1.30 g cm^{-3}, where the characteristic run length is approximately 17 mm. As the density approaches 100% TMD, the run length increases dramatically and appears to approach a vertical asymptote where run lengths effectively go to infinity. Since Luebcke *et al.* only report data for PETN pressed to up to 88% TMD, we cannot say with certainty from their work whether or not PETN at higher densities has the ability to undergo DDT.

The PETN found in LANL's EFI detonators is pressed to 1.65 g cm^{-3} (93% TMD) and are right circular cylinders with dimensions of 7 mm diameter x 1.6 mm tall. It is important to note that 93% TMD lies outside of the density range measured by Luebcke *et al.* It is also important to note that, ignoring considerations for critical diameter, the longest run length available in such a geometry is the diagonal across the center of the pellet, which in this case is just shy of 7.2 mm; less than half the minimum characteristic run length measured by Luebcke. What is more, studies of DDT run length in PETN typically require very strongly confining environments, *e.g.*, thick-walled steel tubes. PETN in detonators are often confined by thin aluminum cups or by plastic headers, providing much less confinement than a thick walled steel tube. Simply put, with our existing knowledge, we can make a convincing argument that there is not enough run length for PETN to be able to undergo DDT in this geometry because it is just too small and too dense.

In this study, we explore the effect of confinement on the 1.65 g/ cm^{-3} PETN to undergo DDT by confining it in a variety of tube materials: steel, sapphire, and polycarbonate tube. The steel and sapphire tubes are meant to over-confine the PETN at least with regard to the confinement provided in an actual detonator, and the polycarbonate tubes are meant to provide a more realistic confining environment closer to what is found in an actual detonator.

EXPERIMENTAL

The DDT tubes used in this study were right circular cylinders with a 7.01 -0.00/+0.05 mm borehole, chosen to match the diameter of the common 7 mm x 1.6 mm PETN pellets in LANL EFIs. DDT tube wall thicknesses varied depending on the specific experiment. Tubes were made of polycarbonate, sapphire, or steel. To monitor the reaction inside of the steel tubes, the tubes featured a slit that ran the length of the tube. A glass or polycarbonate window was glued into the slit to prevent gas venting through the slit during the experiment. The polycarbonate and sapphire tubes are optically clear, and so no slit was required to image or streak the reaction.

Two densities of PETN were examined in this study: (1) detonator density, which were prepared by pressing consolidated pellets to a density of 1.65 g cm^{-3} with dimensions of 7 mm diameter x 7 mm tall, and (2) roughly 73% TMD, which is the density that will undergo DDT in the shortest run length according to research by Luebcke. Pressed pellets come out of the pressing die with low variability in the diameter: 7.00 ± 0.01 mm. For the 73% TMD experiments, PETN is hand pressed into the tubes. The low-density experiments were used as proof-of-concept for our initiation methods.

Two methods for initiating a deflagration in our explosives were used in these experiments: (1) a thermal ignition process that consisted of a Bosch glow plug in physical contact with the PETN, and (2) piston driven ignition in which a projectile is fired into the PETN, driving a compaction wave that eventually starts a deflagration as a result of shear heating and intergranular friction[4]. It has been demonstrated that these two methods produce the same outcomes in 1D DDT experiments[5,6].

The thermal ignition process consisted of a fixture that located a Bosch glow plug to the center of the borehole. A small mass of thermite was tamped into the tube to form a flat surface above the curved tip of the glow plug. The PETN was then loaded into the tube either by dropping consolidated pellets into the borehole or by hand pressing powder on top of the thermite. After the HE was loaded into the tube, a top plate was mounted on top of the tube and the top and bottom plates were bolted together. The assembly is then placed on a stand in a boom box. A DC power supply provided the current to the glow plug to ignite the thermite and generate a burn in the PETN. A schematic of the thermal ignition fixture is shown in figure 2.

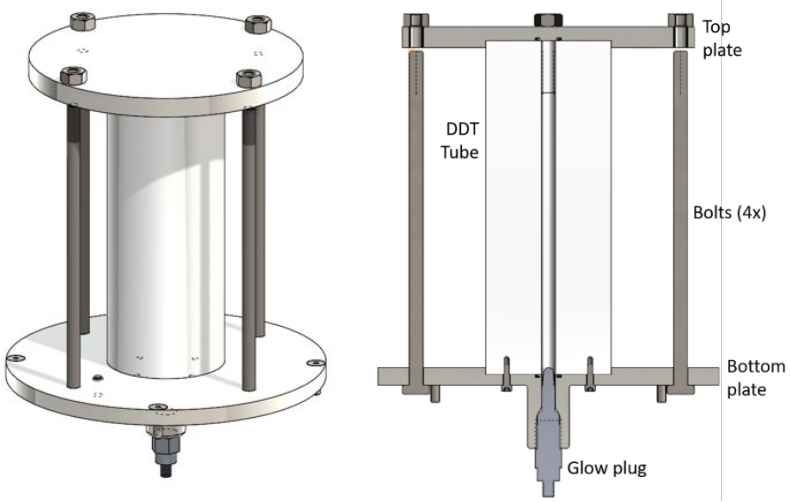

Figure 2. left: assembled thermal ignition fixture; right: cross sectional view of the thermal aging fixture.

Figure 3. top: model of the piston impact ignition gun; lower left: gun with loaded DDT attached; lower right: close-up of loaded DDT tube.

The piston driven ignition experiment consisted of a brass projectile shot down a gun barrel. The projectile is propelled by smokeless powder lit with an e-match in the breech of the gun. Projectile speeds of approximately 150 m s^{-1} are routinely achieved. The DDT tubes mount to the end of the gun barrel and are aligned so that the DDT borehole is concentric with the gun barrel. Prior to firing, the barrel of the gun and the DDT tube are evacuated of air. As the projectile leaves the gun barrel, it enters the DDT tube and impacts the PETN, generating a compressive burn in the explosive. The velocity of the projectile is monitored and recorded throughout the test duration.

Detonation is characteristically a reaction wave that that is supersonic with respect to the speed of sound in the unreacted medium. In PETN, the sound speed is on the order of 2 km s^{-1} depending on its density. The steady state detonation velocity for pressed PETN ranges from 6-8 km s^{-1}, also depending on the density. Therefore, detonation can be identified by luminous burn fronts travelling at characteristic detonation velocities. Hence, the main diagnostic was high speed cameras. The cameras were common to both the thermal and piston ignited DDT experiments. Typically, two cameras were used in each experiment. The first was a color v2512 Phantom camera running at up to 300,000 frames per second to produce wide field recordings of the experiments. Oriented at a right angle to the first camera, a monochromatic v710 Phantom camera was ran at 980,000 fps to produce a streak record of

the experiment[7]. When possible, post mortem analysis of the tube and borehole surface finish was also employed to corroborate the findings from our high speed video.

RESULTS AND DISCUSSION

A total of 30 shots were fielded during the course of the experiment. For the sake of brevity, we will only discuss six of these tests in detail to convey the trends in the PETN response and also provide the reader insight into the reasoning we used to arrive at our conclusions.

A. Thermal Ignition Experiments

The first experiment conducted consisted of 1.65 g cm^{-3} PETN pellets dropped into a polycarbonate tube fitted with the glow plug ignition fixture. A DC power supply was used to supply roughly 10V and 10A current to the glow plug. A loud bang from the PETN was heard roughly 30s after turning on the power supply. The streak record is shown in figure 4. A luminous burn travels up the side of the tube at approximately 0.8 mm μs^{-1}. The horizontal striations seen in the streak record at later times are the pellets burning almost simultaneously at their surfaces. When opening the boombox after the experiment, we observed a great quantity of unreacted PETN scattered inside the box. This indicates irrefutably that there was no transition to detonation in this experiment, and also that polycarbonate provides insufficient confinement to contain flame and product gases from burning PETN, at least when thermally ignited where pressurization rate is slow.

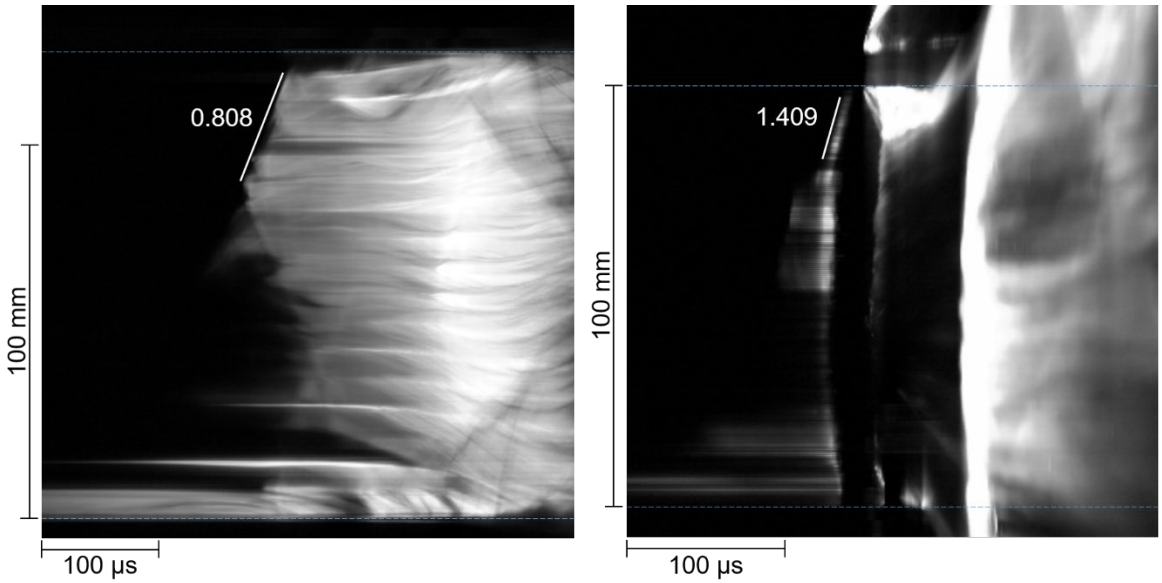

Figure 4. left: streak record for thermally ignited 1.65 g cm^{-3} PETN confined in polycarbonate tube; right: streak record for thermally ignited 1.65 g cm^{-3} PETN confined in steel tube. DDT does not occur in either shot.

In the second test, 1.65 g cm^{-3} PETN pellets were loaded into a steel tube and ignited using the glow plug/thermite system. The streak record for this test is shown in figure 4(b). There is no sustained continuous luminous feature; however, there is a short luminous front travelling at approximately 1.4 mm μs^{-1} followed by simultaneous burning of all the pellets along the length of the steel tube. In contrast with the analogous experiment in polycarbonate, steel provided sufficient confinement to contain the burning PETN long enough that it was entirely consumed during the experiment. No

unreacted PETN was found at the bottom of the boombox post-test. Nonetheless, we can conclude that no DDT occurred in this shot because of the absence of a supersonic luminous burn that would indicate detonation in the streak record.

The experiment was repeated again in a sapphire tube, which has comparable yield strength to steel, but is transparent and therefore allows for wide field of view for high speed videography. We again observe the subsonic luminous burn that travels up the side of the tube. The wide field video reveals that this burn is flame slippage in the thin gap between the lateral surface of the HE and the tube. Figure 5 shows a frame from the high speed video.

Figure 5. Evidence of flame slippage in thermal ignition DDT experiments with 1.65 g cm^{-3} PETN. Flame slippage indicates that flame and product gas are venting along the sides of the tube, thus preventing a transition to detonation.

In figure 5, the glow plug and the thermite are on the right side of the photo, and the flame is travelling leftward. Flame can be seen travelling up the sides of the tube, and pellet boundaries can be seen in both the already burning half of the tube and in the as yet unburnt section. This shot provided evidence that the subsonic luminous burn observed in the thermally ignited polycarbonate and steel DDT tubes are flame slippage, and not compressive burning that is necessary for DDT to take place.

In all three confiners tested, (polycarbonate, steel, and sapphire) thermal ignition appears to not be able to produce the compressive burn that is a requisite first step towards DDT.

B. Piston Impact Ignition

I. 75% TMD PETN in Steel

PETN was loaded into at steel tube at 1.33 g cm^{-3}. Again, this is the density at which PETN is known to undergo DDT in the shortest run length. The steel tube was 6” tall, with an OD of 1.375” and borehole ID of 0.276”. A 0.020” wide slit was cut down the length of the steel tube to allow for streak videography of the experiment. 0.5 g smokeless powder was used to accelerate the projectile to 155 m s^{-1} before impacting the PETN. The streak record for this shot is shown in figure 5.

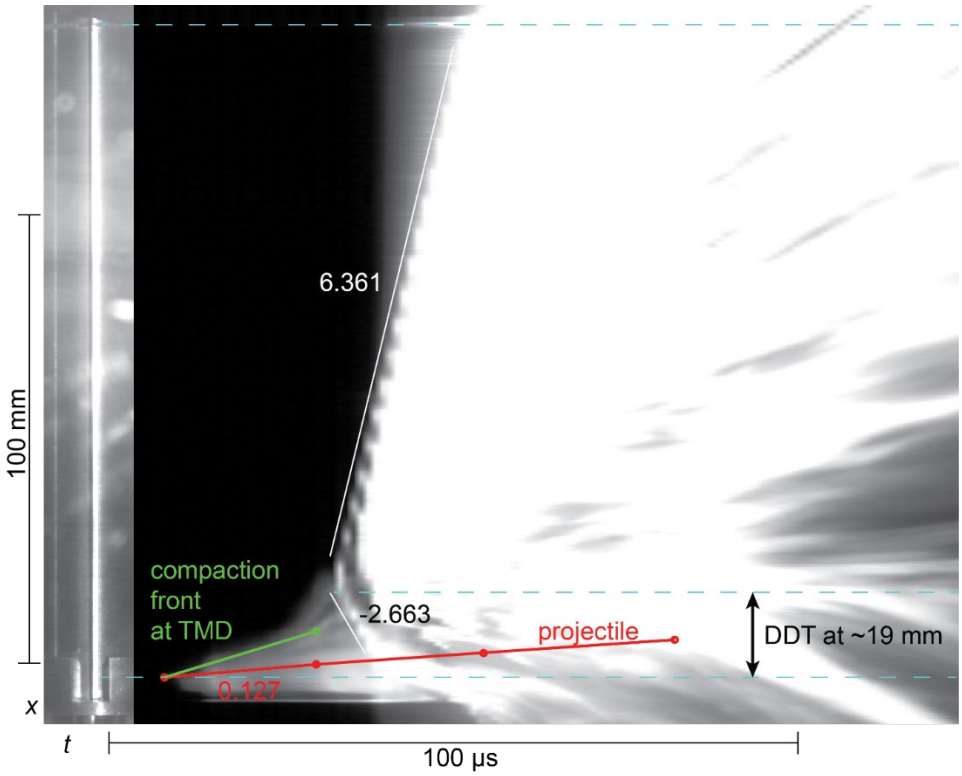

Figure 6. Streak record for 75% TMD (1.333 g cm^{-3}) PETN in steel tube. This shot was intended to demonstrate the efficacy of the piston impact ignition method for causing DDT. Indeed, piston-impacted 75% TMD PETN undergoes DDT in 19 mm, in agreement with Luebcke.

In figure 6, DDT occurs promptly. The run length to DDT is 19 mm, which is in excellent agreement with the results of Luebcke *et al.*, thus demonstrating that the piston impact ignition method is a reliable means of generating a compressive burn causing DDT in PETN. A photograph of the DDT tube post-shot is shown in figure 8. The DDT tube is fragmented into many pieces. Inspection of the borehole revealed the characteristic darkening that occurs when the transition to detonation occurs.

II. 93% TMD PETN in Steel – Piston Impact Ignition

The piston impact ignition experiment was repeated using 1.65 g cm^{-3} PETN. The streak record for this shot is shown in figure 7.

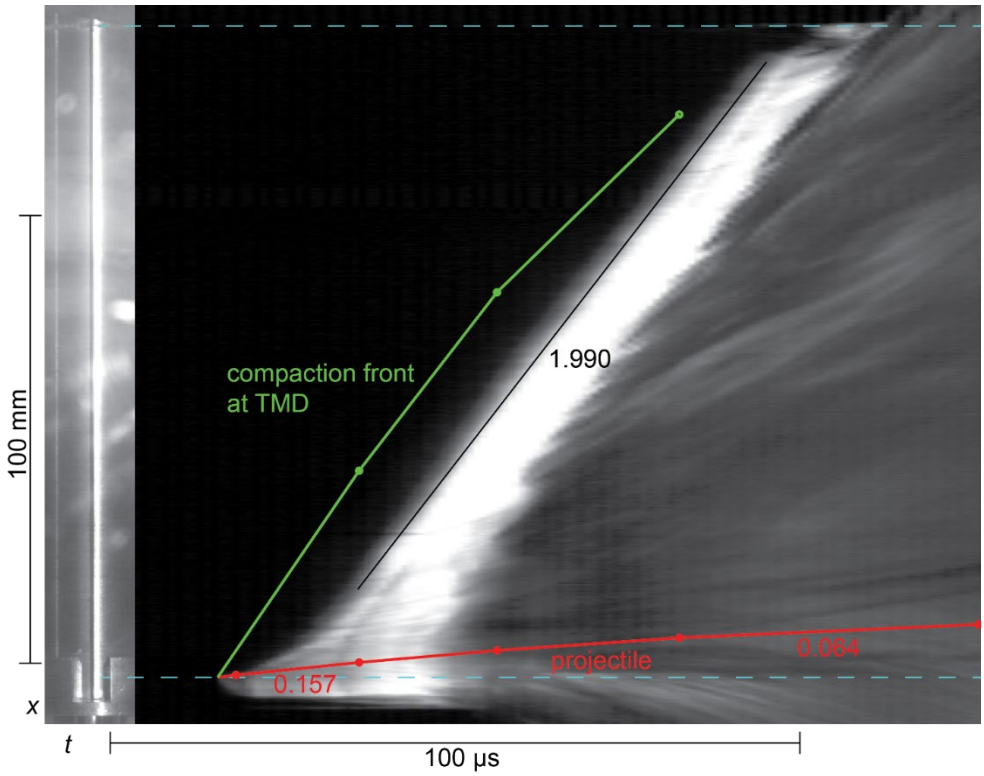

Figure 7. Streak record for 93% TMD (1.65 g cm^{-3}) PETN confined in steel. No DDT occurs.

It is clear from inspection of the streak record that no DDT occurs as indicated by the absence of a supersonic luminous burn. Instead, the slope of the luminous burn indicates a velocity of 1.990 mm µs^{-1}. We were never able to produce DDT via impact ignition in 93% TMD PETN. In all experiments, we only ever observed approximately 2 km s^{-1} steady burning. Compaction wave velocities, u_c, can be calculated using the following equation:

$$u_c = \frac{\rho_1}{\rho_1 - \rho_0} u_p \tag{1}$$

where ρ_1 is the compressed density of the PETN, ρ_0 is the original density of the PETN, and u_p is the projectile/particle velocity. The calculated position of the compaction wave front is shown as the green line in figure 7. The compaction wave front travels at approximately 2 km s^{-1}. Assuming that the PETN is compressed from 1.65 g cm^{-3} to 1.778 g cm^{-3} (100% TMD), this compaction model predicts a compaction wave of 2.153 mm µs^{-1}. This prediction is in good agreement with our measured velocity of the luminous burn, suggesting that what we are observing is a subsonic, or possibly sonic compressive burn.

Figure 8. left: steel tube after compressive burning of 1.65 g cm^{-3} PETN, but no DDT; right: steel tube after DDT occurs in 1.33 g cm^{-3} PETN. Both experiments are very violent and break the steel tube into many parts.

The burn is steady and traverses the entire length of the tube without any indications that it will transition to detonation. However, the compressive burn completely consumes all of the PETN and also does significant damage to the tube. A photograph of the tube post shot is shown in figure 8. Because of the damage that the tube sustains, we initially termed the compressive burn a high explosive violent reaction (HEVR). Figure 8 demonstrates that the damage that both tubes sustain is significant. Even though DDT did not occur in the 93% TMD PETN confined in steel, the compressive burn was similarly destructive.

C. The High Explosive Violent Reaction

A final series of experiments was aimed at better understanding the steady compressive burn observed in 93% TMD PETN piston impact ignition experiments. To investigate whether the steady compressive burn observed in the 93% TMD PETN steel piston experiments could cause any reaction in a booster high explosive, a 6” long steel tube was loaded with a 3” column of 93% TMD PETN followed by a 3” column of 1.87 g cm^{-3} PBX 9012. The same steady compressive burn was obtained in the PETN column by impacting the HE with a projectile at 155 m s^{-1}. The response of the PBX 9012 was then monitored in figure 9.

The steady compressive burn in the PETN is seen once again travelling at 1.9 km s^{-1}. As soon as the luminous burn reaches the bottom of the PBX 9012 column, the reaction is quenched. The compressive burn in PETN does not cause any significant reaction in the PBX 9012. This is corroborated from the framing camera record, in which unreacted PBX 9012 can be seen extruding through the streak slit, and from the presence of large pieces of unreacted PBX 9012 inside of the boombox post-test.

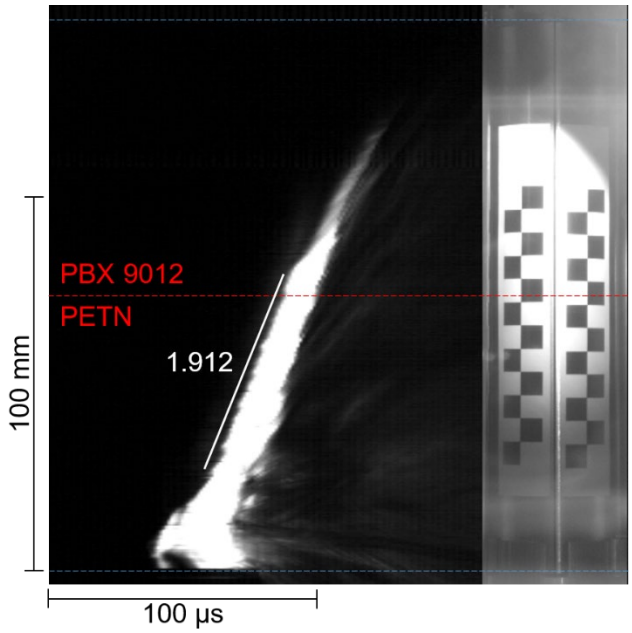

Figure 9. The streak record for 93% TMD PETN compressive burn transitioning into PBX 9012. The location of the boundary between the PETN and the PBX 9012 is shown as a blue dashed line. As soon as the compression wave travels into the PBX 9012, the compressive burn following promptly quenches.

CONCLUSION

In this study, we investigated whether or not 1.65 g cm^{-3} Type 12 PETN could possibly undergo a transition from burning to detonating, and if so, measure the length required to do so. DDT tubes were designed to provide significantly more confinement than a PETN pellet would encounter in a detonator, so that we could have the best possible chance at measuring the DDT length in 1.65 PETN. Of the two methods for initiating a burn in PETN, we found that the piston driven ignition was the best method to examine the issue. In all three confiners when the thermal ignition mechanism was employed to initiate a burn in the 1.65 g cm^{-3} PETN (polycarbonate tube, sapphire tube, and steel tube), product gases from burning PETN would slip up the side of the tube and light the lateral surface of the entire column of pellets, and in the case of the polycarbonate and sapphire tubes, cause the tubes to fail before the compaction-driven DDT mechanism could be initiated. In the case of thermal ignition in steel confinement, the steel was strong enough that the PETN was completely consumed by the reaction, but the flame slippage prevented the bed from being compressed by product gases, and ultimately served as the failure mechanism for DDT to occur. From this series of thermal experiments, we conclude that if the PETN is either (1) not pressed into its housing or (2) is housed in weak confinement, product gases will preferentially vent instead of compressing the powder. We did not observe any DDT in thermally ignited 1.65 g cm^{-3} PETN. Additionally, we learned that (3) a piston impact ignition test is a more severe test and was better suited to achieving our objectives.

We demonstrated that DDT was possible in 73% TMD PETN and the run length to the transition agreed with Luebcke. In steel tubes, piston ignition proved to be necessary to generate a 1D compressive burn in 1.65 g cm^{-3} PETN without any gas slippage. Piston impact shots were conducted for 1.65 g cm^{-3} PETN in both steel and polycarbonate. In all tests where 1.65 g cm^{-3} PETN was confined in a steel tube, a steady deflagration was achieved within roughly a centimeter of the impacted surface of the PETN. This

luminous deflagration travelled at roughly 2 mm μs^{-1} along the full length of the tube, and did not show any signs of acceleration that might have indicated conditions approaching DDT. If we assume that the 1.65 g cm^{-3} PETN is compressed to 100% TMD by the impact of the piston, theory predicts that the generated compaction wave will travel at approximately 2 mm μs^{-1}, which is also the sound speed in 1.65 g cm^{-3} PETN. This suggests that the luminous front is compressive burn. As with thermal ignition, DDT did not occur in 93% TMD piston ignition experiments.

Finally, even though no detonation occurred as a result of the rapid compressive burn in 1.65 g cm^{-3} PETN, the deflagration was nevertheless violent (as evident by postmortem analysis of the steel tubes). Additional testing was executed to probe the nature of the deflagration and its potential ability to cause violent reaction in booster materials. When the 2 mm μs^{-1} compressive burn from 1.65 g cm^{-3} PETN transits into a sample of PBX 9012, the burn completely quenches as indicated by the disappearance of the luminous burn. Therefore, deflagrating PETN did not initiate a deflagration in PBX 9012, and therefore, would not cause PBX 9012 to detonate.

In conclusion, we did not observe DDT for 93% TMD/1.65 g cm^{-3} PETN in any of our experiments, despite the fact that the PETN was in much more strongly confined configurations than it would have been in a detonator. It is distinctly possible from results of our tests and other that this density of PETN lies past an asymptote on the PETN density versus DDT run length curve, above which it is simply not possible to transition from deflagration to detonation. Nevertheless, we showed that the run length to DDT is at least greater than 6" (the longest tube that was tested), which is approximately 20 times more run length than is available in most detonators. Even in our trials where we adjusted density to lie on the run-length curve, the length required for DDT was too great to be a concern.

REFERENCES

1. Luebcke P. E., Dickson P. M. & Field John Edwin. An experimental study of the deflagration-to-detonation transition in granular secondary explosives. *Proc. R. Soc. Lond. Ser. Math. Phys. Sci.* **448,** 439–448 (1995).
2. Luebcke, P. E., Dickson, P. M. & Field, J. E. Deflagration-to-detonation transition in granular pentaerythritol tetranitrate. *J. Appl. Phys.* **79,** 3499–3503 (1996).
3. McAfee, J. The Deflagration-To-Detonation Transition. in *Shock Wave Science and Technology Reference Library, Vol. 5: Non-Shock Initiation of Explosives* 483–535 (Springer-Verlag, 2010).
4. Panchadhara, R. & Gonthier, K. A. Mesoscale analysis of volumetric and surface dissipation in granular explosive induced by uniaxial deformation waves. *Shock Waves* **21,** 43–61 (2011).
5. Campbell, A. W. *Deflagration-to-Detonation Transition in Granular HMX*. 32 (Los Alamos Scientific Laboratory, 1980).
6. McAfee, J., Asay, B. & Bdzil, J. Deflagration-to-Detonation Transition in Granular HMX: Ignition, Kinetics, and Shock Formation. in *Proceedings of the 10th International Detonation Symposium* (1993).
7. Parker, G. R., Asay, B. W. & Dickson, P. M. Note: A technique to capture and compose streak images of explosive events with unpredictable timing. *Rev. Sci. Instrum.* **81,** 016109 (2010).