The Deflagration-to-Detonation Transition Process for High-Energy Propellants—A Review

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Introduction

THE deflagration-to-detonation transition (DDT) in solid energetic materials has been of interest to researchers for many decades since it has a variety of areas of applicability, ranging from industrial to military. A recent example of the former is the detonation that occurred during the production of propelling charges for hunting ammunition.[1] In the military area, the applicability extends from gun systems and projectile impact hazards to rocket motors. Hence, the DDT process has been investigated for both voidless (cast) and porous systems. We shall use the rocket motor environment to assess the DDT hazards associated with high-energy propellants.

In the area of solid propellant rocket motors, an oft-asked question is "Will a cast, well-manufactured rocket propellant grain undergo a transition to detonation from the burning mode?" The answer is no, to the best of our knowledge. Although there are few journal articles directly providing this assessment, our knowledge of the DDT mechanism in gases, liquids, and solids provides a rationale for reaching this conclusion. The rationale is based on the thesis that the deflagration process must ultimately produce a shock wave to drive the system to detonation.[2,3] That is, the shock-to-detonation transition (SDT) is the final stage in any DDT process. Consequently, in the solid-propellant rocket motor situation, the confinement provided by the motor case must be sufficient to allow the pressure from deflagration to build up to a sufficiently high shock pressure to initiate the cast propellant. As will be shown below, these shock amplitudes cannot be reached for cast propellant systems confined in rocket motor cases.

DDT Process in Cast Energetic Materials

Very little documentation is available in the literature concerning the DDT characteristics of cast propellants. Consequently, we must rely upon the insight gained from studies with cast explosives. Among the cast secondary explosives, very few will undergo DDT, even in high-confinement (steel tube) systems. Thus, most of the mechanistic insight has been gained using diethylnitramine dinitrate (DINA) and pentolite (50/50 PETN/TNT). The model of the DDT mechanism for cast explosives proposed by Macek[4] showed that 1) an "exponential" pressure-time (p-t) relationship resulted from simple conductive (laminar) burning at the ignited end of a highly confined cylindrical charge and 2) compressive waves emanating from the ignition region coalesced into a shock at some location (ℓ_s) beyond the burning region. His experimentally measured p-t data were consistent with, but not exactly identical to, his theoretical predictions. With these p-t data, it was shown that the calculated ℓ_s values were in basic agreement with the experimentally observed location for the onset of detonation ℓ. Gipson and Macek[5] subsequently demonstrated the propagation of compressive waves immediately before the onset of detonation. Thus, the model's predictions and the experimental observations were consistent with the thesis that SDT is the final stage in the DDT process.

Price and Wehner[6] extended these experimental DDT studies and also included cast Comp B (60/40/1 RDX/TNT/wax) and cast TNT in their analysis. As a result of the failure of these latter materials to undergo DDT, it was concluded that if a transition to detonation is to occur, then the confinement must be maintained until the pressure reaches a critical value to effect the transition. That is, "the critical pressure in the burning region appears to be equal to the critical pressure for shock initiation." They concluded that to predict the occurrence of a transition to detonation, one needed knowledge of 1) the rate of pressure buildup in the ignition region, 2) the maximum pressure reached before tube rupture, and 3) the shock sensitivity of the test explosive *in situ*. However, recent calculations by Jacobs[7,8] have indicated that conductive burning *could not* have produced a shock wave for the cast pentolite and DINA. He found that the material velocity of the explosive interface had not been referenced to the conductive burning rate in the Macek model. When this is properly included, the model will not produce the experimentally observed exponential pressure rise. Tarver et al.[9] modified the Macek model to provide a more realistic treatment of the accelerating flame. Their calculations showed that the deflagration velocities in cast pentolite during DDT must be 250-500 times faster than those expected from normal burning rates. Thus, it was concluded[8,9] that a mechanical means of increasing the burning

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surface should be included in a realistic model of DDT in cast explosives.

One possible explanation for the breakup of material in the ignition region is based upon the deformation of the confining tube. Because of the finite yield strength of the confining tube, deformation of the tube from internal gas pressurization could produce a thermal environment around the free (burning) surface of the charge that leads to breakup. Experimental evidence of such thermal breakup of TNT-based cast explosives can be found in the closed-bomb combustion studies of Wachtell and McKnight.[10] On the other hand, Kooker and Anderson[11] have postulated that breakup in mildly porous explosive charges confined in steel cylinders could arise as the result of shear-induced stresses from the confined burning. Consequently, the present accumulated knowledge indicates that there is very little likelihood that a cast explosive or propellant could undergo DDT from conductive burning. Only if there is breakup (introduction of porosity) in the ignition (burning) region of the cylindrical sample will sufficient pressurization rates be attained to produce the SDT portion of the DDT process for these cast explosives.

There is additional information in the literature to support the conclusion that the burning of porous material in the ignition region (e.g., resultant from breakup) can drive a cast system such as Comp B to detonation. These data come from the studies of Price and Bernecker[12-15] ranking explosives by their susceptibility to DDT. They developed a *gas loading* procedure in which a porous column of a rapidly burning, but nondetonating, material is located between the ignitor and the test material in the DDT tube. By varying the length of the gas loader, the pressure and pressurization rates (dp/dt) experienced by the test material were systematically changed. This technique has been successfully applied to porous[12,13,15] and cast systems.[14,15] For both systems, it was found that the distance to detonation (relative to the gas loader interface ℓ' or the ignitor interface ℓ) decreased with the length of the gas loader ℓ_g.* In this manner, when pentolite was gas loaded ($\ell_g \sim 40$ mm), the predetonation column length ℓ was significantly shorter with the gas loader. For a given length of gas loader (e.g., 50 mm), the order of DDT susceptibility for cast explosives decreased in the order: pentolite $\gg$ Cyclotol 75/25 > Comp B. Cast explosives that failed to be gas loaded to detonation included TNT and various cast plastic-bonded explosives (PBXs).[15] Since the latter materials are elastomeric systems (similar to some propellant systems) and most of the PBXs are more shock sensitive than the propellant systems, it is inferred that most cast propellants will not undergo DDT—even under severe gas loading and high-confinement conditions. Hence, the accumulated data strongly suggest that pressure from the confined burning of cast or broken propellants may not be sufficient to drive a *cast propellant* system to detonation.† If the broken material is present in both the burning region and the incipient shock region, then the DDT process is representative of DDT in a porous system. As shown below, a porous system has a much greater likelihood of undergoing DDT.

The occurrence of DDT in a rocket motor is thus based upon damaging of the propellant grain to produce region(s) with some porosity.[17] According to Butcher et al.,[17] "The type of damage and extent of damage have been reasonably well identified by examination of research motors purposely damaged in cold blowdown testing." Thus, in at least one instance, there exists a reasonable description of the type of granular material to be used in laboratory DDT experiments pertaining to motor hazards.

DDT Process in Porous Propellant Systems

The study of the DDT process in porous samples of energetic materials (explosives and propellants) has a long history.[2,3,18] The nature of the confined burning in porous charges was postulated to differ from the laminar (conductive) burning description used by Macek[4] for cast systems. It was recognized that the presence of pores in a granular sample could facilitate the flow of hot combustion gases through interconnected pores.[2,18] Because of these flow properties, as reflected in the permeability of the sample, the hot gases from confined burning could convectively transfer their enthalpy to the pore walls. Continuous flow of the hot gases could lead to ignition and combustion within the granular sample. Because of the greater burning surface and gas generation rate resulting from this process (called convective combustion), the pressure buildup near the ignitor/explosive interface could increase dramatically under the proper experimental conditions. (The convective combustion process is the basis for the gas loading procedure described earlier.) As seen from this description, permeability (which is a function of the sample's porosity, particle size distribution, and particle shape) is an important factor in the DDT process in porous energetic materials.

In practice, convective combustion can be studied in nondestructive experiments (as opposed to a DDT tube experiment) using a high-pressure combustion vessel. A variety of researchers[19-22] have conducted extensive studies in convective combustion (CC) of explosives, delineating the importance of composition, permeability, and charge dimensions in influencing the onset of CC and its propagation characteristics. Based upon these investigations, analytic models of CC were developed.[23-25] In the field of gun propulsion, various American researchers have conducted experimental studies of CC.[26-28] This latter work provided data for the development and verification of theoretical/numerical models of CC[29-31] As a result of this modeling of the CC process, numerical models of DDT have been formulated in recent years.[32,33] Because of the complexity of the DDT process in porous materials, experimental studies have led the development of these theoretical/numerical models of DDT. Ultimately, meaningful theoretical models must be developed that will yield accurate predictions based upon the chemical, mechanical, and physical properties of the porous medium. Only with the comparisons between the numerical calculations and the experiments will the DDT process be sufficiently quantitative for application to evaluations of the hazards in rocket motor situations.

Experimental Parameters

While insight into the boundary conditions needed for CC can be obtained from nondestructive experiments, much larger samples are needed to assess whether the important pressure buildup from CC can drive the porous system to detonation. The number of important experimental parameters for studying DDT in porous systems is larger than with cast systems; these experimental variables are listed in Table 1.

As in the case of cast systems, confinement is at the top of the list. Most DDT experiments have been conducted in high strength (metallic) tubes.[34-37] High-confinement conditions yield data that are more one-dimensional, thus simplifying data analysis and parametric correlations. Plastic tubes had also been used in the earlier studies[35,38] to demonstrate the effect of confinement on effecting a transition to detonation. Calzia and Carabin[39] worked with porous columns of RDX and RDX/wax in a thick-walled plastic tube. They found that it was necessary to use venting to avoid rupture of the tube before a transition to detonation could be attained in

*Of course, too short a column of gas loader ($\ell_g < \ell_g^{crit}$) would fail to boost the materials to detonation. In these measurements, $\ell = \ell_g + \ell'$.

†On the other hand, numerical calculations by Forest[16] indicate that the burning of a porous region can produce a transition to detonation in adjacent cast regions for certain geometric configurations.

Table 1 Variables influencing experimental studies of porous systems

1) Confinement
High: steel tube
Low: plastic tube
2) Ignition strength
Weak: gasless or low dp/dt
Strong: gaseous or high dp/dt
3) Physical properties of porous medium
Permeability: particle size, particle shape, porosity
Compressibility
4) Chemical composition
Decomposition kinetics: pyrolysis, ignitability, and burning rates
5) Charge dimensions
Diameter
Column length
6) Detonability
Shock initiation pressure P_i

these relatively shock-sensitive systems. It was evident from all of these studies that the ability of a porous material to transit to detonation depended strongly on the confinement. Plastic deformation of the tube can diminish the pressurization rate produced from confined burning and thus limit the pressure levels available for shock initiation (the final stage of DDT).

If one is evaluating the possibility of DDT occurring in a rocket motor configuration, then the confinement representative of the motor system must be used. To this end, Pilcher et al.[40] and Butcher et al.[41] have evaluated several experimental configurations for the study of DDT in porous beds of damaged propellant samples. Of the three tube configurations evaluated [thin-walled steel tube, Kevlar (fiber) case, and thick-walled plastic (Lexan) tube], the plastic tube proved more valuable, primarily because the DDT process could be monitored optically. The wall thickness (50.8 mm) of the Lexan tube (50.8 mm i.d., 152.4 mm o.d.) was selected to simulate the inertial and stress confinement of a typical full-scale motor.[41]

The second area of importance for the study of DDT in porous systems is the nature of the ignition aid. Most DDT experiments use an ignition aid (ignitor) to start the burning of the energetic material being studied. Until recently, insufficient attention was given to the characteristics of the ignition system. However, based upon observations made in CC studies[42] and the gas loading DDT experiments,[12,13] it was noted[43] that compaction of very porous columns of energetic materials may occur during the burning of the ignitor and thus may precondition the column. This preconditioning would produce density gradients within the column before the transient combustion of the energetic material could occur.

This preignition compaction can, consequently, change the boundary conditions for each of the stages in the DDT mechanism. The extent to which compaction occurs depends on (besides the ignitor's burning and gas generation characteristics) the physical properties of the granular energetic material—item 3 in Table 1. Both the permeability‡ and the compressibility of the porous column are very important. A decrease in permeability (as reflected in the use of a smaller particle size at a constant sample density) will cause the accumulation of gases from the ignitor's combustion process (and any preignition pyrolysis) and change the pressure loading experienced by the sample. On the other hand, keeping the permeability constant, but changing the compressibility of the sample, changes the extent of the preignition compaction for a given pressure level. As the compressibility of the sample increases, the extent of preignition compaction increases; this change results in a dramatically reduced permeability. It is easy to see how this feature would influence the onset of CC, which depends on convective heat transfer.

The final items from Table 1 to be discussed are the dimensions and detonability of the sample. Sample diameter is important because there are critical diameter dependencies for the onset of both CC[20] and steady-state detonation. The detonability is determined by the critical diameter for detonation and the shock initiation pressure P_i. It is possible for a system to be compatible with the first, but not the second item. An example of this is a low confinement DDT tube arrangement having a large inner diameter and using a fairly shock-insensitive material. As stated above for DDT in cast explosives, it is important to have some idea of the magnitude of P_i *in situ* for the energetic material being studied. Shock initiation pressure is a relative term that depends upon several experimental parameters, one of which is the time duration of the shock loading. For the purposes of this paper, P_i is defined as that pressure at which detonation occurs for a fixed "run up" distance X_D (X_D should be at least one charge diameter). In a SDT experiment, X_D is the distance from the initially shocked end to the location where detonation begins. In terms of a DDT experiment, the region traversed by a shock wave is usually a small fraction of the predetonation column length ℓ.

Hazards Considerations

The most practical information pertaining to hazards that can be obtained from DDT studies is knowledge of the permeability range over which DDT will occur and some quantitative measure of the propensity for that granular mixture to undergo DDT. A representation of this propensity is a correlation of ℓ as a function of the initial permeability of the system. Permeability has only occasionally been measured for DDT samples.[34] Instead, this type of correlation has been made by displaying ℓ as a function of porosity for various particle sizes and shapes or as a function of particle sizes at a constant density or percent theoretical maximum density (% TMD).

The most complete description of the susceptibility of a *porous propellant system* to DDT at low confinement lies in the work of Butcher et al.[41] In this work, the particle size and shape were combined into a single variable, the surface-to-volume (S/V) ratio. In this manner, the granulated propellant§ used in the laboratory experiments could be correlated with propellant fragments from naturally damaged propellant. The S/V ratio was based upon the closed-bomb pressurization rate data and correlated with materials of known dimensions and conductive burning rates. Typically, the granulated particles had a S/V ratio about four times that calculated from their dimensions.[41] This fact illustrates the "damaged" nature of the surface of the propellant granules.

The DDT arrangement used by Butcher et al. was the thick-walled Lexan tube mentioned earlier (i.e., 50.8 mm wall); their ignitor could be classified (based upon the analyses described below) as a strong ignitor that could produce extensive preignition compaction. Their data established a fairly well-defined pattern for the limits of DDT of systems with 50% porosity or less. In Fig. 1 these limits are shown as a function of the permeability (% TMD and S/V ratio); the dashed line encompasses the region where DDT occurred for their experimental configuration. The highest permeability of a sample is at the lower left side of the

‡The initial permeability of a column is usually characterized by measuring the flow of a gas through the column and calculating a permeability coefficient as defined by the Darcy equation. Andreev and Chuiko[19] measured permeability coefficients for a variety of explosives having different sizes and porosities.

§Granulated propellant is defined here as any material manufactured from cast propellant by dicing, shredding, chopping, or any other mechanical means.

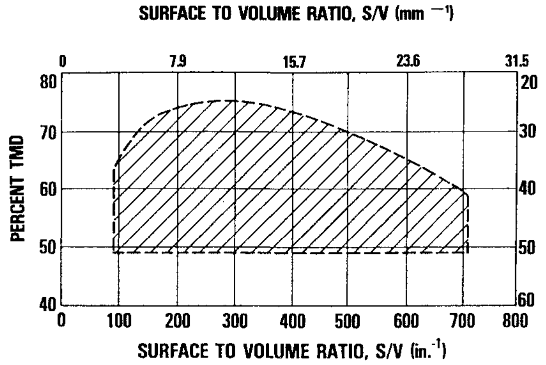

Fig. 1 Limits of DDT for granulated propellant samples using experimental apparatus of Ref. 41.

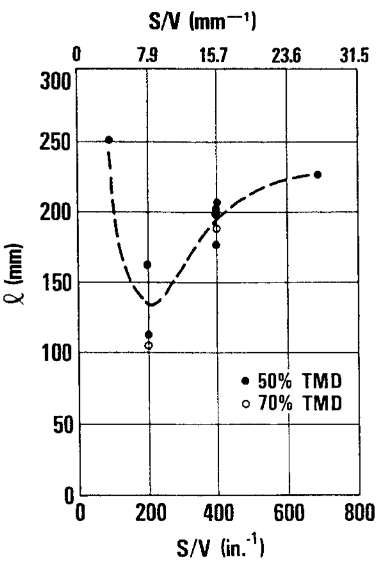

Fig. 2 Dependence of ℓ on surface-to-volume (*S/V*) ratio of granulated propellant.[41]

figure; the lowest permeability is at the upper right side. The data in Fig. 1 show that at 50-60% TMD, all samples tested (*S/V* limits of 100-700 in.$^{-1}$) underwent DDT. Thus, DDT occurred irrespective of the initial permeability of the sample at these two densities.

However, this observation does not mean that all samples in this grouping were equally susceptible. The variation of this susceptibility (as indexed by the predetonation column length ℓ) is shown in Fig. 2 as a function of permeability. The data show a minimum in the ℓ-*S/V* curve for 50% TMD samples. That is, as the permeability decreased [*S/V* ratio increased from 100 to 200 in.$^{-1}$ (3.94 to 7.87 mm^{-1})], ℓ decreased; a further decrease in permeability resulted in larger ℓ values.¶ Consequently, there is a permeability (identified by the 200 in.$^{-1}$ material) at which 50% TMD material is most susceptible to DDT. For 70% TMD material, we see that the two data again indicate that the 200 in.$^{-1}$ material is more susceptible than the 400 in.$^{-1}$ (15.7 mm^{-1}) material. With respect to motor hazards application, these observations mean that material representative of 3.0 mm cubes (*S/V*~200 in.$^{-1}$) essentially represents a worst case for DDT. However, in large rocket motors where the damaged bed length may be a meter or more in length, the difference between ℓ values of 100 and 254 mm (as seen in Fig. 2) may be insignificant. Finally, the observation, for this low-

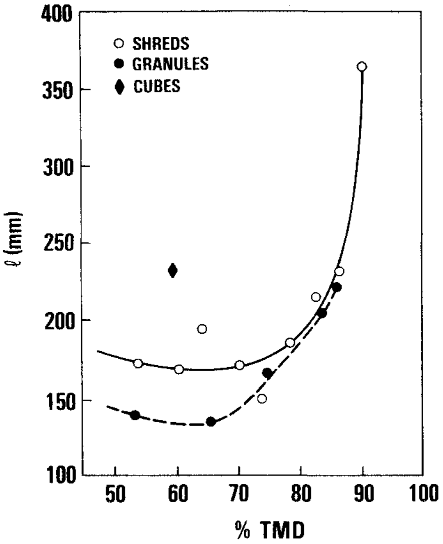

Fig. 3 Dependence of ℓ on initial compaction for various propellant particle sizes and shapes.[44]

confinement system, that there is a minimum permeability at which the propellant was most susceptible is in agreement with earlier observations made with crystalline explosives under high-confinement conditions.[34-36]

A cross-linked double-base (XLDB) propellant has been studied at high-confinement conditions by Bernecker et al.[44]; the ignition system used there can also be classified as a strong ignitor. The XLDB propellant was in the form of shreds, granules (powder), and cubes. The variation of ℓ with permeability is shown in Fig. 3; here the permeability is varied by either changing the density for a given form of propellant or by changing the particle shape at a fixed density (i.e., at 60% TMD). While the data for the shreds and granules indicate that ℓ increases with decreasing permeability, the scatter of the data preclude the observation of any minimum. The *increase* of ℓ with *decreasing* permeability in Fig. 3 most likely can be associated with the increase in ℓ with *S/V* from 200 to 700 in.$^{-1}$ (7.87 to 27.6 mm^{-1}) in Fig. 2. On the other hand, evaluation of the data in Fig. 3 at 60% TMD shows that ℓ *decreases* with *decreasing* permeability. That is, for the more permeable column of 1.6 mm cubes, ℓ is larger than the ℓ values for the less permeable columns of ~0.5 mm granules or the thinner shreds. With reasonable justification, it might be postulated that this correlation is consistent with the variation in ℓ with *S/V* from 100 to 200 in.$^{-1}$ (3.94 to 7.87 mm^{-1}) in Fig. 2. However, at the end of the paper, it shall be shown for very compressible propellant samples, used with strong ignitors (i.e., systems representative of Fig. 3 and possibly Fig. 2), that the influence of permeability considerations may be very small and that the SDT behavior of the porous samples is the dominant feature which determines the ℓ values.

The data in Fig. 3 also show that DDT could be attained in samples having only 10% porosity (90% TMD). This was possible because the higher confinement conditions could lead to higher shock levels for SDT (for a given system, P_i increases as % TMD increases). On the other hand, the very rapid increase in ℓ as TMD increases above 80% suggests that it would be very difficult to observe DDT in porous systems approaching TMD. Thus, the data are consistent with the earlier conclusions concerning the probability of DDT in cast propellant systems.

Improved Experiments for Evaluation of DDT Hazards

A complicating feature in relating laboratory experiments to the hazards associated with motor malfunctions is the fact that an idealized representation of the damaged propellant

¶Assuming that the particles are cubes, the *S/V* ratios can be used to give estimated dimensions. Thus, the edge dimensions are calculated to be: 6.1, 3.0, 1.5, and 0.9 mm (*S/V* 100, 200, 400, and 700 in.$^{-1}$).

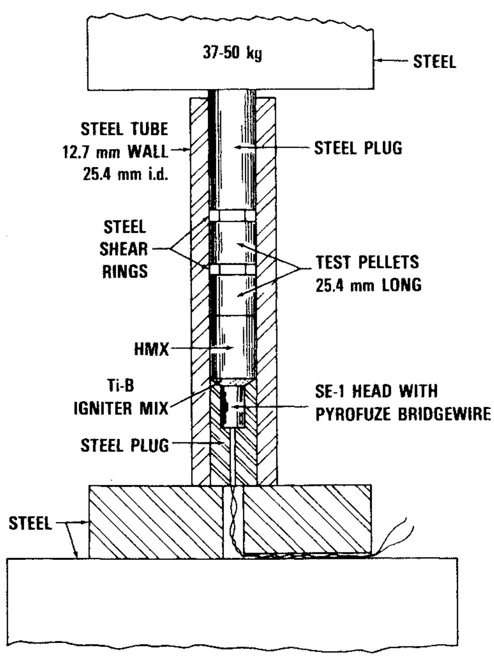

Fig. 4 Dynamic shear/DDT experiment.[45]

sample is used. That is, the damaged sample is made by consolidating regularly shaped granules into a column with essentially uniform porosity. This idealization is necessary in order to correlate data from DDT experiments with numerical model predictions or to provide insight for the development of these models. However, from the hazards viewpoint, the typical DDT experiment is only a first (and not necessarily always a good) approximation. A much better correlation of DDT susceptibility to motor hazards is an experiment in which the damaged sample is generated *in situ* during the experiment. An experiment of this type has been developed and described by Campbell.[45]

Campbell[45] was interested in defining the role of shear in producing rapid reaction ("hot-spot" burning) and possibly DDT. A variety of experimental arrangements were described in which no reaction or a subdetonation process was induced in the test materials. However, the experiment most pertinent to this discussion is that shown in Fig. 4 (this is Fig. 13 in Ref. 45). In this arrangement the burning of a 64.2% TMD column of HMX is used to induce dynamic shearing of test materials confined in steel tubes. (One material was a pressed PBX with very low porosity; the other was a cast, compliant propellant.) Shear rings were placed between adjacent pellets of the test material. It was shown that detonation could be induced in both materials under various conditions. (The confined burning arrangement is similar to the gas loading system[12-15] described earlier.) For example, using a 12.7 mm column of porous HMX, detonation could be induced with a shear ring thickness of 6.35 mm. Using a longer column (25.4 mm) of HMX, the minimum shear ring thickness that would produce DDT was about 3 mm (2.3 mm produced a failure, 3.2 mm produced detonation). It is illuminating to consider the short "predetonation column lengths" involved in these experiments. For example, ℓ' is less than 50.8 mm (excluding the length of the rings); correspondingly, ℓ is less than 76.2 mm. Very few low-porosity systems have such low ℓ values (see, for example, Figs. 2 and 3). The observation that a 3.2 mm thick shear ring could cause enough shearing action and breakup to produce detonation is most intriguing and worthy of further research.

The dynamic shearing DDT experiment developed by Campbell is a good example of the type of experiment that relates DDT aspects to potential mechanisms that might occur during motor blowdown. More experiments of this type are needed for the more practical aspects of hazards evaluation. In addition, they should be more extensively instrumented such that the models of CC and DDT can be used to provide mechanistic insight into these dynamic processes.

Table 2 Stages of DDT mechanism for porous charges

1) Preignition
2) Ignition/conductive burning
3) Convective burning
4) Compressive ("hot-spot") burning
5) Shock formation
6) Compressive burning
7) Detonation

DDT Mechanism(s) in Porous Propellant Systems

The objective of most recent experimental DDT studies on propellants has been to provide useful data that aid in the development of physical models of the DDT process and guide (or confirm) the development of theoretical/numerical models of the DDT process. The stages of the DDT process in porous columns of energetic materials are outlined in Table 2. These stages are the same as discussed in Refs. 37 and 43 with the exception of the preignition stage, which has been added to address the observations from recent plastic tube experiments.[46,47]

High-confinement apparatuses provide data that are more one-dimensional and more directly useful for the one-dimensional computational models. Unfortunately, the use of high-confinement systems has generally precluded extensive diagnostic instrumentation and the recent important observations concerning the DDT mechanism have come from low-confinement experimental arrangements. The low-confinement (plastic tube) arrangements have utilized flash radiography and high-speed photography to outline quantitatively the propagation of reaction fronts and to measure the nature (i.e., density profile) of the reacting propellant column at various predetonation times. Associated instrumentation (e.g., piezoelectric transducers) provided important *p-t* histories during the predetonation period. The use of flash radiography has demonstrated (for the high-porosity samples used in plastic tube arrangements) the importance of compaction during the DDT process. Compaction has been shown to occur in every stage of DDT for very porous samples.

Postignition compaction in the DDT process has been inferred from indirect experimental work with explosives in steel tubes.[35,36] Direct measurement of extensive compaction from the radiographic studies was first reported[48] for XLDB propellants and a nitrocellulose (type A) fluid ball powder in a Lexan tube arrangement with a strong ignitor.** Two different compaction modes were observed at very similar porosities. In the first mode, only axial compaction occurred, with no detectable separation between the propellant column and the Lexan tube. This mode was observed for a 57.6% TMD charge of the nitrocellulose (NC) powder. In the second mode, *radial* as well as *axial* compaction occurred. The radial compaction and the expansion of the Lexan tube led to the presence of a large annular space between the expanded tube and the lateral surface of the compacted column. For a 58.2% TMD column of a shredded XLDB propellant, the cross section of the compacted column was distinctly asymmetrical. For experiments with a cuboid XLDB propellant, the radial compaction was much more symmetrical for 69 and 80% TMD charges. For the initially 80% TMD charge (which did not detonate), the density

**In all reported flash radiographic data, it has been necessary to insert metallic x-ray tracers into the porous column during assembly in order to obtain quantitative density measurements.

along the compacted column was essentially TMD by the time the compaction front had traveled less than a third of the charge length.

In these experiments, it was not possible to separate pre-ignition compaction from the extent of compaction associated with reaction (pyrolysis or burning) of the propellant. This situation arose because of the absence of a measure of time of ignition and because these porous columns of XLDB propellants are very compressible. However, it is important to be able to make this compaction distinction since it serves to delineate the boundary between stages 1 and 2 of the DDT process (see Table 2) and is important for modeling comparisons. Thus, there was a need to deduce experimentally the time of ignition, as well as to quantify the extent of compaction.

In order to reduce and quantify the extent of preigniton compaction, Butcher et al. evaluated various ignition systems, varying both the amount and type of ignitor components.[17,47] A weak ignitor was defined as having a dp/dt of 10^3 psi/s (6.9 MPa/s); not surprisingly, this mixture of B/KNO_3 and Mg/Teflon produced very little gas. A strong ignitor was defined as a material having a dp/dt of 10^5 psi/s (690 MPa/s) or greater. Some of these ignitors were used in DDT tests. Contrasting pressurization data from weak and strong ignition systems were presented for experiments where, unfortunately, transitions to detonation did not occur. It was observed with the weak ignitor[47] that a much higher pressurization rate resulted from confined burning than with a strong ignitor. However, with the weak ignitor, the first pressurization started at the far end of the tube (away from the ignitor) in contrast to the case with the strong ignitor. The latter showed the typical pressurization characteristics (see, for example, Refs. 35 and 36 in which pressurization begins near the ignition region and progresses downstream toward the far end of the tube). Consequently, the unusual pressurization and other DDT characteristics from weak ignition systems need to be explored further for the completeness of mechanistic insight.

A different type of pressurization was observed for a XLDB propellant confined in a steel tube, using a strong ignitor; in this case, it occurred *after* significant pressurization in the ignition region.[44] The pressure in the ignition region increased slowly and linearly with time. Accelerating pressure buildup (a necessary but not sufficient condition for DDT) was observed to occur shortly before (both in time and distance) the onset of detonation. This region of accelerating buildup was relatively far from the ignition region and led to the propagation of compressive waves both upstream (toward the ignitor) and downstream. It had been observed in an earlier study[49] that both coarse and very fine tetryl exhibited the same mechanistic characteristics as discussed here for the the XLDB propellant. This DDT mechanism (called a Mode II mechanism[50]) contrasts with the DDT mechanism discussed in Refs. 34-36 for materials such as HMX, PETN, and waxed RDX samples. The latter mechanism (called a Mode I mechanism[50]) is based upon convective combustion as the process whereby the pressure builds up rapidly in the ignition region, leading to the propagation of ever increasingly stronger compressive wavelets that reinforce the head of the compressive wave. The propagation of this compressive wave initiates hot-spot burning. Eventually, the head of the compressive wave sharpens into a shock front and the SDT portion of the process begins.

In a DDT study of HMX and tetryl in low-confinement plastic (Lexan) tubes,[50] it was evident from the flash radiographic data that the extent of CC associated with the Mode I mechanism was significantly greater than that observed for the Mode II mechanism. Hence, it was concluded that the difference between Modes I and II reflects a change from CC to compressive burning as the dominant factor for a transition to detonation. This difference is very important for DDT under low-confinement conditions. For a Mode I material, the gas pressure generated in the ignition region will have to overcome the competing factor of tube expansion. Only if the porous material is very reactive (on a relative basis) can a transition occur despite extensive deformation of the plastic tube. If the material is less reactive (e.g., a waxed RDX composition), then only the compressive burning stage (stage 4 in Table 2) can be reached by the time of tube rupture; this was the case for 97/3 and 94/6 RDX/wax confined in Lexan tubes.[51] For Mode II materials (tetryl and the XLDB propellant), the pressurization in the ignition region is much lower and the deformation of the plastic tube is reduced and less localized. The gas pressure, which results from some CC and some burning along the lateral surfaces of the radially compacted charge (at least in the case of the propellant), produces compressive wavelets that initiate compressive burning. If the hot spots are readily formed and grow sufficiently rapidly at low pressure levels, then a transition to detonation can occur because the pressure growth from hot-spot reaction is very localized[44] and it is the inertial aspects of the confinement that then are important. From these mechanistic considerations, it was evident that the extent of CC and the shock reactivity and sensitivity of a propellant in the porous state are important processes needing to be evaluated in order to quantify our knowledge of the fundamentals affecting the susceptibility of various propellant formulations to DDT.

In our work at Naval Surface Weapons Center (NSWC), a more complete understanding of CC and compressive reactivity has been obtained from the comparison of the very compressible propellants (e.g., the XLDB system) and nitrocellulose/nitroglycerin (NC/NG) ball powders. The latter materials were selected because the energy content could be varied easily, as could the particle size. The fact that these Olin ball powders were nearly spherical made them ideal for numerical modeling purposes. In addition, we acquired (from a local ammunition store) samples of two double-base (DB) powders [herein called Winchester (WC) reloading powders], which were similar in NG content to two of the ball powders, but which had been rolled such that their height-to-diameter ratio was about 0.25. The goal was to compare these nonspherical WC powders to the spherical Olin ball powders. One of these reloading powders (WC 231) proved very useful in providing us with an understanding of the *total* DDT process under low confinement, since *none* of the Olin ball powders underwent a transition to detonation in our Lexan DDT tube (25.4 mm i.d., 76.0 mm o.d.).

In order to evaluate the susceptibility of all of these NC/NG propellants to DDT, we also conducted some high-confinement (steel tube) experiments. However, we did not use the steel tube (16.3 mm i.d.) arrangement used earlier.[36] Instead, we simply replaced the Lexan tube in our low-confinement arrangement (for example, see Ref. 50) with a steel tube of nominally identical dimensions. In this manner, we increased the lateral confinement while retaining the same ignition and end clamping arrangements. This new steel tube system is called a "modified steel tube arrangement."

The summary of the susceptibility of the various Winchester and Olin propellants to undergo DDT is given in Table 3. Again using ℓ as the index of susceptibility, it can be seen that ℓ is influenced both by the NG content and the particle size. For the single-base materials (WC 140 and type A fluid), the finer material has a significantly smaller ℓ value. Consequently, one could make the general conclusion for these materials that, for a constant NG level, the finer material will be more susceptible to DDT.†† In this manner,

††A small difference in % TMD of the magnitude of 3-5% does not influence the relative susceptibilities unless ℓ values differ by less than 30-40 mm. However, when there is a large difference in %TMD, such as the WC 231 columns, a comparison of relative susceptibilities is unwarranted. Moreover, WC 231 and WC 630 differed from the Olin propellants in that they had been rolled during manufacture. In the case of WC 231, the rolling process had produced particles with cracks; consequently, the fractured surface of the WC 231 particles was unique for the materials listed in Table 3.

Table 3 Summary of DDT results for NC/NG powders[52]

Propellant	%NG	Average particle size, μm	Density, g/cm³	%TMD[a]	Tube arrangement	ℓ mm
Winchester 630	35	~650	1.10	~67.5	Mod. steel	160
Winchester 231	25	~800	0.82	~50.2	Mod. steel	121
Winchester 231	25	~800	0.82	~50.2	Lexan	210
Winchester 630	35	~650	1.08	~66.3	Lexan	>280
Olin WC 140	0.0	411	1.00	60.6	Mod. steel	>368
Olin TS 3660	12.3	714	0.94	57.6	Mod. steel	>330
Olin TS 3659	21.6	434	0.99	60.4	Mod. steel	199
Olin TS 3661	34.9	724	1.03	63.2	Mod. steel	298
Olin type A fluid	0.0	~40	0.95	57.6	Mod. steel	228
Olin type A fluid	0.0	~40	0.95	57.6	Lexan	>280

[a]Based upon voidless densities; actual particles are somewhat porous.

we can rationalize the relative ℓ values for TS 3661 and 3659. Thus, the higher energy content of TS 3661 is negated by the relatively smaller particle size of TS 3659 such that the latter is more susceptible. On the other hand, the DB material with comparable particle sizes (TS 3660 and TS 3661), the more energetic propellant is more susceptible. The same trend is noted in comparing values for TS 3659 (21.6% NG) and WC 140 (0% NG).‡‡

While data from these initial modified steel tube experiments provided insight into the influence of various properties of the powders on the tendency to undergo DDT, essentially no information was obtained concerning the various stages of their DDT mechanism. Fortunately, information for the earlier stages in Table 2 could be obtained from the Lexan tube experiments. Preignition compaction, from the burning of our B/KNO_3 ignitor, was evident in the flash radiographic data. The extent of the pre- and postignition compaction could be readily ascertained for these powders using the methodology of x-ray tracer displacement (as illustrated in Ref. 50 for very coarse HMX). Using this methodology, convective combustion (CC) was characterized experimentally by the presence of a low-density region upstream of a highly compacted region of the column. It was established[52] that CC occurred readily in porous charges of WC 140, TS 3659, TS 3660, TS 3661, WC 231, and WC 630; this pressure buildup from CC led to a loss of confinement (for all materials except WC 231) before a transition to detonation could be attained. As stated earlier, the Mode I DDT mechanism has the characteristic of extensive CC, which is the driving force for a transition to detonation. By inference, it would appear that all of the Olin and Winchester propellants listed in Table 3 would follow a Mode I mechanism. This definitely was proved to be the case for the Winchester powders and TS 3659 based upon both the low- and high-confinement studies; further work is being conducted with the other materials.

Considering that tetryl (a Mode II material) exhibited little CC,[50] it was of interest to reexamine the XLDB propellant data[44] and conduct additional experiments to evaluate the extent of CC using the x-ray displacement method of Ref. 50. The new experiments used a weaker ignition system in which a 5.0 cm³ cavity was introduced between the B/KNO_3 ignitor and the surface of the propellant. Our analyses confirmed that there was very little indication of CC with the XLDB propellant for both ignition systems. Any additional surface for combustion did not come from the interior of the charge at the ignitor end, but from the external lateral surface as a result of radial compaction and tube expansion. Consequently, with our regular ignition system, extensive preignition compaction of very compressible propellants would readily occur and eliminate (or drastically reduce) the possibility of CC.

The conclusion that columns of Mode II materials would undergo a transition to detonation *in plastic tubes* with minimal convective burning implied that these materials must be fairly shock sensitive. In addition, since the shock to detonation transition is the final stage of the DDT process, one might expect a consistent trending in DDT and SDT sensitivities for Mode I materials. Thus, using run-up distance X_D as a measure of SDT sensitivity and ℓ as a measure of DDT sensitivity, both X_D (for a given input pressure) and ℓ would be expected to increase as one went from very coarse HMX to a DB propellant to a single-base propellant (i.e., as the shock sensitivity decreased) for columns with similar permeabilities. On the other hand, for Mode II materials, it was not apparent that X_D and ℓ would track in a similar manner. This idea was based upon the observation that DDT started at the boundary between compacted and uncompacted material; the compacted region is associated with the propagation of compressive and/or shock waves. Since pressure builds up more slowly for the Mode II material, it takes longer for the shock initiation pressure level to be reached with a Mode II material (e.g., tetryl). Thus, this material would have a larger ℓ value than a Mode I material (e.g., coarse HMX) if these materials were assumed to have equal shock initiation pressures.

To check this deduction, the shock sensitivities of various propellants were evaluated using a modification of the Naval Ordnance Laboratory (NOL) large-scale gap test (LSGT). The same donor (pressed pentolite) and attenuator material (polymethylmethacrylate) of the LSGT were used.[53] The confinement of the acceptor was a shortened (~145 mm) version of our Lexan DDT tube; in this way, we evaluated the relative shock sensitivities of various propellants *in situ* (i.e., in the confinement of our Lexan DDT tube). The distance to detonation X_D was determined from a streak camera film; the input pressure P_E for the propellant was obtained from the impedance method using data on the attenuator pressure, etc., from the LSGT.[53] Hugoniots for the porous propellants were calculated by the method of Erkman and Edwards.[54] A summary of the pertinent SDT and DDT data is shown in Table 4.

To evaluate relative shock sensitivity, it should be noted that for the same shock input pressure P_E, the more sensitive material will have a smaller X_D value. For example, in the case of Olin in TS 3660 and a type A fluid powders, the latter is relatively more shock sensitive; as qualitatively expected from a Mode I DDT mechanism, ℓ for the fluid powder is less than that for TS 3660. The same trend is observed for other Mode I materials. Thus, ℓ for WC 231 is smaller than

‡‡This type of information is of interest for hazards associated with manufacturing commercial ammunition propellants.[1]

Table 4 SDT and DDT sensitivities of porous columns of energetic materials[55]

Material	%TMD	SDT		DDT	
		P_E, GPa	X_D, mm	ℓ (steel), mm	ℓ (Lexan), mm
TS 3660	57.6	1.11	F[a]	>330	—
Type A fluid	57.6	1.11	24	228	F
TS 3659	60.4	0.46	F[a]	199	F
WC 231	~50.2	0.35	72	121	210
HMX (very coarse)	61.0	0.37	39	53	100
Tetryl (coarse)	57.8	0.37	23	128	210

[a]Failed in ~135 column.

for TS 3659 because the former is more shock sensitive; at 0.46 GPa WC 231 would have an X_D value less than 72 mm. On the other hand, the HMX sample is more shock sensitive than WC 231 and, thus, ℓ for HMX is relatively smaller. A contrasting picture is observed in the comparison of the very coarse HMX with tetryl. For tetryl (a Mode II material) and HMX, we observe that the more shock-sensitive material (tetryl) has a significantly larger ℓ value. Consequently, these combined shock sensitivity data and DDT data confirm the postulated differences between the Modes I and II DDT mechanisms.

These shock initiation data also demonstrate the need to know *in situ* shock sensitivities for the porous media of interest for DDT considerations. Previously, we described how permeability would influence the susceptibility of various propellants to DDT (e.g., see Figs. 2 and 3). For the 60% TMD data of Fig. 3, it was inferred that differences between ℓ values for the cubic material and the granular (or shredded) material was dependent on the variation of permeability and, hence, on the extent of CC associated with these columns. However, a shorter ℓ value could also result from a higher shock reactivity and sensitivity for the granular material. Consequently, we also measured the relative shock sensitivity of the granular and cubic XLDB materials in the same manner associated with data in Table 4. For 83% TMD charges, the cubic material (larger particle size) was found to be significantly less shock sensitive than the finer granular material at 2 GPa.

The above shock sensitivity data thus serve to illustrate again that knowledge of both the CC process and shock sensitivity are needed to understand the susceptibility of individual propellant systems to the DDT process. Having demonstrated that CC is minimal for the very compressible XLDB propellant, the data from Ref. 44, as shown in Fig. 3, for the shreds and granules were re-evaluated. It is now concluded that the rapid increase in ℓ with increasing % TMD is completely associated with increasing P_i values and not with a reduction in the extent of CC. The importance of the permeability of the charge for gas flow is negated by our strong ignition system. (Based upon the compressibility of the propellants used by Butcher et al., as reflected in Refs. 17 and 47, the same interpretation may hold for most of the S/V range in Fig. 2.)

Future Research Considerations

These low- and high-confinement DDT experiments have indicated that significant differences in convective combustion (CC) characteristics can be observed for columns of very porous propellants. This observation is important for at least two reasons. First, there is the practial problem of whether sufficient pressure buildup can occur and drive a damaged propellant bed to detonation under low-confinement conditions. The shock sensitivity of the damaged (or granulated) propellant is an important consideration for this problem. Second, the contrasting nature of the confined burning (i.e., convective combustion at the free (ignitor end) surface of the column *and* burning along the lateral surface) must be fully appreciated and documented for the development of valid models of the DDT process. Consequently, these considerations point to the need for many complementary experimental studies. These complementary studies, moreover, include a wide range of disciplines such as thermal decomposition, ignition, material science, compressive reaction, etc. However, since the boundary conditions of these complementary expeiments will, in large part, differ from an actual DDT experiment, additional data are also needed from DDT experiments to characterize the nature and extent of "in-depth" reaction (i.e., as opposed to reaction seen on the lateral surface). Some examples of these complementary experiments are 1) the study of pyrolysis and reaction leading to ignition and transient combustion[56]; 2) quasi-static compaction studies[57]; 3) the study of compressive ignition and reaction[58]; and 4) the measurement of *in situ* pressure histories in DDT studies.[59] The latter work is an example of the more sophisticated intrumentation that is also needed to define more accurately the temporal boundaries of the various DDT stages for a proper correlation of the experimental data with predictions from numerical models.

Finally, from the hazard assessment area, the experimental data from Lexan tube studies suggest that the current Lexan tube configuration is not the best arrangement to assess the tendency to DDT during motor malfunctions. That is, the Lexan tube experiments have shown extensive radial compaction of very compressible porous propellant columns because of the preferential flow of gas along the interface between the Lexan tube and the propellant; this apparently occurs irrespective of the strength of the ignition system. It has not been demonstrated that this radial compaction occurs in the DDT process for a damaged grain. Hence, other experimental configurations for DDT tube experiments must be considered. It would appear best to conduct experiments in which damaged propellant is generated *in situ* under conditons representative of dynamic breakup.

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