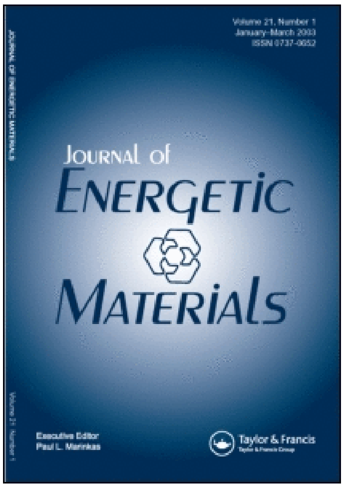

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EFFECT OF PARTICLE SIZE ON THE SHOCK SENSITIVITY OF POROUS HE

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Abstract

Literature data show that in most gap tests, coarse porous high explosives (HE) seem more shock sensitive than fine whereas in most wedge tests, the reverse is true. It is proposed that gap tests measure threshold for ignition, and that the reversal occurs because the time to ignition is shorter and the time of buildup of chemical reaction is longer for the coarse than the fine material. In other words, for any pair of fine and coarse HE in any specific experiment, there is a pressure (P_r) at and above which ignition for the fine and coarse is simultaneous. At and above this pressure, the finer material appears more sensitive than the coarse.

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INTRODUCTION

There seems to be much confusion and many contradictions in the current literature on the effect of particle size on the shock sensitivity of explosives. This paper is the first phase of a study of available literature data in an attempt to eliminate such confusion. It is restricted to pure, pressed explosives.

About forty years ago, shock sensitivity was assessed by gap tests.[1] The experiment consisted of a standard donor explosive separated from the test high explosive (HE) by an attenuator material (the gap), the thickness of which was varied until the test explosive detonated in 50% of the trials. That 50% point thickness could be translated into pressure at the end of the gap and pressure entering the explosive (initiating pressure P_i) provided that a test calibration was made and Hugoniot data were available for both gap material and HE.

More recently, with the invention of the wedge test[2], it has become fashionable to measure shock sensitivity by the run distance required for a specified initial shock wave to cause detonation of test HE.

EARLIER RESULTS & POSSIBLE EXPLANATION

In 1961 Campbell et al.[3] reported from wedge test data that fine TNT (20 - 50 μm) was more shock sensitive than coarse (200 - 250 μm). But in 1963, Dinegar et al.[4] reported the gap test data of Figure 1 in which they showed shock sensitivity decreasing with increasing specific surface and therefore decreasing particle size of 0.95 g/cm^3 PETN. Moreover, they reported that comparable experiments on PETN at 0.75 g/cm^3 and at 1.4 g/cm^3 had shown the same trend. Since then, it has been thoroughly documented that gap test shock sensitivity values show the coarser HE to be more sensitive than the finer. (However, the test must be well designed. A very coarse explosive tested in a very small diameter gap test can produce weird results.[5])

To resolve the contradictions, more detailed information about the effect of shocking the explosive is required. Scott[6] provided some of this in 1970. He used the design of the NOL small scale gap test to supply various strength shocks to the same acceptor HE. He used the depth of the resultant steel plate dent as a measure of the shock induced reaction. Figure 2 shows some of his results on fine (through screen 325 and retained in pan) and coarse tetryl (through screen 40 and retained on screen 60); both charges were compacted to 1.50 g/cm^3. As Figure 2 shows clearly, the stages of ignition and of buildup of reaction are differentiated in this experiment. Ignition is signalled by the first appearance of a dent in the witness plate and the rate of reaction is indicated by the slope of the subsequent curve. Hence, in the case of tetryl, as well as RDX and PETN, Scott found that the coarser particles ignite more easily, i.e., at lower initial shock pressure, than the fine. But, once ignited, the fine particles show more rapid buildup to detonation than the coarse.

In 1976, Howe and his colleagues at BRL[7] used projectile impact to provide the stimulus, and measured free surface velocity on the opposite side of the HE target; this is approximately twice the particle velocity of the shocked explosive and hence a measure of the degree of induced reaction. Figure 3, free surface velocity u_{fs} as a function of shock pressure, shows the data for fine (58 μm) and coarse (254 μm) TNT compacted to 1.55 g/cm^3. It shows the same differentiation between ignition and buildup reported by Scott. Here ignition is the first departure of the data from the straight line response to be expected when an inert solid of the same impedance as the HE is shocked. Again the coarse material ignites at lower pressure than the fine, but the latter, once ignited, shows much more rapid reaction.

From the above data, a simplified schematic of the degree of reaction as a function of shock pressure is shown in Figure 4. The curves for fine and coarse explosives cross at P_r before detonation is achieved by either charge. At $P < P_r$, the ease of ignition determines the response, and the coarser material appears more sensitive than the fine. At $P \geq P_r$, ignition will be simultaneous for the coarse and fine; hence, rate of reaction predominates and the fine appears more sensitive than the coarse.

This suggests that gap tests carried out under conditions favoring propagation of steady state detonation and used to measure the lowest pressure leading to detonation in 50% of the trials are a measure of the minimum pressure for ignition. Seely[5] concluded this was the case from results he obtained on high porosity charges in 1963. Now there is some stronger evidence.

Ignition requires some reaction. Consequently separating ignition from combustion is impossible. In studies of ignition by radiation[8], curves are obtained in a log-log plot of time vs energy flux as shown in Figure 5. There is a region between the curve for first light and the curve for sustained ignition (go/no-go); hot spots producing light will fade out in that region. Similar detailed study has not been carried out for shock ignition of many HE--probably because the greater practical interest has been in initiating detonation rather than causing ignition. For the present study, therefore, ignition will be defined according to Liddiard and Jacobs[9]; that is also the definition used by Howe et al. for Figure 3 data.

Liddiard[9] developed a modified gap test with a short, unconfined acceptor on which he measured free surface velocity as a function of shock pressure to determine a threshold of burning. For the four pressed explosives he tested, the threshold for burning was nearly equal to the threshold for initiation of detonation measured in the NOL large scale gap test (LSGT). That test has a longer and a confined acceptor. However, Tasker[10], who developed a test based on Liddiard's, showed that the threshold pressure for initiating burning was not affected by confinement or by acceptor thickness, whereas the threshold pressure for initiating detonation was affected by both. Tasker considered that the burning threshold was the most important parameter of shock sensitivity because any sufficiently large charge will detonate when shocked even by the low amplitude shock required to initiate burning.

Unfortunately, most of Tasker's data are for cast charges. The one exception is 95/5 TATB/Kel F at ρ_o = 1.91 g/cm^3. This is a pressed explosive although not a pure one. It is of interest that the LSGT value for 96/4 TATB/Kel F at 1.89 g/cm^3 is about 7.0 GPa[11] at the end of the 50% point gap or about 8.5 - 9.0 GPa entering the explosive. This is about the threshold pressure for burning measured by Tasker. Because of this and the preceding discussion, it seems likely that the NOL LSGT, and most gap tests, measure a critical pressure for the sustained ignition which can grow into detonation. In other words, in comparing two particle sizes of the same porous HE, the low amplitude gap tests are in pressure ranges below P_r of Figure 4. Obviously P_r will differ for each pair of particle sizes chosen as well as for each HE, each porosity, and each change (including dimensions) of each experimental test design.

MOST RECENT DATA

It is now customary to speak of long and short duration shock sensitivities. However, different sensitivities to ignition and reaction buildup after ignition seem more likely to be responsible for reversal of sensitivity ratings. Rather than long and short duration shock sensitivities, low and high amplitude shock sensitivities seems more accurate. To be sure, most gap tests of porous HE use low amplitude, long duration shocks, whereas most foil flyer impact tests use high amplitude, short duration shocks. Nevertheless, in the proper pressure range, flyer impact and gap test can give the same relative sensitivity rating. The reversals with pressure range can be demonstrated by examples from the work on HNS and TATB, two explosives for which shock sensitivity has been most extensively studied.

HNS

In 1981, Schwarz[12] studied three batches of HNS: HNS-I (1.59 m^2g), HNS-SF (2.56 m^2/g) and HNS-HF (10 m^2/g). With an 1.02 mm diameter flyer and 1.60 g/cm^3 charges, he obtained the results shown in Figure 6; the 50% probability of detonation initiation ranged from 7.6 GPa for the coarsest to 6.2 GPa for the finest. In this range, the fine was more sensitive than the coarse. He also showed (Figure 7) that $P^{2.4}\tau$ = constant for durations of 0.01 to 0.10 μs, but only for that short duration range.

Then in 1984, Setchell[13] published a study of the shock sensitivity of HNS-I (2.1 m^2/g) and HNS-FP (8.2 m^2/g), both pressed to 1.60 g/cm^3. He used both sustained shocks and those of 0.19 μs duration, and measured the velocity profiles they produced in the HE. In both cases, he was amazed to find that his measured wave forms showed the coarse HNS-I more shock sensitive than the finer HNS-FP. Figure 8 shows his results for the 0.19 μs pulses at his highest pressure of 4.0 GPa. In this experiment, an order of magnitude difference in the pulse width did not reverse the relative sensitivities.

TATB

In 1981, Honodel et at.[14] reported both flyer impact and gap test investigation of the insensitive HE, TATB. By varying the thickness (and thereby the velocity and impact pressure) of the 25.4 mm diameter flyers, they determined threshold velocities required to initiate detonation of 25.4 mm dia cylinders of 1.80 g/cm^3 TATB; cylinder lengths of 10 - 19 mm gave the same results. This LLNL group used eight different lots of TATB: ultrafine (4.3 and 4.6 m^2/g) and coarser samples (0.3 - 0.7 m^2/g). Table 1 contains a portion of the raw data obtained. As they show, for the thinnest flyer (0.051 mm) and highest pressure, the fine material is more sensitive than the coarse. It is not until the flyer thickness becomes 0.5 mm that the coarse TATB is relatively more sensitive than the fine.

The LLNL group also ran gap tests on the same lots of TATB. For this, they used the Pantex gap test shown in Figure 9. Figure 10 shows the results as 50% gap thickness vs charge density; they show that, according to this test, the coarse TATB is more sensitive than the fine. In other words, the gap test ranks the two with the same relative sensitivity as that found in the lower pressure range by flyer impact e.g., with the 1.27 mm thick flyers.

The flyer impact threshold velocities were used to obtain threshold pressure-time data. These are shown in Figure 11 for one of the coarser samples of TATB at 1.70 g/cm^3. The solid lines are fits to the data; the dashed are for constant flyer kinetic energy normalized to the maximum flyer velocity. As the curves show at lower velocities and pressure, "data deviate substantially from the critical energy criterion". In the log-log plot of P vs τ, the deviation for 1.70 g/cm^3 coarse TATB occurs at 6.0 GPa. For the range 6.0 - 25.0 GPa, the relation $P^2\tau$ = constant is followed.

TABLE 1

Raw Data from Thin Flyer Shock Initiation Experiments on TATB (Reference 14)

Explosive, Density Mg/m^3	Flyer Thickness mm	Threshold Velocity km/s
B-226	0.051	5.4 ± 0.2
1.80	0.127	3.9 ± 0.2
(58 μm)	0.254	2.9 ± 0.2
	0.508	2.45 ± 0.2
	1.27	2.22 ● 0.08
B-592	0.051	4.1 ± 0.2
1.80	0.127	3.2 ± 0.2
(~9 μm)	0.254	2.6 ± 0.2
	0.508	2.6 ± 0.2
	1.27	2.6 ± 0.2

Seitz[15], in 1984, carried out wedge tests on three samples of TATB for which he gave the sieve analyses but no specific surface areas. Samples 1 and 2 were relatively coarse; 3, very fine. He used sustained pulses to obtain Pop plots for 1.80 g/cm^3 charges, and also carried out a few experiments with short (0.02 μs) pulses. His results are shown in Figure 12. As in previous work[14], the coarse samples were indistinguishable from each other but differed definitely from the very fine. Pop plots cross at a P_r of about 10.5 GPa, above which the very fine is more sensitive and below which it is less sensitive that the coarser TATBs. The short pulse data (points above the curves) were taken in the range P > 10.5 GPa. They do not change the relative sensitivity (fine greater than coarse) in this range although they do require a higher threshold pressure as might be expected. The increase in required pressure is least for the very fine material as might also be expected. Nevertheless, the relative rating is fine more sensitive than coarse for both long and short duration shocks at pressures greater than 10.5 GPa.

In 1985, Grief et al.[16], a group from AWRE, reported on studies on TATB wedges supplied by Wackerle and Seitz (LANL). For this study, an electric gun and flyers (plates and cylinders) were used to produce shocks of 11.0 - 26.0 GPa for 0.08 - 0.11 μs duration. The TATB used was fine (4.5 m^2/g) and coarse (0.5 and 0.7 m^2/g), and the wedges were at 92% TMD or ρ_o = 1.783 g/cm^3 as compared to 1.801 g/cm^3 in Seitz's work.[15] Figure 13 shows the AWRE results and compares them to those of Seitz. It is of some interest that for TATB, an insensitive HE, at these high pressures the 0.1 μs pulse is effectively a sustained shock for production TATB; the separation of the two curves seems to be of the order of magnitude of the experimental scatter. The curve for the superfine - 0.1 μs pulse is slightly lower whereas Seitz found no difference between the production grade and the superfine TATB, as noted in Figure 13.

The most important feature of Figure 13 for the present analysis is that the curve (0.1 μs pulse) crosses Seitz's curve for the ultrafine (sustained pulse). Grief et al. attributed this to the fact that their highest pressure, smallest run distance to detonation (x*) point is much less accurate than the rest of their data, and suggest that the estimate of x* ~ 0.4 mm is in fact an overestimate. That may be true and, if so, would tilt the curve to agree better with the slope of the corresponding Seitz curve. Another possibility is that the TATB used was of a different particle size distribution. As Seitz noted, an extremely fine TATB is required to demonstrate a particle size effect. But if we assume that the ultrafine TATB supplied by Wackerle and Seitz is the same that Seitz[15] used at about the same time, and also that the designs of the two sets of experiments were such that the same numerical pressures are equivalent, an obvious conclusion can be drawn. The AWRE data show no reversal of sensitivity between fine and coarse TATB because there were no pressures below P_r = 10.5 GPa. Hence, at the pressures $P > P_r$ used, the ultrafine TATB always appears more sensitive than the coarse.

PROPOSED THEORIES

Honodel et al.[14] suggested a qualitative explanation of the observed reversals of relative sensitivity with particle size on the basis of hot spots resulting from void collapse. They argued that void size would be proportional to particle size and that large hot spots survive longer than small, the energy of which dissipates very rapidly. Hence at lower pressures, the coarser material would ignite more easily. However, at much greater pressures, hot spots of all sizes would become hotter and the total number of hot spots would predominate over hot spot size in determining the time of reaction. Hence, in this range the finer HE would appear more sensitive than the coarse.

Hayes[17] also used the concept of hot spots formed by pore collapse to build a numerical model with which he predicted that a fine grained 91.2% TMD HNS will react more rapidly (be more sensitive) than a coarse grained HNS exposed to the same shock. His data from impact of HNS on fused silica showed this relative sensitivity between 21 μm and 37 μm particle sized HNS. However, the data of Setchell[13] showed that this result is not generally true.

Inasmuch as most of the data have been from charges near 90% TMD, it would seem likely that shear processes have contributed to hot spot formation as much as or more than void collapse. The role of shear and viscoelastic work is being investigated by a number of people in the field.[18-21] Of these investigators, Frey[19] has made the most progress toward developing a numerical model.

In contrast to the Hayes numerical model developed for HNS, Cochran and Tarver[22] combined the ignition and growth reactive flow model of shock initiation and detonation with Cochran's statistical treatment of hot spot formation and subsequent reaction. Among the assumptions made is that the initial hot spot size in production (standard grind) TATB is 1 μm; in SF TATB, 0.14 μm, and that the maximum volume of hot spots equals the initial void volume. With this model, the computed wave forms matched closely those measured with manganin gages.

They also demonstrated quantitatively "the validity of the classical argument that coarse particles ignite more readily than fine ..., but fine particles react faster once ignited". However, for a 7.5 GPa shock, after 2 μs, there seemed to be little indication of different distance to detonation (x*) for the two samples, and it was remarked that other works find no difference in x* values. Reference 9 of Reference 14 identifies the SF TATB as B-474. In Reference 14, TATB B-474 was compared to production TATB B-226, both at 1.80 g/cm^3. For the 0.051 mm flyer, Honodel et al. found the following threshold velocities for initiating detonation:

B-226	5.4 ± 0.2 km/s
B-474	5.35 ± 0.2

In other words, in this high pressure region, the particle size effect on shock sensitivity was negligible as it was also on the two coarser TATB samples investigated by Seitz.[15] Incidentally, although B-474 contained many more smaller particles than B-226, its specific surface area (0.513 m^2/g) was less than that of the production TATB (0.539 m^2/g). And as in Seitz's work, it is possible that an order of magnitude difference in particle size would be necessary to show a difference in distance to detonation (x*) caused by change in particle size. It is possible that the common sensitivity of the coarser samples is caused by a reduction in particle size during pressing of the charges such that the average particle size in the compacted charges is the same. A reduction of the original particle size, after pressing to charge density, has been observed and reported by several investigators including Setchell[13] who found no increase in surface area when he compacted the ultrafine to the same density. In addition, Elban et al.[23,24] used the Tinius Olsen Machine to compact a bed of #20 sieve cut HMX. They found widespread fracture of particles at a stress as low as 1.1 MPa.

Finally, I pose the possibility that Figure 4 can be further generalized. As used, it shows a difference in required energies for ignition of fine and coarse samples of the same material. But it might also represent a single sample capable of undergoing two different reactions requiring different activation energies.

SUMMARY

Ignition can be distinguished from the subsequent rate of buildup of reaction in shocked porous HE.

When tested at relatively low pressures and long durations as in gap tests or by the impact of thick flyer plates, coarse porous HE appears more shock sensitive than fine. However, when tested at relatively high pressures (sustained or short duration) fine HE seems more shock sensitive than coarse. Most wedge tests have been carried out in the high pressure regime and there they consistently show the fine HE more shock sensitive than the coarse. A reversal of the relative rating is seen only when the large range of pressure down to and including very low amplitudes is used. The reversal found for TNT, HNS, and TATB is probably a general phenomenon. It can be explained in terms of ease of ignition and rate of subsequent buildup of reaction to detonation.

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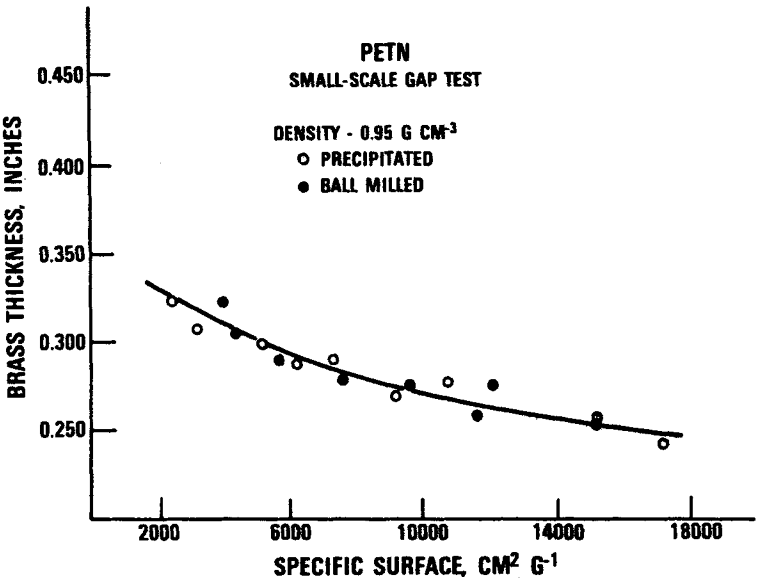


FIGURE 1. LANL SMALL SCALE TEST RESULTS FOR SAMPLES OF PETN OF VARIOUS SPECIFIC SURFACES. (REF. 4)

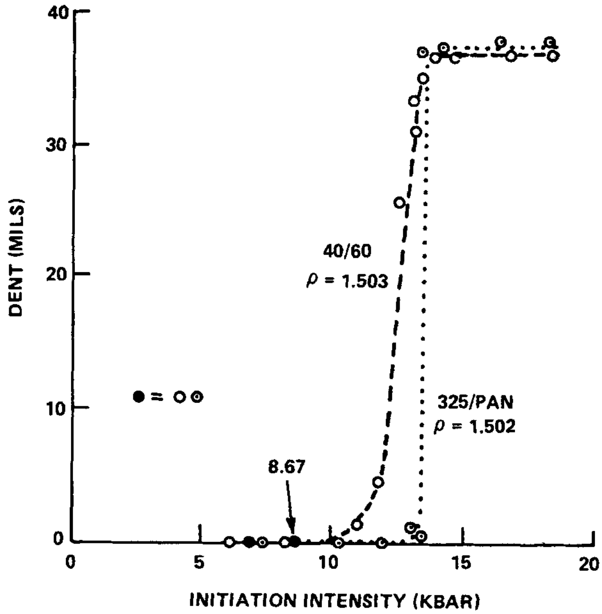


FIGURE 2. OUTPUT OF TETRYL OF TWO PARTICLE SIZES (40/60 AND 325/PAN). (ρ = DENSITY IN gm/cc) (REF. 6)

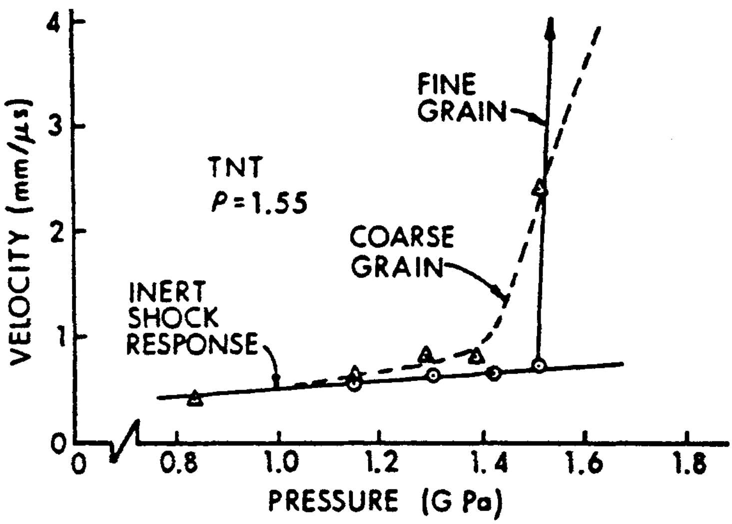


FIGURE 3. FREE SURFACE VELOCITY VERSUS INPUT PRESSURE FOR HIGH DENSITY TNT. (REF. 7)

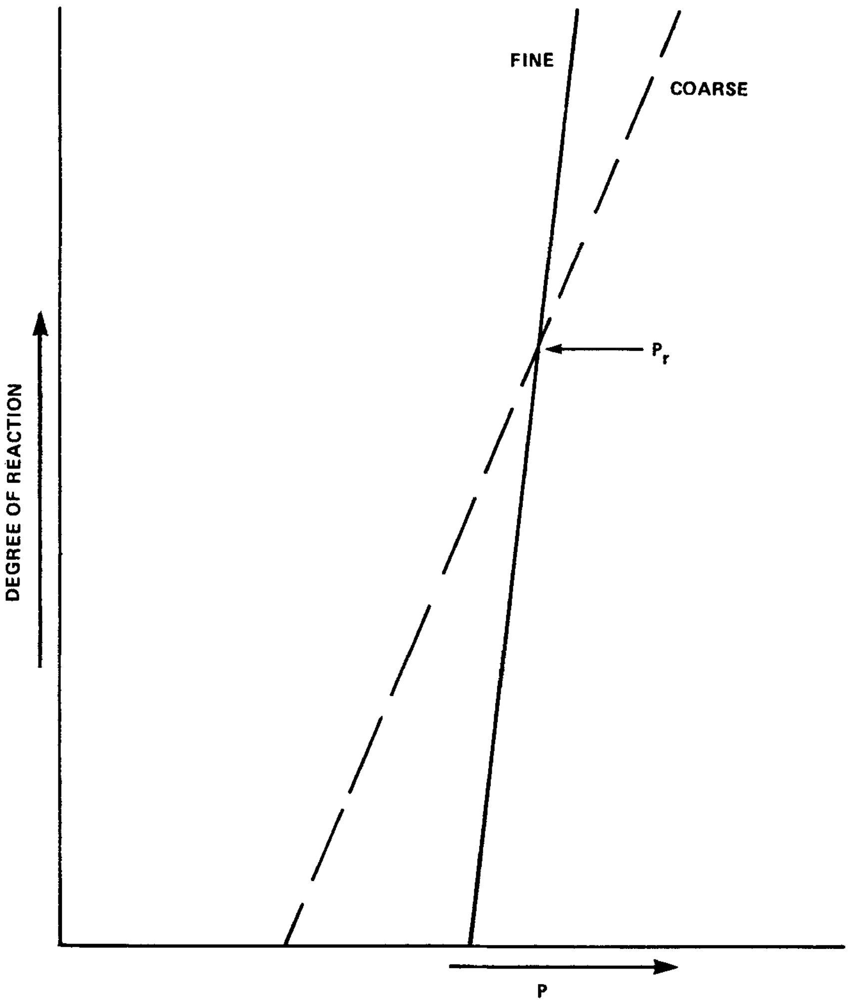


FIGURE 4. GENERALIZED CURVE FOR SHOCKED EXPLOSIVES AT A CONSTANT DENSITY

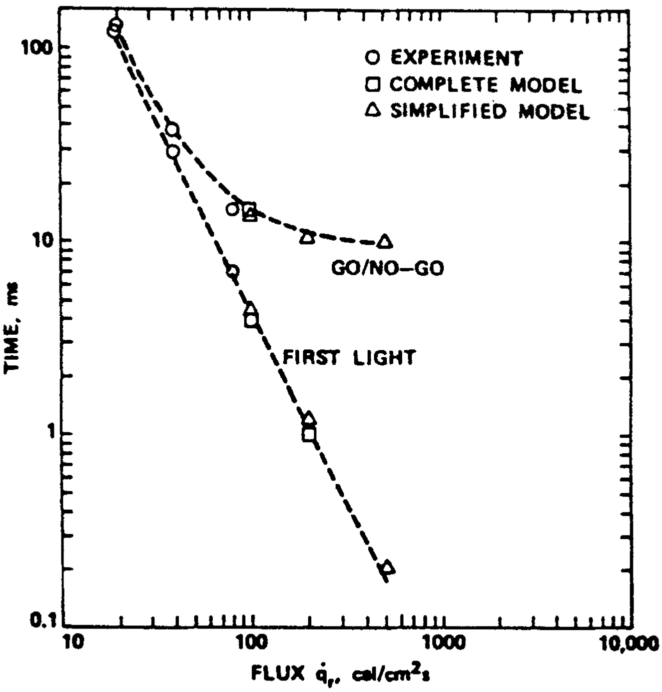


FIGURE 5. THE FLUX-TIME IGNITION HISTORY FOR HMX. THE EXPERIMENTAL DATA OBTAINED FROM XENON ARC IMAGE EXPERIMENTS. (REF. 8)

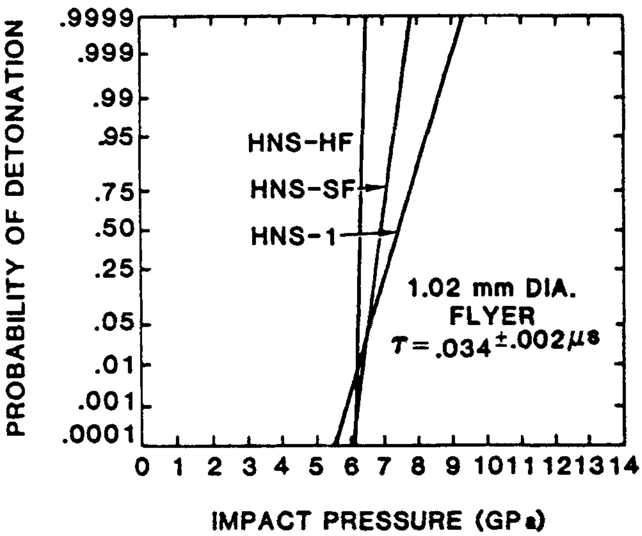


FIGURE 6. EFFECT OF MORPHOLOGY ON SENSITIVITY. (REF. 12)

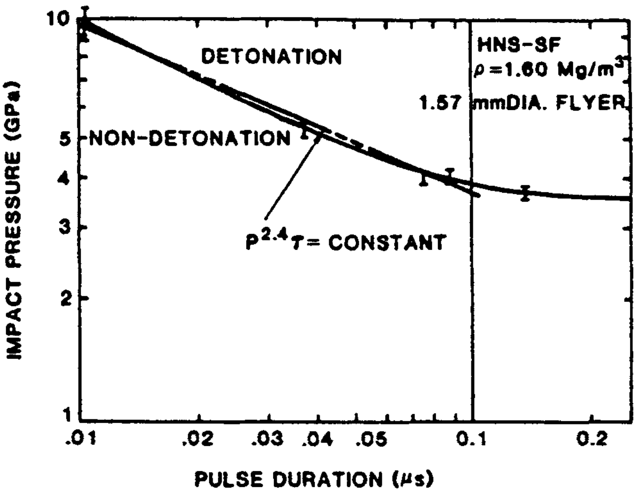


FIGURE 7. EFFECT OF PULSE DURATION ON INITIATION SENSITIVITY. (REF. 12)

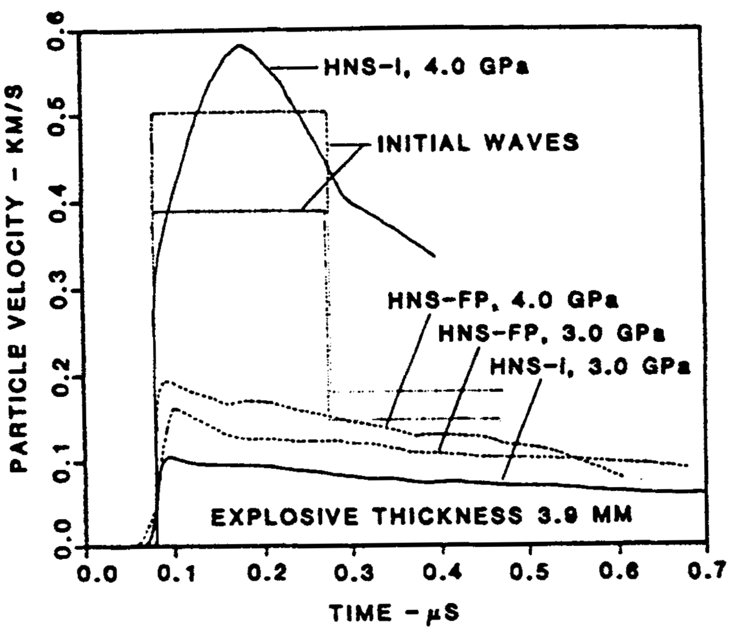


FIGURE 8. PARTICLE VELOCITY HISTORIES RECORDED AFTER PROPAGATION THROUGH 3.9 mm OF EXPLOSIVE. (REF. 13.)

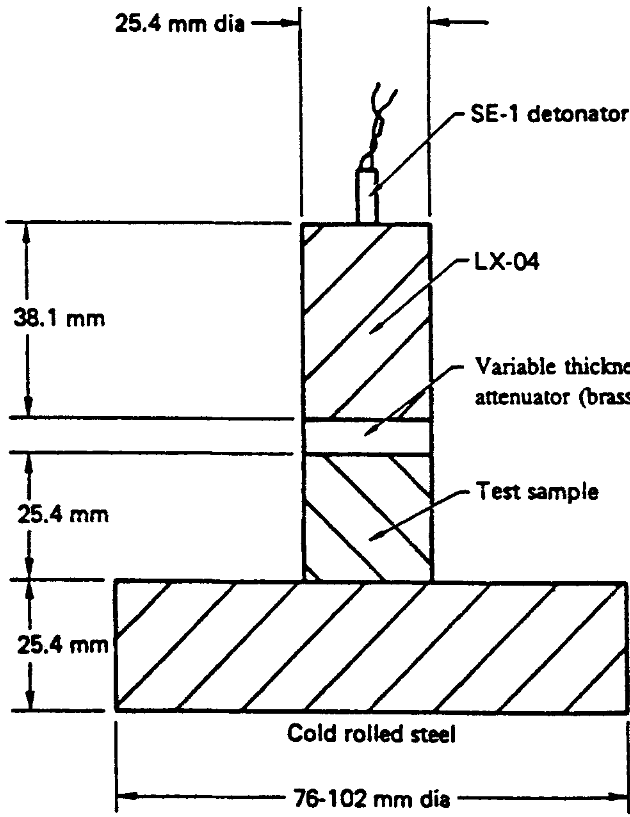


FIGURE 9. SCHEMATIC DRAWING OF PANTEX ONE-INCH GAP TEST. (REF. 14)

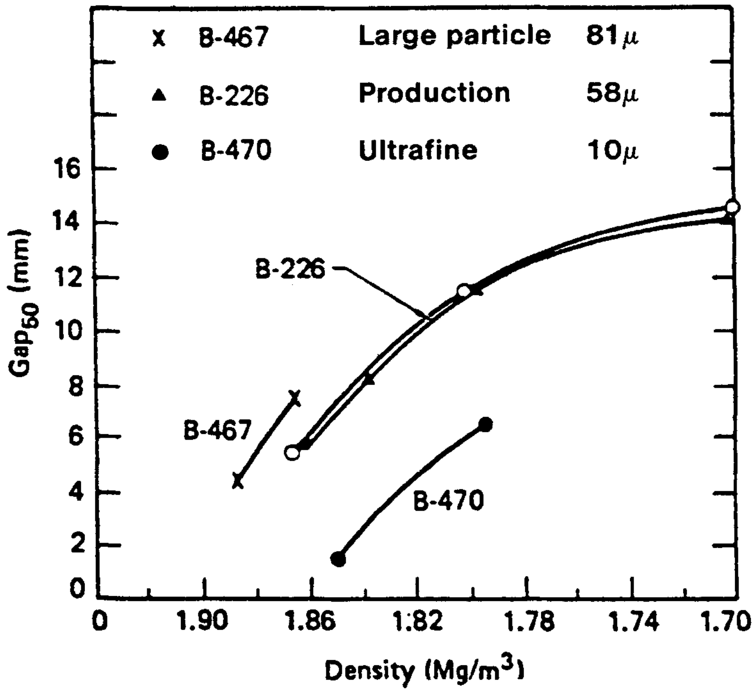
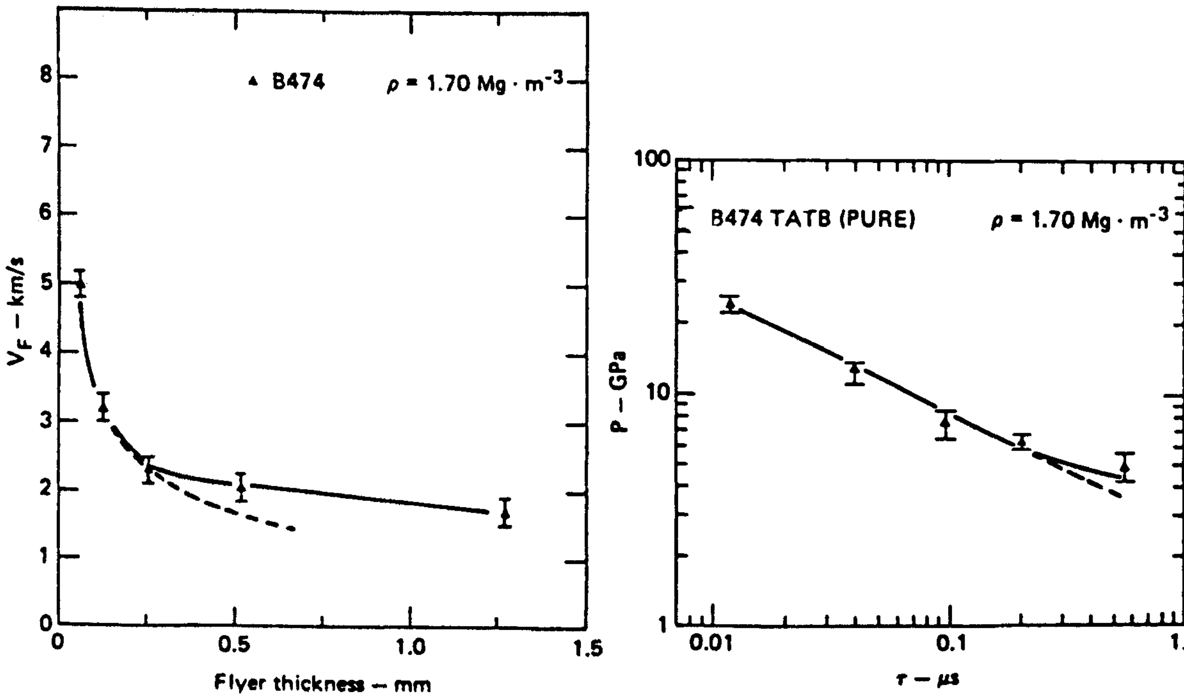


FIGURE 10. GAP TEST DATA. (REF. 14)



a. THIN FLYER THRESHOLD DATA

b. P-τ RELATIONSHIPS FOR TATB

FIGURE 11. IMPACT DATA. (REF. 14)

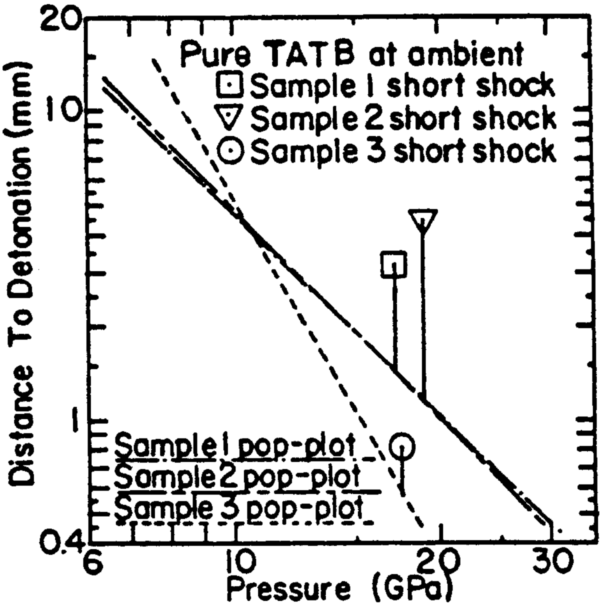


FIGURE 12. SHORT-DURATION SHOCK DATA FOR PURE TATB, SHOWING THE EXTENSION OF DISTANCE TO DETONATION OVER THE SUSTAINED-SHOCK POP PLOTS. (REF. 15)

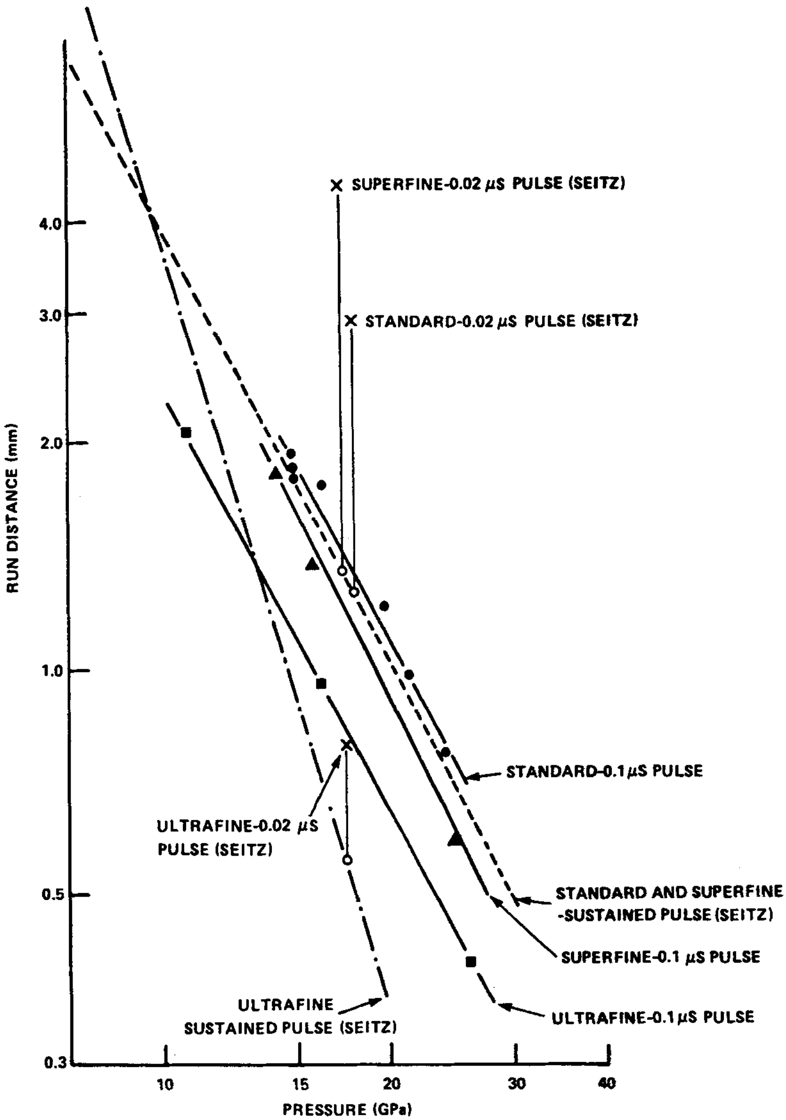


FIGURE 13. DATA OF GRIEF ET AL. COMPARED WITH THAT OF SEITZ ON THE SAME EXPLOSIVES. (REF. 16)