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TITLE: IN-SITU STUDY OF THE CHEMICALLY DRIVEN FLOW FIELDS IN INITIATING HOMOGENEOUS AND HETEROGENEOUS NITROMETHANE EXPLOSIVES

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IN-SITU STUDY OF THE CHEMICALLY DRIVEN FLOW FIELDS IN INITIATING HOMOGENEOUS AND HETEROGENEOUS NITROMETHANE EXPLOSIVES†

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Electromagnetic gauging has been used to make in-situ measurements of particle velocity and impulse at five Lagrangian positions in nitromethane (NM) during gas-gun-driven, shock-to-detonation experiments. Homogeneous initiation experiments were conducted using NM that was chemically sensitized (using an organic base) and heterogeneous initiation experiments were done with physically sensitized NM (using silica particles). In the homogeneous initiation experiments, some of the features we observe are consistent with the classical homogeneous initiation model, however, our measurements show that the superdetonation does not form immediately after an induction time. Considerably behind the initial shock, reaction causes a wave to build up over a discernible length and this wave evolves into a superdetonation which catches the initial shock. In the heterogeneous initiation experiments, the waveforms indicated that wave growth occurs primarily in the shock front, similar to earlier observations in other heterogeneous explosives.

† Work performed under the auspices of the U.S. Department of Energy

INTRODUCTION

The conclusions drawn in the classical studies of Campbell, Davis, Travis, and Ramsay[1,2] concerning the initiation process in homogeneous and heterogeneous explosives depend primarily on inferences drawn from shockwave trajectory measurements. Essentially all of our intuitions concerning the initiation of detonation in homogeneous condensed-phase materials have their origin in such measurements; some of these ideas remain controversial. Although considerable in-situ measurements have been made on condensed-phase heterogeneous materials, little work has been done on heterogeneous materials in which the inhomogeneities are controllable. It is increased understanding of the evolutionary wave processes in homogeneous and controllable heterogeneous explosives that this research addresses.

Classical homogeneous initiation can be illustrated using a time-distance diagram (first drawn by Chaiken[3] and later by Campbell, Davis and Travis[1]) as shown in Figure 1. The explosive is shocked and, after an induction period (which depends on the initial shock pressure), a thermal explosion occurs at the explosive/driver interface. After the explosion, a superdetonation runs forward into the precompressed explosive. Eventually, the superdetonation overtakes the initial shock and then decays

to a steady ZND detonation. It seems reasonable that establishment of the superdetonation is more complex than this picture. Recently Kapila et al.[4,5] have studied the initiation process theoretically (in gaseous systems) and have obtained both analytical and numerical evidence of a more complex series of steps that lead to the superdetonation. Their ideas lead to modifications of the classical homogeneous model as shown in Figure 2. They have found an unsteady process in which the superdetonation develops from a weak detonation that, in turn, results from the thermal explosion at the input boundary. The weak detonation slows down and evolves into a superdetonation which then overtakes the initial shock. In this work we are looking for experimental evidence of how the superdetonation develops. This has been done by making in-situ measurements in condensed-phase homogeneous nitromethane (NM) explosives.

Studies of initiating NM have been conducted by a number of researchers in the past 30 years. Chaiken[3] (1957) studied shock-initiated NM and developed the time-distance diagram (shown in Figure 1) that describes the presently accepted model of homogeneous initiation as described above. Campbell, Davis, and Travis[1] (1961) produced considerable evidence that corroborated, amplified, and extended Chaiken's conclusions. Dremin, et al.[6] (1965) reported embedded electromagnetic gauge measurements that also support the ideas of the model;

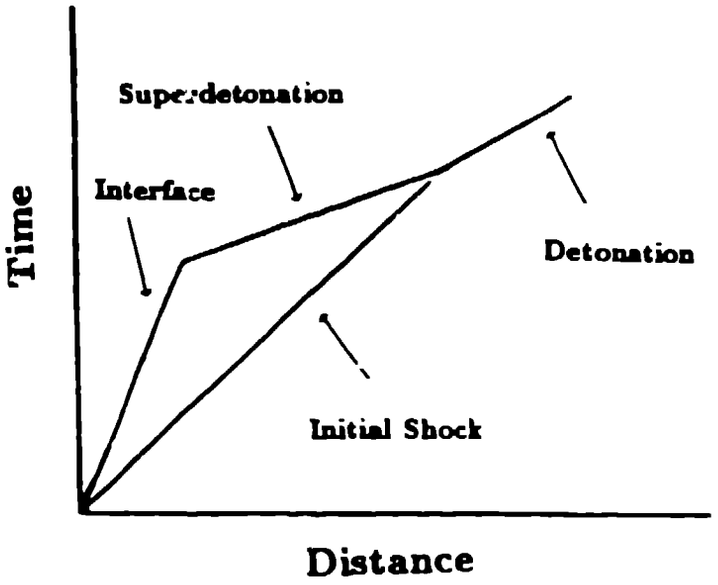

FIGURE 1. TIME-DISTANCE DIAGRAM FOR THE CLASSICAL HOMOGENEOUS EXPLOSIVE INITIATION PROCESS.

unfortunately, they were only able to make one measurement per experiment and because of inconsistencies in the explosive drive it was impossible to get a clear picture of the wave evolution process. Berke, et al.[7] (1970) studied NM, along with ten other liquid explosive materials, looking for clues as to the chemistry involved in the initiation process. Although there were definite clues, this work was apparently stopped before in-depth understanding developed. Hardesty[8] (1976) reported gun driven experiments in which a velocity interferometer system was used to measure the input boundary velocity and compare it to emitted light information obtained on the same experiments. Walker and Wasley[9] (1970) did very large scale experiments to study the initiation process at lower pressures than in the previous studies. Walker[10] (1979) also reported work in which initiation sensitivity was measured (at the same low input pressures) as a function of diethylenetriamine addition to the NM.

In all these studies, very few successful in material measurements have been made to measure the evolutionary buildup process. Since all the experiments involve explosive driver systems (with the exception of those by Hardesty), the inputs to the NM were not constant and sometimes ill defined. The measurements usually made were time histories of shock trajectories, motion of boundaries, or emitted light, the latter being the most

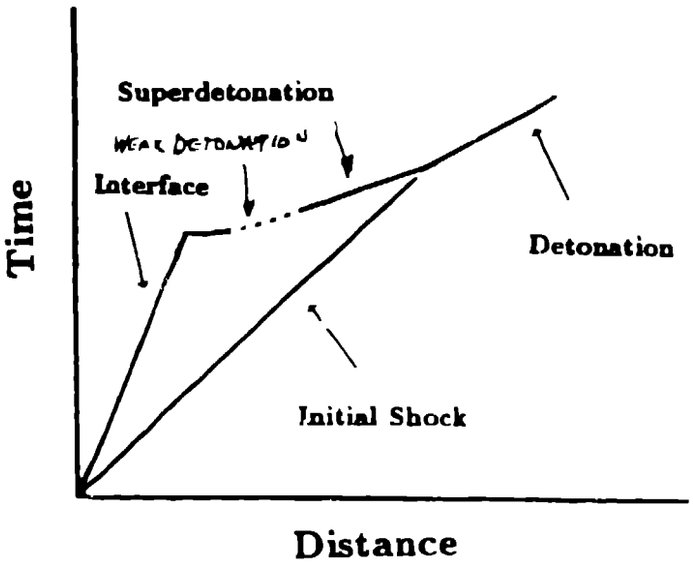

FIGURE 2. TIME-DISTANCE DIAGRAM OF KAPILA, ET AL.[5] FOR GASEOUS EXPLOSIVE INITIATION, SHOWING THE DEVELOPMENT OF THE SUPERDETONATION.

common. These studies all generally support the classical homogeneous initiation model but details of the buildup process remain unknown. In addition, conflicts exist in superdetonation velocity measurements and in thermal-explosion-time data, depending on how the measurements were made and by whom. It was to clarify some of these conflicts and to begin to understand the details of the buildup process that brought us to use a multiple, embedded particle velocity and impulse gauge technique to look again at this evolving reactive process.

A heterogeneous initiation process[2] is different from the homogeneous case in that wave growth occurs at the front, as well as behind the front. Such a wave has a relatively orderly growth to detonation, rather than the abrupt changes caused by a superdetonation. Little is known about the mechanisms that produce the wave growth other than that shock induced hot spots develop at the inhomogeneities. This makes the explosive much more sensitive than it would be without the inhomogeneities. To date, the size and nature of the inhomogeneities have not been sufficiently controllable to allow detailed understanding.

Engelke[11] and Engelke and Bdzil[12] have made detailed studies on simple prototypical NM based condensed explosives concerning how chemical and physical sensitization of the NM affect steady two dimensional

detonation. It seems reasonable to expect corresponding sensitization effects in the initiation regime. We have studied the initiation process using these *same* materials. Our experiments are superior to earlier initiation work in two important respects: (1) the chemically driven flow fields were measured directly by use of in-situ, multiple electromagnetic gauges and (2) chemical reaction was induced in the test materials by use of gas-gun-driven projectiles allowing precise control of the initial fluid-dynamic state and following flow.

In this paper we describe the experimental setup, followed by a discussion of the homogeneous experiments and then a discussion the heterogenous experiments.

EXPERIMENTAL SETUP

The liquid explosive NM was our homogeneous prototype; it was chemically sensitized to allow initiation with the available gas gun. (Chemically sensitized NM has long been known to be more sensitive to initiation than neat NM.[13]) The heterogeneous materials were produced from NM by controlled addition of solid heterogeneities to gelled NM. Since the homogeneous and heterogeneous explosives are chemically similar, results obtained from the two cases help to separate the chemical from the physical effects. Addition of heterogeneities allows more rigorous control of hot-spot characteristics (e.g., size, spatial distribution, shape) than is feasible with standard (e.g., pressed) explosives.

The homogeneous, chemically sensitized NM was made by adding 5 wt% of the liquid organic base diethylenetriamine ($NH_2(CH_2)_2NH(CH_2)_2NH_2$ hereafter called DETA) to liquid commercial-grade NM to make a 95/5 wt% NM/DETA material. The heterogeneous material was made by gelling commercial-grade NM (with guar gum) to which silica beads had been added; the final composition was 92.75/6.0/1.25 wt% NM/silica/guar gum. For details concerning the materials and how they were prepared see References 11 and 12.

All the experiments were done using an 8 m-long, 72 mm-diameter bore single stage gas gun capable of projectile velocities up to 1.43 mm/μs. An electromagnet was installed in the gun target chamber to provide the magnetic field ($\approx$ 825 gauss) required for the electromagnetic gauging setup.

Electromagnetic gauging in shock experiments was first reported by Zaitsev, et al. in the Soviet Union in 1960.[14] The system that we are using was developed by Vorthman and Wackerle in the early 1980's.[15] The gauge package includes, in addition to particle velocity gauges, impulse gauges which were first reported by Young, Fowles, and Swift.[16] (We refer to this gauge package as an MIV gauge, for Magnetic Impulse and

Velocity gauge.) The MIV gauge package is an $\approx 60\ \mu m$ thick membrane, which is suspended in the liquid so that a particle velocity and an impulse measurement are made at each Lagrangian position. The use of this gauging system is discussed in some detail in Reference 15. In theory it is possible to measure particle velocity, impulse (pressure), and shock velocity in an unreactive material with the MIV gauge technique. In our experiments, reaction causes changes in the flow field from one gauge to the next, so only average shock velocities are obtained. The impulse data are used primarily as an indicator of the pressure because of the difficulties encountered in numerical differentiation. The particle velocity data are the most reliable measured quantity and are the primary information obtained.

Since the NM materials were liquids, a 68.6-mm outside diameter cell made from Plexiglas was used to clamp the gauge package and contain the sample material. (Details of the gauge and cell construction are shown in Figure 3.) The cell fronts were either Kel-F or Plexiglas (depending on the pressure desired in the sample material) and were approximately 6-mm thick. The MIV gauge membrane was suspended in the cell at a 30 degree angle with the cell front, giving 5 particle velocity and 5 impulse gauge measurements in each experiment (one of each gauge type at a common axial position), with each of the 5 axial positions separated by approximately 1 mm. The inside of the cell, which comes in contact with the NM, was lined with Teflon film to eliminate any NM attack on the PMMA. Before the cell front was attached, the positions of the gauges in the cavity were measured with a depth measuring microscope. After a cell was filled with explosive, care was taken to eliminate all the air bubbles in the explosive before sealing. In all cases, the shock initiation experiment was completed within 3 hours of filling.

Input to the NM was by a well controlled gas-gun-driven Kel-F projectile with a sapphire facing. Inputs ranged from 5.8 to 7 GPa (projectile velocities of 1.26 to 1.42 mm/μs). The target was positioned between the pole pieces of the electromagnet in a region where the magnetic field was uniform to within 1%. A schematic of the projectile and target shortly before impact is shown in Figure 4.

RESULTS AND DISCUSSION

Shot data for each of the experiments are given in Table 1. Because Hugoniots for the two materials have not yet been measured, we used the "universal" liquid Hugoniot[17] for the shock velocity–particle velocity relationship, along with the appropriate densities, to obtain the input conditions by impedance matching. The input

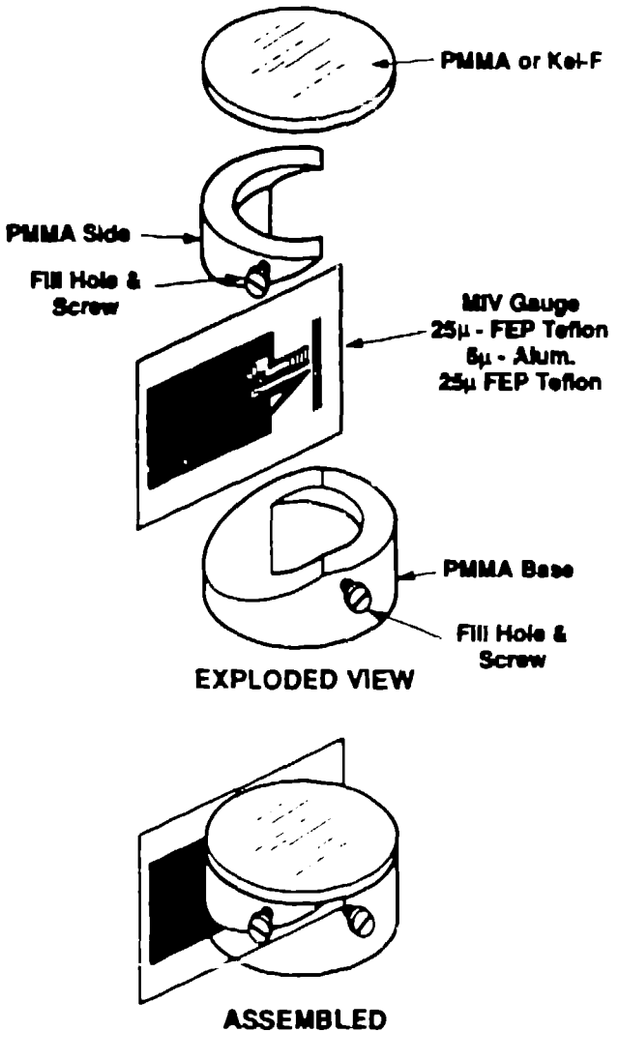

FIGURE 3. CELL AND GAUGE CONSTRUCTION DETAILS.

conditions are estimates with an accuracy of a few percent. These will be corrected later after the Hugoniots are measured.

HOMOGENEOUS NM

Particle velocity profiles and the resulting time-distance (t-x) diagram from Shot 747 (a 95/5 wt % NM/DETA experiment with a NM input pressure of 6.8 GPa) are shown in Figure 5. (The third gauge record was noisy because the recording digitizer differential comparator was set incorrectly.) Notice the constant level initial shock, followed by changes in the waveform due to shock-induced reaction considerably behind the shock front. In this experiment transition to detonation occurred between the third and fourth gauges, as indicated by the changes in particle velocity waveform profiles. Times at both the start and the top of the growing

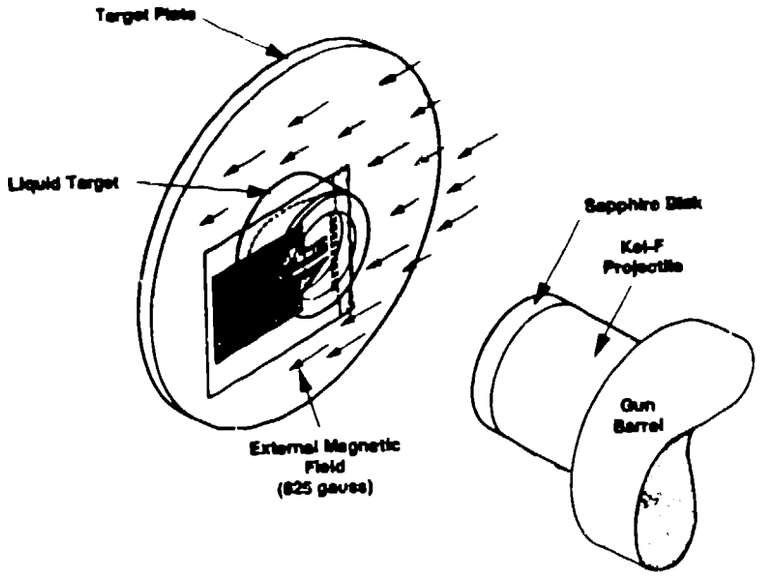

FIGURE 4. PROJECTILE AND TARGET JUST BEFORE IMPACT.

TABLE 1
NITROMETHANE SHOT DATA

Shot No.	Impactor Mat'l	Cell Front Mat'l	Proj. Vel. mm/μs	Initial Density Mg/m^3	Impedance Match NM Input Conditions Part. Vel. mm/μs	Stress GPa	Shock Vel. mm/μs
Homogeneous (95/5 wt% NM/DETA)							
747	Sapphire	Kel-F	1.41	1.11	1.48	6.8	4.12
755	Sapphire	Kel-F	1.26	1.11	1.34	5.8	3.89
Heterogeneous (92.75/6.0/1.25 wt% NM/silica/guar gum)							
748	Sapphire	Kel-F	1.42	1.17	1.47	7.0	4.10
754	Vistal	PMMA	1.42	1.17	1.33	6.0	3.88
756	Sapphire	Kel-F	1.39	1.17	1.44	6.8	4.06

wave (at each gauge position) were determined from the particle velocity waveforms, plotted on each gauge trajectory, and then joined to produce the wave paths shown in the t-x diagram. Notice they coalesce into a single shock.

This diagram indicates the detonation-wave evolution process occurs over a relatively long time and distance, with the wave starting out as a compressive wave and growing to form a shock. As shown, a super-

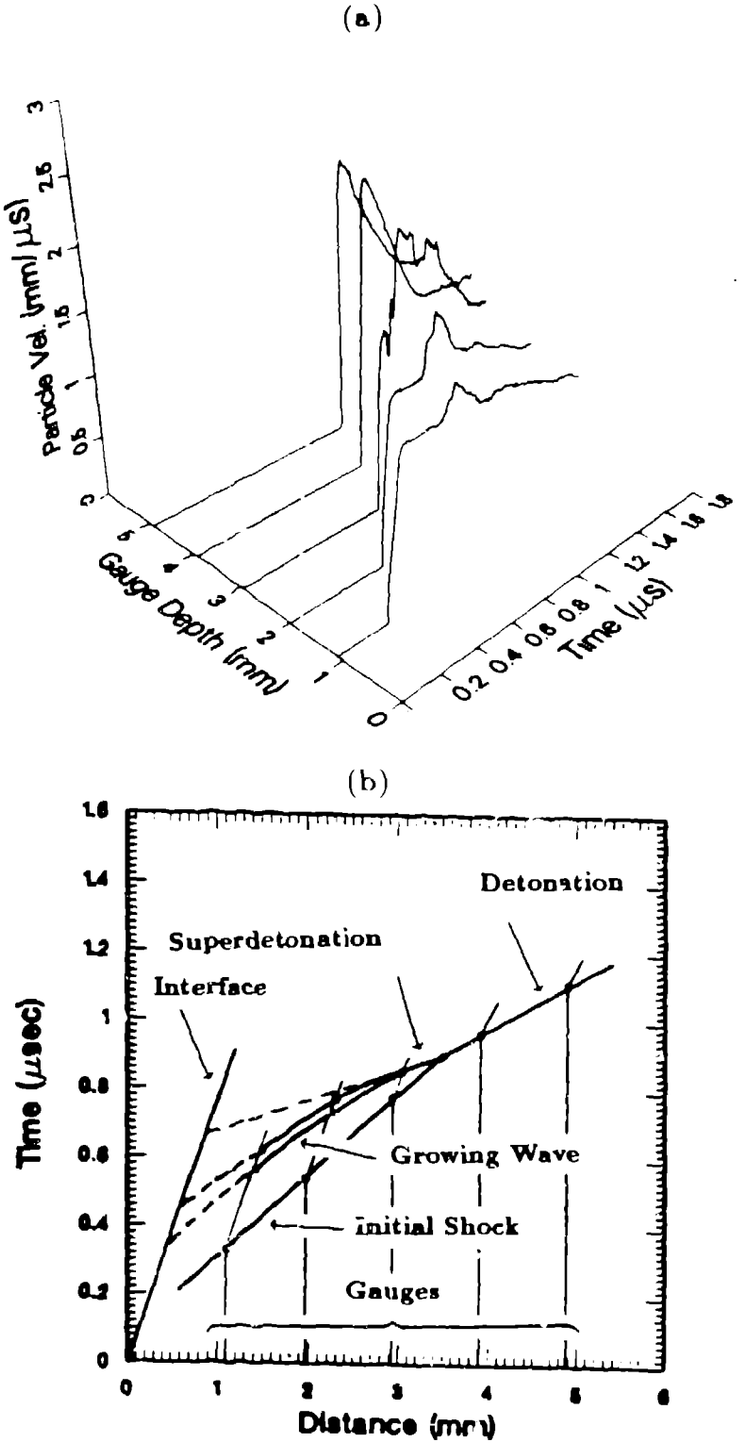

FIGURE 5. PARTICLE VELOCITY WAVEFORMS FROM SHOT 747 ARE SHOWN IN (a) AND THE RESULTING TIME-DISTANCE DIAGRAM IN (b).

detonation does not form immediately at the input boundary but rather after a relatively long buildup process. In fact, we can only infer from our records that a superdetonation occurs just before overtake of the initial wave. This inference is made because a wave of superdetonation velocity is required to link up the information from the particle velocity gauges into the consistent picture shown in Figure 5(b). In this figure the dashed line with the shallow slope represents the locus of a superdetonation that would be required to agree with the initial shock overtake time observed from the waveforms and indicates clearly that a relatively long induction time would be required when compared to our data. In previous studies, streak camera measurements of emitted light as the initiation progressed led to the model of the superdetonation being formed immediately after the thermal explosion at the input NM boundary. In our experiments, we do not yet know at what point light would begin to be emitted with an intensity that a streak camera could record.

A similar analysis was done for Shot 755 but since the wave did not completely grow to a detonation before the end of recording, it was only possible to estimate the condition where detonation would have been attained. The evolving wave in this experiment achieved about the same wave shape at the position of the fifth gauge as was observed in the second gauge of Shot 747. The time-to-overtake data are tabulated in Table 2 for these two experiments.

TABLE 2
HOMOGENEOUS NM RELAXATION & OVERTAKE DATA

	Calculated NM Input Conditions			From Gauge Meas.		Overtake Conditions	
Shot No.	Part. Vel. mm/μs	Pressure GPa	Shock Vel. mm/μs	Part. Vel. mm/μs	Pressure GPa	Distance mm	Time μs
747	1.48	6.8	4.12	1.30	6.2	3.3	0.9
755	1.23	5.8	3.89	1.24→1.18	5.5→5.1	≈7.8	≈2.0

To give some idea of how our work fits with previous studies, we compare it to the work of Hardesty[8]. He reported a series of very nice powder-gun-driven experiments in which the NM input boundary particle velocity was measured using a VISAR velocity interferometer system[18]. In the same experiments, the emitted light was measured with a streak camera in the same way that it had been done in several previous studies. These experiments were done on neat NM with pressures generated in the NM being between 7 and 9 GPa. Because the inputs were the result of projectile impact, a (known) constant state was developed in the NM after the initial shock

Hardesty observed that particle velocity decay at the input interface occurred considerably before any emitted light was visible on the streak camera record; in fact the decay occurred in about 50 to 60% of the time to first light. He stated that using the emitted light to indicate the thermal explosion may lead to overestimates, by nearly a factor of two, in the induction time. He did not, however, indicate what effect this observation would have on the accepted homogeneous initiation model. It is obvious that decreasing the thermal explosion time by a factor of two, while leaving the time at which the superdetonation overtakes the initial wave the same, would lead to a low velocity superdetonation, too low to be physical.

If we fold our data into this picture, interpreting the information so that the reaction (i.e, the particle velocity relaxation) occurs earlier and the reactive wave does not give off sufficient light to record with the streak camera until it has built up to a compression wave, then our information agrees rather well with the observations of Hardesty. In this case it becomes necessary to modify the homogeneous initiation model to include these features. This new model is shown by the t-x diagram of Figure 5(b). If the two models shown in Figures 1 and 2 are contrasted to our diagram, it is easy to identify considerable differences. In our model the superdetonation develops from a growing compression wave which starts from the runaway chemical reaction at the input interface, with the buildup occurring over a considerable time and distance. This sharply disagrees with the idea that the superdetonation forms immediately, giving off sufficient light to record, and indicates that emitted light is not a good measure of the induction time (time to run away chemical reaction or a thermal explosion).

We are not yet prepared to guarantee that Figure 5(b) is the correct model for homogeneous NM initiation. Additional experiments will be required to eliminate the possibility of gauge perturbations and demonstrate that this model is correct. It should also be remembered that we are working with NM/DETA homogeneous explosive rather than neat NM, although the agreement between our ideas and Hardesty's neat NM data seems to indicate they have similar behavior. We also note that addition of 5 wt% of DETA to NM produces a small amount of the DETA/NM salt in suspension in the NM. We plan to eliminate this complication, in the future, by working with lower DETA concentrations.

Another interesting aspect of our measurements is that in each of the homogeneous NM experiments, the condition in the initial shock was somewhat different from that expected for the particular impact conditions and the estimated Hugoniots of the materials involved. The gauge measurements of the initial wave indicate a lower pressure (and also particle velocity) state than would be expected based on estimating the input NM

conditions by impedance matching (by about 10% in both pressure and particle velocity). We feel this difference is more than the uncertainty in the Hugoniot for the NM, indicating that something (perhaps endothermic chemistry) may be happening in the initial wave. In fact the particle velocity and impulse waveforms recorded on Shot 755 show a generally decreasing state from the first to the fifth gauge. To illustrate this difference, the expected input state and the measured state in the initial wave are compared in Table 2

Time-to-overtake values obtained from the t-x diagrams are also tabulated in in Table 2. In Shot 747, overtake occurred between the third and fourth gauges and it appears that, to make all the times come out correctly the reactive wave grew to a superdetonation prior to overtaking the initial wave. In the case of Shot 755, we extrapolated the measured wave behavior to where we thought the overtake would have occurred if a longer measurement were possible. These data have been plotted on a Pop-plot, along with some of the available neat NM data (taking some liberty in interpreting the data, because all the previous experimenters did not report the time-to-overtake). These are shown in Figure 6. Data given in the figure are these: C,D,T are from Campbell, Davis, and Travis[1], Hard. are from Hardesty[8], Vosk. et al. are from the Soviet Union[9], Walk. are from Walker and Wasley[9], SRI are from Berke, et al.[7], and New Homo are from this study. A line has been drawn through the neat NM data (without the benefit of a fitting routine) to give some idea of the slope of the data. A line has also been drawn through the two points of this study for the chemically sensitized NM (remembering that the lower pressure point is an estimate) and, while it is not the same slope as that of the neat NM, it is similar and much different than for the heterogeneous data, which will be shown later. It is interesting to note that adding the 5% DETA to the NM increases the sensitivity greatly, the line moves down in pressure by $\approx$ 3 GPa at an overtake time of 1 μs.

HETEROGENEOUS NM

The buildup in the heterogeneous NM experiments was totally different from that observed in the homogeneous experiments. Figure 7 is a plot of the particle velocity waveforms measured in Shot 748 where the explosive was heterogeneous NM (92.75/6.0/1.25 wt% NM/silica/guar gum with an input pressure of 7.0 GPa) The gauge records indicate a wave that is growing both at the front and behind the front. Because of this growth in the front, it is impossible to estimate from the records what the input condition was. We have calculated this condition using impedance matching techniques and the estimated input conditions are given in Table 1. Although the input conditions varied from 6 to 7 GPa, the waveforms from each experiment were similar. The t-x

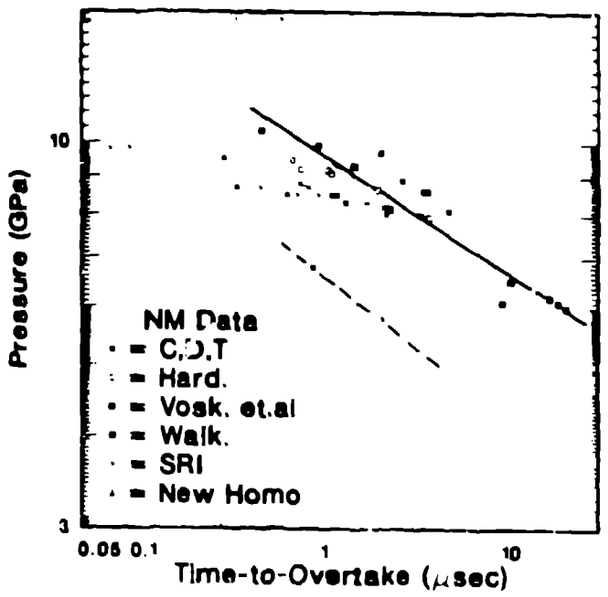

FIGURE 6. POP-PLOT OF OUR NEW DATA FOR SENSITIZED NM AND SOME OF THE AVAILABLE DATA ON NEAT NM.

diagrams were plotted from the gauge data and the time and distance-to-detonation conditions were determined. These are tabulated, along with the estimated input conditions, in Table 3.

TABLE 3
HETEROGENEOUS NM DETONATION TRANSITION CONDITIONS

	Calculated NM Input Conditions			Transition Conditions	
Shot No.	Part. Vel. mm/μs	Pressure GPa	Shock Vel. mm/μs	Distance mm	Time μs
748	1.47	7.0	4.10	3.2	0.72
754	1.33	6.0	3.88	4.57	0.93
756	1.44	6.8	4.06	3.27	0.74

In all three heterogeneous experiments, an electrical burst occurred at about the same time that the apparent transition to detonation occurred. Evidence of this phenomenon can be seen in Figure 7 (in the form of noise on the particle velocity waveforms) indicating that the transition occurred between the third and fourth gauges. The source of this signal is unknown (it was not observed in

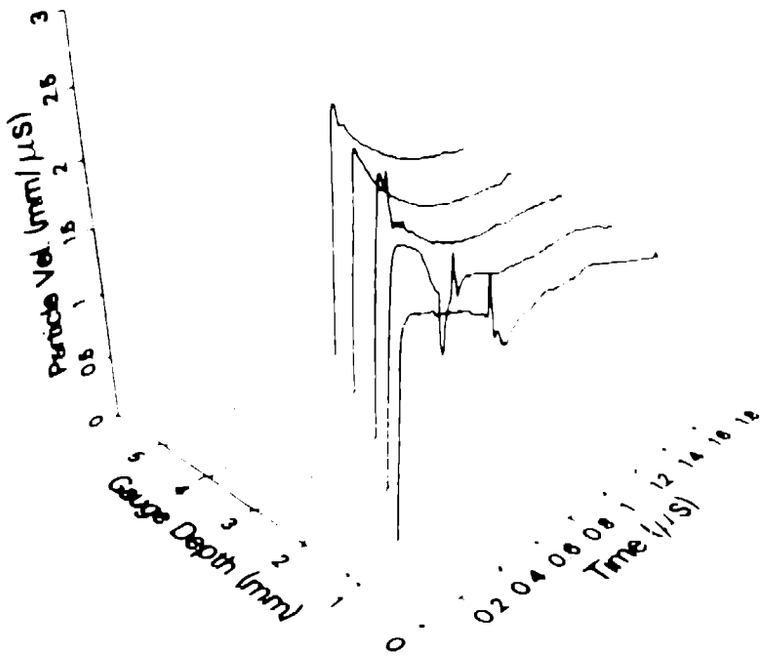

FIGURE 7. PARTICLE VELOCITY WAVEFORMS FROM SHOT 748; INPUT 7.0 GPA.

the homogeneous experiments) although a similar phenomena was observed by Sheffield in shock-induced reacting carbon disulfide experiments done at Washington State University several years ago[20]. We used this electrical burst to pinpoint the transition-to-detonation when drawing the t-x diagrams.

The three time-to-detonation points for heterogeneous NM are plotted, along with the homogeneous data of this study and several data for PBX 9404[21], in Figure 8. Lines have been drawn through all three sets of data to give some idea of the relationship between them. The heterogeneous NM is very close to the PBX 9404 data in both position and slope, showing that it is a relatively sensitive material. The slope of the chemically sensitized homogeneous NM is much different from the heterogeneous NM slope. It is interesting to note that one homogeneous datum and two heterogeneous data are nearly in the same position on the Pop plot so that they achieved detonation in similar times and distances but the wave forms (and the initiation process) of Figures 3 and 7 are completely different.

Waveforms obtained in the heterogeneous experiments are consistent with earlier studies on solid, heterogeneous explosives, i.e., the wave grows at the front (as well as behind the front) into a detonation. However, we believe that a great deal can be learned about heterogeneous initiation by varying the size, the number density,

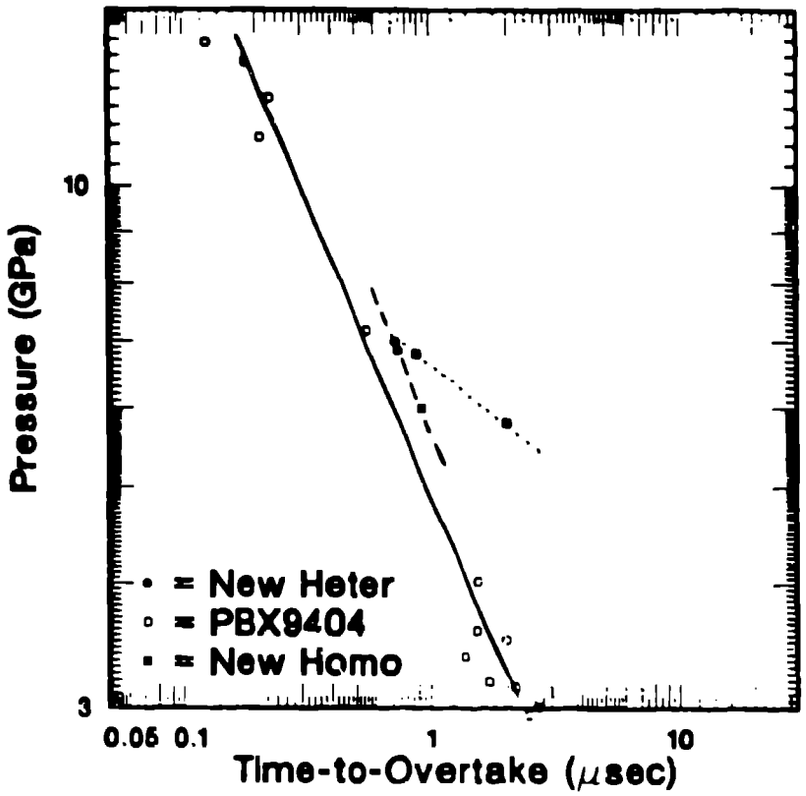

FIGURE 8. POP-PLOT FOR HETEROGENEOUS NM, ALONG WITH OTHER MATERIALS FOR COMPARISON.

the shape, and the nature of the heterogeneities in the gelled NM. We conjecture that it will be possible to tailor initiation waveform profiles by changing the variables associated with the heterogeneities.

SUMMARY AND CONCLUSIONS

In-situ multiple magnetic gauging experiments have been completed on homogeneous and heterogeneous NM Homogeneous explosive was made by chemically sensitizing neat NM. This made it sensitive enough that it could be initiated with our gas gun, allowing a well controlled input to the liquid explosive. We measured particle velocity profiles at discrete Lagrangian positions from initiation to detonation in a single experiment with multiple gauges. Based on our measurements, we have proposed important refinements to the classical homogeneous shock-to-detonation initiation model, including a relatively long reactive wave buildup which evolves into a superdetonation that overtakes the initial shock. This differs considerably from the classical model in which a thermal explosion occurred, immediately producing a superdetonation which traveled a considerable distance in the precompressed NM before overtaking the initial shock. The chemically sensitized NM has a similar slope in the Pop-plot plane to neat NM but offset to lower pressures by $\approx$ 3 GPa.

Heterogeneous NM was produced by gelling the NM and suspending silica particles in it. Again we measured from initiation to detonation in a single experiment. Growth occurred both at and behind the front in a manner similar to that which has been measured in other heterogenous materials. The sensitivity of this physically sensitized heterogeneos NM was similar to that of PBX 9404 (ρ_0 = 1.84 Mg/m^3) both in slope and position on a Pop-plot. We hope this is the first step in a program in which considerable understanding can be developed concerning the role of hot spots in heterogeneous initiation, as we can control the size, shape, number density, and impedance of the heterogeneities.

That these experiments could be done over the full detonation regime without gauge failure has been gratifying. This technique will almost certainly lead to a more detailed understanding of the buildup process in both homogeneous and heterogeneous systems. We are, however, concerned about gauge perturbations and must check this possibility out carefully before making categorical statements. Much remains to be done to completely understand the processes observed in the waveforms we have already obtained. It will take a number of gun experiments on both the homogeneous and heterogeneous materials to rule out gauge perturbation effects and determine how the process varies with input conditions. At the present time, the consistency of our data, both between the experiments we have done and also with the earlier studies, suggests the perturbations are small.

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