INITIATION OF DETONATION IN LIQUID EXPLOSIVES BY A REFLECTED SHOCK WAVE

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Initiation of detonation by shock wave reflection from a high impedance anvil was investigated in amine-sensitized nitromethane. Initiation was observed by luminosity and a distinct pressure "hump" appearing approximately 1 μsec after shock reflection. The critical shock pressures required for incident and reflected initiation were determined by varying the strength of the shock wave transmitted into the explosive. The incident and reflected shock temperatures are estimated using different equations of state for nitromethane, and the temperatures required for incident and reflected initiation are found to be approximately equal. This result suggests that the initiation of detonation is governed by temperature, independent of the particular shock-loading path used.

INTRODUCTION

While the initiation of detonation in homogenous liquid explosives by planar shock waves has been studied extensively over the past four decades, there remains considerable disagreement over the mechanism and details of the initiation process. The classic picture of initiation by Chaiken[1] and Campbell, Davis, and Travis[2] suggests that initiation of detonation is governed by the thermal decomposition of the first layer of the shock explosive which, after an ignition delay time, begins to release energy. The volumetric dilation of the reacting explosive generates a secondary shock wave, giving rise to a "superdetonation" which propagates into the explosive already compressed by the incident shock, eventually overtaking the incident shock. Sheffield et al.[3] proposed a modification of this picture in which the superdetonation develops gradually due to continuous compressions generated by the reacting explosive. Walker and Wasley[4] and more recently Pujols and Pouligny[5] have questioned the existence of superdetonations and suggested that detonation initiation occurs at the incident shock front as it propagates into the explosive. Thus, there is at present no consensus as to whether bulk temperature or localized reactions (e.g., hot spots) govern initiation.

In order to further investigate the mechanism of shock initiation of homogeneous explosives, it is of interest to

explore other methods of shock loading (e.g., ramp compression via train of weak shock waves, reflected shock wave, etc.). The use of multiple or reflected shock waves allows "off-Hugoniot" states to be realized, permitting the pressure and temperature of the explosive to be varied along thermodynamic paths other than the single shock Hugoniot. Reflected initiation of detonation in heterogeneous explosives has been obtained by Marlow[6], and more recently by Tarver et al.[7], and significant energy release without detonation was reported by Winter et al.[8] Although attempts at initiation of detonation in nitromethane by reflected shock were made by Presles et al.[9], no successful initiation was observed. Presles et al. concluded that since the reflected shock does not result in significant temperature increase, the shock reflected from aluminium in their experiment was not sufficiently strong to initiate detonation.

Work by Gruzdkov, Winey, and Gupta[10] involved multiple shock reverberations between two stiff anvils to compress nitromethane to pressures of up to 19 GPa. The onset of reaction in pure nitromethane is observed at temperatures above 940 K, apparently independently of the pressure. While the reverberating shock wave technique provides a means to study the onset of chemical reaction in off-Hugoniot (quasi-isentropically compressed) states, the samples used were too small (typically 100-300 μm) to permit the initiation and propagation of detonation to be observed.

The present study examines the initiation of detonation in a homogeneous liquid explosive by reflecting a subcritical incident shock off a high impedance anvil. The charge is significantly larger than the critical diameter of the test explosive, permitting detonation initiation and propagation in a shock-compressed explosive to be observed. These experiments seek to identify and compare the critical pressure and temperature to initiate detonation in the incident versus reflected mode of initiation.

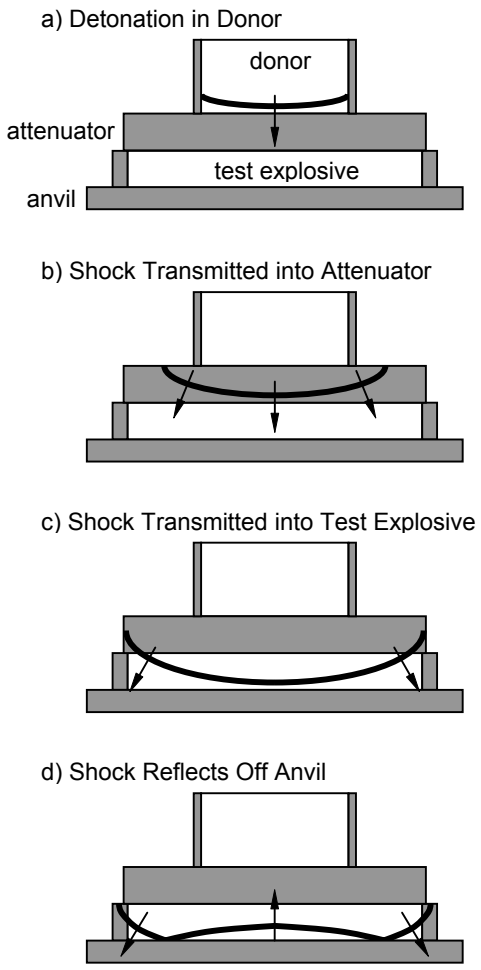

FIGURE 1. SCHEMATIC OF EXPERIMENTAL SEQUENCE.

EXPERIMENT

The experimental arrangement used in this investigation is shown schematically in Figure 1. An explosive donor is used to transmit a shock wave into a capsule containing the text explosive via an attenuator of inert material. The attenuator thickness can be varied to control the strength of the transmitted wave. The capsule is larger in diameter than the donor charge to prevent initiation from occurring via interaction of the transmitted shock with the capsule walls.[11,12] For experiments in which the incident shock

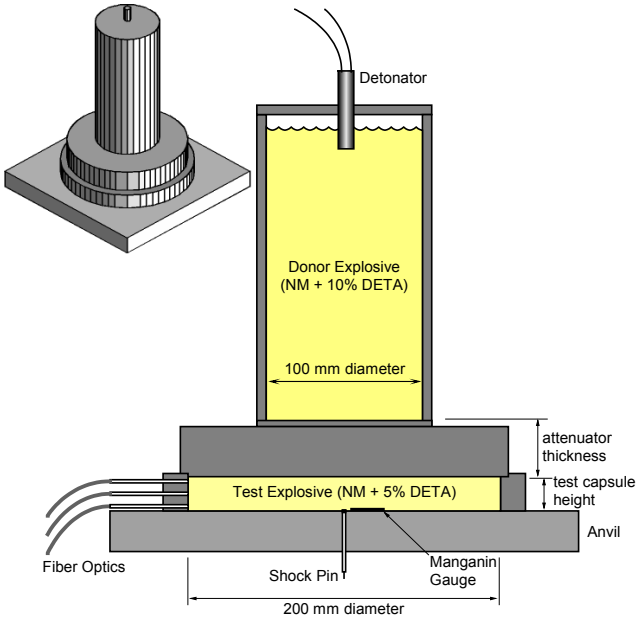

FIGURE 2. CHARGE DESIGN.

wave is not sufficiently strong to initiate detonation, the shock propagates through the test explosive and reflects off an anvil. The strength of the reflected shock is governed by the impedance of the anvil material. In this study, mild steel was predominately used as the anvil, with additional experiments being performed using PVC, aluminum, and titanium anvils.

A detailed schematic of the charge is shown in Figure 2. Gray polyvinyl chloride (PVC) was used as the material for the donor charge and test explosive containers, as well as the attenuator, because of its compatibility with nitromethane. The donor consisted of a 100 mm by 200 mm charge of nitromethane (commercial grade) sensitized with 10wt% DETA (Aldrich). The test explosive was nitromethane sensitized with 5wt% DETA. This explosive has been used in prior shock initiation studies.[3,13] Since the critical diameter of NM + 5wt% DETA (2-3 mm for light confinement[14]) is much smaller than the scale of the charge used, we can expect that a detonation will propagate if initiated.

The mixture of nitromethane and DETA was prepared within 1-2 hours of the experiment. The test capsule was carefully filled and any visible bubbles were removed prior to the experiment. Most tests were performed with the text explosive temperature controlled to between 23 and 27 °C, independent of the ambient temperature. This is important, as the shock sensitivity of liquid explosives is known to be extremely sensitive to initial temperature.

Luminosity from an experiment was monitored by fast-response photodiodes that received light from fiber optics mounted into light pipes. The fiber optic/light pipe was separated from the liquid test explosive by a Mylar window. High impedance (50 Ω) Manganin gauges (Dynasen MN4-50-EK) were used to record the shock pressure history for incident and reflected events.

One concern in experiments of this type is the occurrence of Mach reflection as the curved shock wave interacts with the anvil (Figure 1). Mach reflection of an oblique shock can result in much higher shock temperatures and pressures than would result from normal reflection.[15] Assuming a spherically expanding shock front is transmitted into the test explosive, simple geometric considerations show that the angle of the interaction between the shock and anvil does not reach values necessary for the formation of a Mach reflection until more than 10 μsecs after the central portion of the shock has reflected off the anvil. Since the initiation events reported below typically occurred within 2 μsecs of shock reflection off the anvil, Mach reflection is not believed to have played a role in the initiation event.

RESULTS

Incident Initiation

Upon shock entry into the test explosive, the photodiodes detected luminosity within 1 μsec of shock entry if initiation occurred, or no

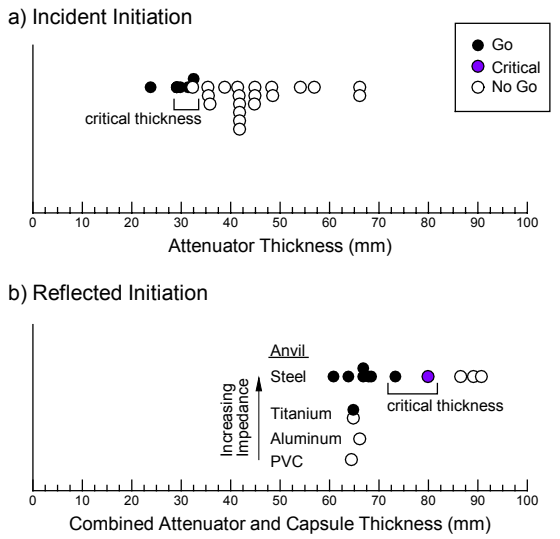

FIGURE 3. SUMMARY OF RESULTS.

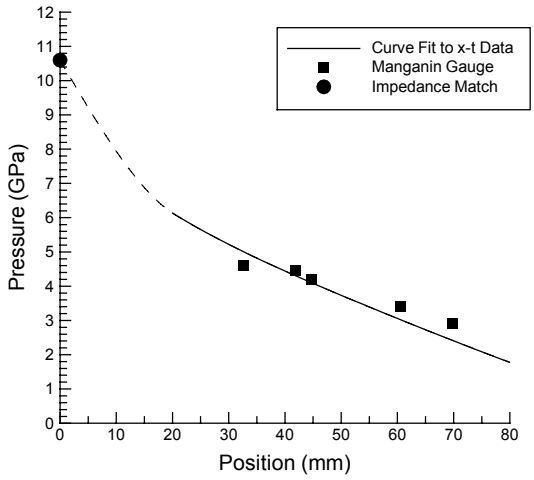

FIGURE 4. SHOCK PRESSURE IN PVC ATTENUATOR.

luminosity at all was detected if initiation did not occur.[16] The photodiodes were found to be not sensitive enough to observe shock-initiated reaction without detonation. After repeating this experiment to determine the critical thickness of attenuator necessary for initiation, it was determined that the critical thickness was in the range of 31-33 mm. A summary of these experiments is shown in Figure 3.

In order to relate the critical thickness of attenuator to the critical shock pressure necessary for initiation, the shock pressure in the PVC attenuator was determined by measuring the trajectory of the shock transmitted into the attenuator using shock pins (Dyansen CA-1135) and self-shorting contact gauges embedded in the attenuator. Approximately 30 arrival time measurements were obtained at locations between 17 mm and 80 mm inside the PVC; these arrival times were fit with a 4th order polynomial. The polynomial was differentiated to obtain shock velocity and, with the known U_s-u_p Hugoniot for PVC[17], the shock pressure in the PVC was computed. The shock pressure as a function of attenuator thickness for this curve fitting technique is shown as a curve in Figure 4, along with the symbols representing direct manganin gauge measurements of the shock pressure in PVC made at select locations in the attenuator. Finally, the pressure of the shock transmitted into the PVC by detonation of the NM + 10%wt donor charge (as calculated by impedance matching calculations with the expansion adiabat of the donor explosive) is shown as solid circle on the *y*-axis of the figure. The good agreement obtained between these various methods lends confidence to the values of shock pressure in PVC. With the shock pressure known for a given thickness of attenuator, the shock pressure transmitted into the nitromethane test explosive can be calculated by impedance matching techniques. The equation of state for pure nitromethane[17] is used for the test mixture of NM + 5%wt DETA under the assumption that the DETA content does not affect the shock impedance of the mixture until the onset of chemical reaction. For the critical PVC attenuator thickness of 31-33 mm, the shock pressure in PVC is 5.0 GPa, which transmits a 4.3 GPa shock into the test explosive.

Reflected Initiation

If the attenuator is thicker than 33 mm, the transmitted shock will propagate through the

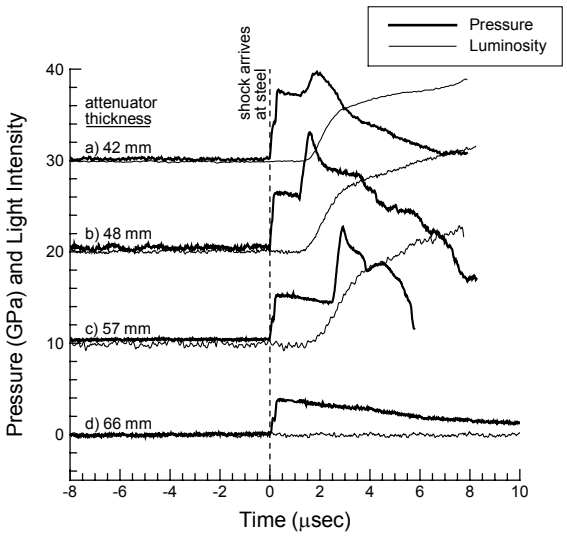

FIGURE 5. REFLECTED INITIATION FOR DIFFERENT INCIDENT SHOCK STRENGTH.

test explosive without initiating detonation. Upon reaching the bottom of the capsule, the shock reflects off of the anvil. Results of experiments with a 23 mm thick capsule and different thickness of attenuator are shown in Figure 5. The manganin gage was located at the center of the anvil at the interface between the test explosive and the anvil. The signals in Figure 5 are synchronized such that time zero corresponds to the instant of shock reflection. The first trace (Figure 5a) corresponds to an experiment with a 42 mm thick attenuator. Prior to the arrival of the shock at the anvil, no luminosity was observed. Upon shock reflection, the manganin gauge recorded a reflected shock pressure of 7.5 GPa. At 1.2 μsec after shock reflection, the photodiode began to detect light. Simultaneous with the appearance of luminosity, the manganin signal shows a distinct "hump" which reached a pressure of 9 GPa.

If the attenuator thickness was increased to 48 mm (Figure 5b), a weaker shock was transmitted into the test explosive, resulting in a weaker reflected shock (6.5 GPa), but a reflected initiation event was still observed. Again, after a delay of 1.2 μsec following the shock reflection, a pressure "hump" appeared in the manganin signal coincident with the appearance of luminosity. In this experiment, the amplitude of the reflected event reached approximately 13 GPa. The high-impedance magnanin gauges used here are only intended for use up to shock pressures of approximately 10 GPa. Thus, they cannot be expected to accurately record the propagation of detonation in shock precompressed nitromethane. As the attenuator thickness was increased further to 57 mm, the reflected pressure was lowered to 5.2 GPa and the delay before initiation increased to 2.5 μsec. Finally, for an attenuator of 66 mm, no luminosity was observed and the pressure signal showed only the reflected shock followed by gradual decay. This result appears similar to results obtained in pure, non-reacting nitromethane published previously.[16]

The results of additional experiments similar to these with attenuators ranging in thickness from 37 to 66 mm are summarized in Figure 3, where the total distance of shock travel (attenuator and capsule thickness) prior to shock reflection is plotted. The occurrence of a reflected initiation event which occurred within 1.5 μsec of shock reflection is reported as a solid symbol. Experiments in which no luminosity or pressure activity was observed (similar to the results of Figure 5d) are represented by an open symbol. The shaded symbol corresponds to the experiment at critical conditions (Figure 5c).

In order to further investigate the details of the reflected initiation event, additional experiments were performed with manganin gauges suspended in the liquid test explosive. The gauges were located at approximately 3.5 mm and 8 mm above the anvil surface as well as on the anvil surface. The results of two experiments with the same attenuator (44 mm) and test capsule thickness (23 mm) are superimposed in Figure 6. The 3 GPa incident shock can clearly be seen propagating toward the anvil. Similar to Figure 5a,

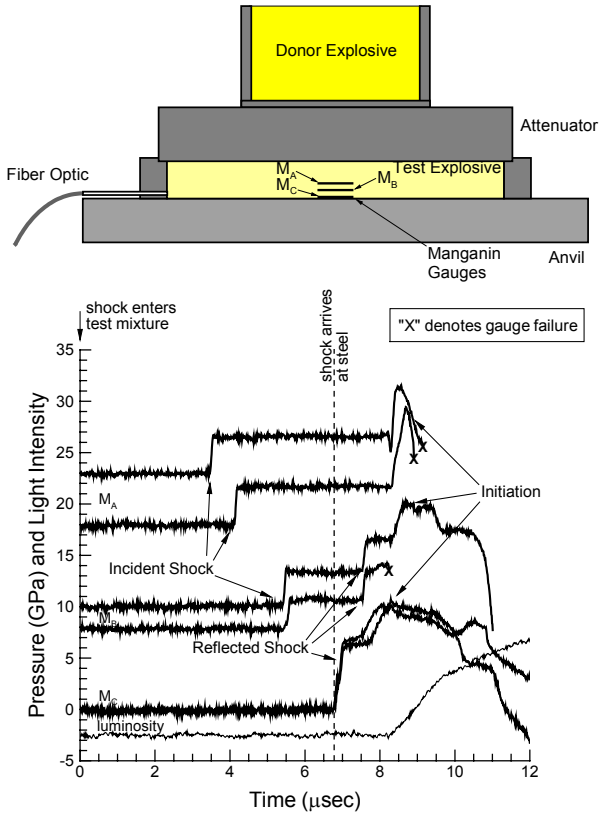

FIGURE 6. DETAILS OF REFLECTED INITIATION.

luminosity and pressure activity begin 1.2 μsec after shock reflection. The reflected shock is visible propagating back up, with the characteristic pressure hump steepening and catching up to the reflected shock at the top manganin gauge (M_A). Again, these gauges are not intended for use at the high pressures that occur in detonation, and several gauges are observed to fail shortly after the reflected initiation.

In addition to varying the strength of the transmitted shock by changing the attenuator thickness, the strength of the reflected shock can also be varied by modifying the impedance of the anvil for a fixed incident shock strength. For these experiments, the attenuator thickness was fixed at 42 mm and the capsule height was 23 mm, giving a total shock travel distance before reflection of 65 mm. As can be seen in Figure 3, these experimental parameters consistently result in

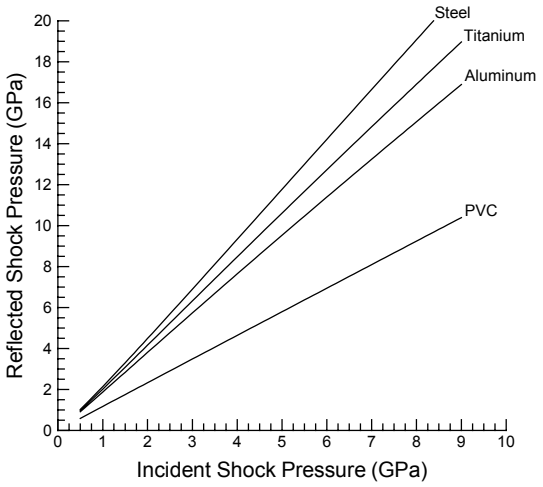

FIGURE 7. SHOCK REFLECTION IN NITROMETHANE.

initiation if a steel anvil is used. Experiments with PVC and aluminum anvils did not result in initiation upon reflection; no luminosity was observed. Two experiments were performed with titanium anvils: one test did not result in initiation, the other test resulted in initiation, with luminosity and manganin pressure signals similar to the result in Figure 5b. This implies that the shock reflected from titanium is near the critical shock pressure. As can be seen from impedance matching calculations for shocks in nitromethane reflected from various materials (Figure 7), titanium results in a slightly lower reflected shock pressure than steel, which may explain why a titanium anvil yielded critical results. Shock reflection from aluminum results in only 80% of the pressure obtained from steel, and PVC results in less than 50% of the pressure from steel. Thus, the fact that no initiation was observed with these materials confirms that the strength of the reflected shock determines if initiation occurs.

ANALYSIS

The results of the incident and reflected shock initiation experiments are summarized in Table 1, which reports the critical shock

TABLE 1. SHOCK PRESSURES AND TEMPERATURES

	Shock Pressure	Estimated Temperature	
		Walsh & Christian	Winey et al.
Incident Initiation	4.3 GPa	662 K	720 K
Reflected Initiation*:			
Critical Incident Shock	2.5-2.8 GPa	492-577 K	576-602 K
Critical Reflected Shock	6.0-6.5 GPa	565-590 K	701-724 K

*For steel anvil.

strengths as measured via manganin gauges and estimated by the observed shock speed and the known Hugoniots of the materials involved. Of course, the critical shock strength required for initiation of detonation is not a unique property of an explosive. Successful initiation of detonation requires that a shock of sufficient strength be applied for a given duration, and the duration of shock can be limited by rarefactions generated from the boundary conditions. For a given scale and geometry of experiment, however, whether detonation is observed or not should be a function of the shock strength. As can be seen from Table 1, the critical pressure for initiation is not the same for incident and reflected shock initiation, showing that pressure is not the determining factor. Since Gruzdkov et al.[10] found that the onset of reaction is a function of temperature, it is of interest to examine the temperatures achieved via incident and reflected shock compression of nitromethane.

The classic Walsh and Christian technique for finding temperature[18] starts with the assumption that c_v and $(\partial P/\partial T)_v$ are approximately constant and their values correspond to those at the initial (unshocked) state. This assumption permits integrating the First Law along the Hugoniot to find temperature:

$$T_1 = T_o e^{\beta(v_o - v_1)} + \frac{e^{-\beta v_1}}{2c_v}\int_{v_o}^{v_1}\left[P - P_o + (v_o - v)\frac{dP}{dv}\right]e^{\beta v}\,dv$$

$$\beta = (\partial P/\partial T)_v / c_v \qquad (1)$$

For nitromethane, the values of c_v and $(\partial P/\partial T)_v$ were taken from Presles et al.[9] The computed temperature along the Hugoniot for the incident shock as a function of pressure is shown in Figure 8. In order to compute the temperature for reflected shocks, it is assumed that the principal Hugoniot in the (*P, v*) plane is still valid, and the integration of Equation (1) is initialized with the new values of (*T, v*). Calculations of the reflected shock temperature as a function of the final pressure are also shown Figure 8 as curves that depart from the principal Hugoniot at the condition prior to shock reflection ($P_{initial}$ = 1, 3, 5, and 7 GPa). As noted by Presles et al.[9], most of the temperature rise occurs with the incident shock. For example, for a 5 GPa incident shock, the shock temperature is 725 K, and a second shock that doubles the pressure to 10 GPa only results in an increase to 800 K.

A more sophisticated equation of state for nitromethane has recently been published by Winey et al.[19] This equation of state makes no assumptions regarding the variation of c_v or $(\partial P/\partial T)_v$ and instead makes maximum use of experimental measurements to model these parameters as functions of temperature and specific volume. This equation of state has been verified by direct temperature measurements of nitromethane compressed by reverberating shock waves.[19] Using the analytic expressions for the specific heat at constant volume c_v, the coefficient of thermal pressure $b = (\partial P/\partial T)_v$, and the isothermal bulk modulus $B_T = -v(\partial P/\partial v)_T$ reported in Ref. 19, the pressure and internal energy can be integrated along the Hugoniot as follows.

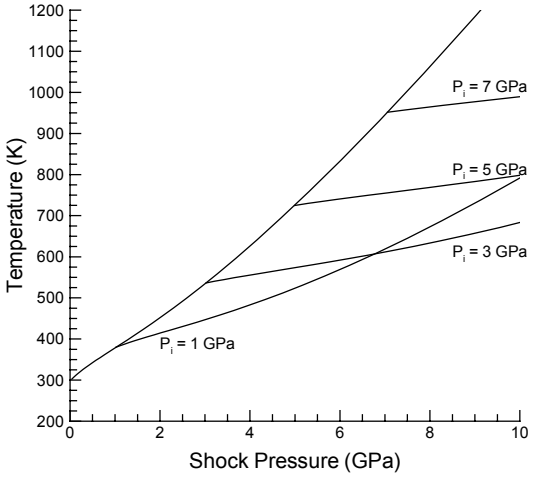

FIGURE 8. TEMPERATURE COMPUTED BY WALSH AND CHRISTIAN.

Starting with the Rankine Hugoniot relation in differential form:

$$de = \frac{1}{2}\left[(v_o - v)dP - (P + P_o)dv\right] \qquad (2)$$

and expressing the differentials dP and de in terms of T and v:

$$dP = \left(\frac{\partial P}{\partial T}\right)_v dT + \left(\frac{\partial P}{\partial v}\right)_T dv = b\,dT - \frac{B_T}{v}dv \qquad (3)$$

$$de = \left(\frac{\partial e}{\partial T}\right)_v dT + \left(\frac{\partial e}{\partial v}\right)_T dv = c_v dT + (Tb - P)dv \qquad (4)$$

the expressions for dP and de from Equations (3) and (4) can be substituted into (2), giving:

$$\left(\frac{dT}{dv}\right) = \frac{(Tb - P) + \frac{1}{2}B_T\left(\frac{v_o - v}{v}\right) + \frac{1}{2}(P + P_o)}{\frac{1}{2}(v_o - v)b - c_v} \qquad (5)$$

which is an ODE with v as the independent variable. This expression is coupled with Equation (3), and together they can be integrated to generate P and T as functions of v along the Hugoniot. The computed values of shock temperature for the Winey et al. equation of state are shown in Figure 9 as a family of curves. Again, the main curve is for the principal shock, and the curves which depart from the main curve are for reflected shocks.

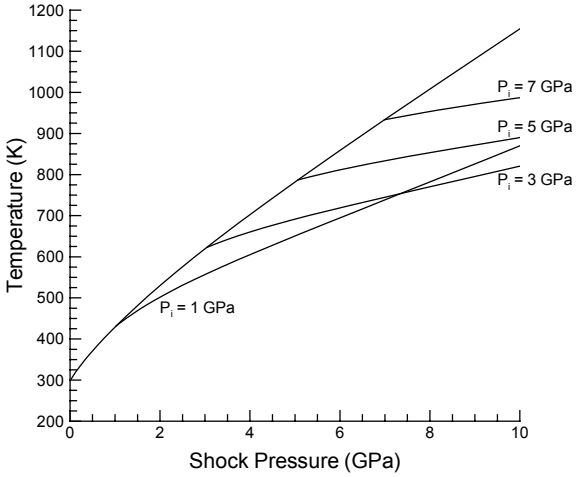

FIGURE 9. TEMPERATURE COMPUTED BY WINEY ET AL.

While the Winey et al.[19] equation of state is qualitatively similar to the Walsh and Christian method, as seen in Figures 8 and 9, the quantitative values of temperature predicted by the two methods disagree significantly for the range of shock pressures encountered in the current experiments (2.5-6 GPa), particularly in computing reflected shock states. The values of temperature have been computed for the critical shock strengths for incident and reflected initiation and are listed in Table 1. Note that the temperatures for incident and reflected initiation as predicted by the Winey et al. equation of state (≈ 700 K) are equal within the error of these experiments. This result is consistent with the hypothesis that temperature is the controlling factor in obtaining initiation. Had only the Walsh and Christian method been used, this conclusion might have been different, since the computed temperatures for incident and reflected initiation differ by as much as 130 ºC.

CONCLUSIONS

The initiation of detonation by reflection of a shock wave from a high impedance anvil has been demonstrated. The initiation event was

characterized by the appearance of luminosity and a distinct pressure "hump" as recorded by manganin gauges. This pressure increase due to reaction becomes steeper as the initiation shock decreases in strength, until a critical shock strength is reached and no initiation occurs. The reactive wave initiated by the reflected shock has been observed to catch up to the reflected shock and overtake it.

The use of a steel anvil resulted in reflected initiation, but the use of anvils of lower impedance materials (titanium, aluminum, PVC) yielded either critical results or no initiation at all. Similar results were noted by Presles et al.[9], who were unable to obtain reflected initiation in pure nitromethane using an aluminum anvil.

The use of a reflected shock wave appears a promising means to study shock initiation by means other than incident shock. The ability to vary pressure and temperature along a different thermodynamic path has the potential to isolate the dominant factor governing initiation. The present results appear to reinforce the conclusion that initiation is a function of the temperatures reached upon shock compression rather than the final pressure.

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