SHOCK INITIATION AND DETONABILITY
OF ISOPROPYL NITRATE

Fan Zhang and Stephen B. Murray
Defence R&D Canada – Suffield, Medicine Hat, Alberta, T1A 8K6, Canada

Akio Yoshinaka
G-Zero, Medicine Hat, Alberta, T1A 1M1 Canada

Andrew Higgins
McGill University, Montreal, Quebec, H3A 2K6, Canada

Shock initiation of detonation and critical diameters were investigated for isopropyl nitrate (IPN). IPN appears to be ideal for fundamental studies of homogeneous liquid explosives because its long detonation length scale facilitates diagnostics. Both direct initiation of detonation and SDT via the onset of detonation were observed. The initial shock pressure versus the time of onset of detonation and the catch-up time were obtained and the critical shock initiation pressure was determined to be in the range of 7-8.5 GPa at 0±5 °C. The critical diameter for neat IPN was found to exceed 310 mm at 16 °C in low impedance PVC tubes. Aluminium particles of 100 nm average size were added to IPN in the belief that the fine aluminium would react significantly within the reaction zone of the IPN detonation. The critical diameter of IPN containing these fine particle additives was found to lie between 29 mm (no detonation) and 48 mm (detonation) in PVC tubes, thus being one order of magnitude smaller than for neat IPN.

INTRODUCTION

Liquid isopropyl nitrate $[(CH_3)_2CHONO_2]$ (IPN) is known to be a monopropellant of low explosive sensitivity. The early work of Brochet[1] indicated that the critical diameter for IPN detonation in a high impedance steel tube was about 20 mm at 20 °C and increased to more than 40 mm at 3 °C. At 20 °C, the measured detonation velocity was about 5.4 mm/µs in a 40 mm steel tube and reduced to about 5.3 mm/µs near the critical diameter. Thus, under high-impedance confinement, the critical diameter for IPN is about an order of magnitude larger than that for nitromethane (NM), while the velocity deficit appears to be somewhat comparable for both. Recently, Sheffield et al.[2] utilized a two-stage gas gun with a high input shock pressure of 7.5 to 8.9 GPa to produce a shock-to-detonation transition (SDT) in a sample of IPN several millimeters in thickness. The resultant SDT was reported as a "superdetonation," which occurred after an induction delay time behind the initial shock wave and which ultimately overtook the shock wave. The samples used were too small to permit propagation of steady, self-sustained detonation to be observed.

Owing to its large detonation length scale versus the resolution times of available diagnostic systems, IPN appears ideal for fundamental studies of homogenous liquid explosives. Furthermore, the critical diameter of homogeneous explosives can be reduced by solid particle additives. Solid particle additives change the behaviour of the explosive from homogeneous to

heterogeneous in two respects: introduction of hot spots and absorption of momentum and heat from the explosive and combustion products. The former effect increases the explosive sensitivity, while the latter reduces the detonation velocity. Lee et al.[3] elucidated these facts in tests employing a packed bed of inert glass beads saturated with sensitized NM. Apart from inert particles, the recent work of Haskins et al.[4] also employed a packed bed of aluminium particles saturated with pure NM, the latter of which has such a thin reaction zone that it does not permit significant reaction of even 100 nm aluminium particles.[5] Thus, the observed effects on the detonation were similar to those with inert beads, suggesting that the shock interaction and transmission around and through the solid particles play a role in determining the critical diameter and detonation velocity. Even during the brief time required for the shock to cross over a particle, significant compaction and deformation occurs for fine particles at enhanced local pressures, and the velocity of the fine aluminium particles increases to 60-80% of the shocked explosive velocity, indicating a significant momentum transfer.[6]

The present paper investigates the shock initiation and detonability of IPN using a revised gap-test device[7] and critical diameter measurements. The charges used for examining shock initiation are close to the critical diameter of IPN, permitting both the detonation initiation and propagation to be observed. Critical diameter experiments are performed in low impedance PVC tubes for both neat IPN and IPN with 100 nm aluminium particle additives. The aluminium particles chosen are small enough that their significant combustion is assumed to be complete within the large reaction zone of IPN. These experiments seek to identify and compare the detonation properties in homogeneous liquids versus heterogeneous particle-additive liquids, with an ultimate goal of understanding the effects of the fine reactive particle additives that equilibrate in the reaction zone of homogeneous energetic materials.

EXPERIMENTAL

Shock initiation set-up

At room conditions, liquid IPN has a density of 1.036 g/m^3, a boiling point of 102 °C, a sound speed of 1.10 mm/μs, and a heat of formation of -229.7 kJ/mole. The shock initiation of detonation in liquid IPN was investigated using a revised donor/gap/receptor type arrangement as shown in Fig. 1. A shock, generated by the detonation of a 100 mm diameter C4 donor charge having a length of 1.5 diameters, was transmitted through a gray PVC attenuator into a cylindrical capsule of test explosive. The cylindrical capsule had a 38 mm height and a larger diameter (200 mm) than the donor to eliminate wall interactions that could promote initiation.[8,9] The capsule height was also chosen to ensure that the shock arrived at the bottom of the charge before the edge effect from the 100 mm diameter donor reached the central part of the charge. Thus, the device was designed to observe detonation initiation and propagation in the axial direction for low- sensitivity explosives. PVC was used for the entire device because of its compatibility with a class of liquid explosives. The thickness of the attenuator was varied to control the strength of the shock transmitted into the test explosive. The bottom of the charge was sealed with a sheet of 0.25 mm thick Mylar mounted on a medium density fiberboard.
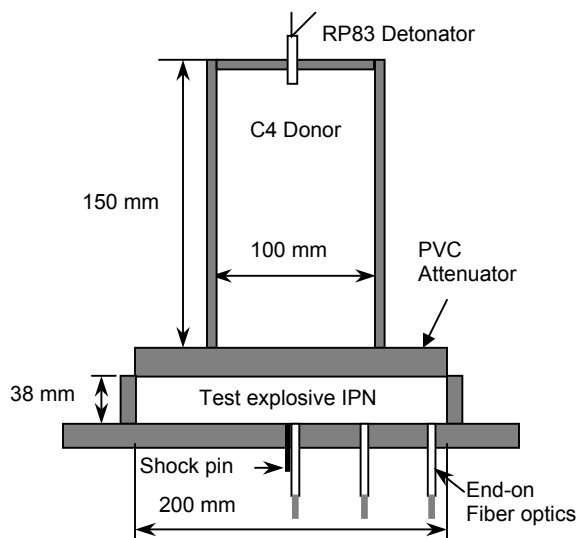

FIGURE 1. SHOCK INITIATION SET-UP.

The luminosity generated by detonation was detected via gauges consisting of photodiodes connected to fiber optics. The arrival time of the shock in the test explosive was determined by

1.5 mm diameter piezoelectric shock pins or by a pair of 0.1 mm diameter twisted wires. The end-on luminosity gauges were mounted at the bottom of the charge in brass light pipes. The photodiodes were sensitive enough to determine the onset of detonation but not weak chemical reactions. In the first series of experiments, three luminosity gauges were employed. These were located on the charge axis, at a radius of 38.1 mm, and at a radius of 76.2 mm, respectively. A shock pin was also mounted near the bottom center of the charge to monitor the arrival of the shock toward the end of the test period. In the second experimental series, only one fiber optic was positioned at the bottom center of the charge and four shock pins or twisted wire pairs were mounted at a radius of 6 mm, but at vertical distances of 5 mm, 10 mm, 15 mm, and 29 mm from the attenuator/IPN interface (not shown in Fig. 1). The shock attenuation curve in the PVC attenuator was also measured using contact gauges embedded in the attenuator. The shock velocities so obtained were then used to calculate the transmitted shock pressure by the impedance matching method with the PVC Hugoniot[10] and the universal liquid Hugoniot for IPN.[2,11]

Critical diameter test set-up

Critical diameters were examined using a donor/receptor arrangement, as shown in Fig. 2. Initiation was achieved via the detonation of a C4 explosive donor charge (1.57 g/cm^3 in density) that had the same diameter as the test explosive and was 1-1.5 diameters long. The receptor charge was 8-10 diameters in length for the neat IPN experiments and 6-8 diameters long for the IPN-aluminium mixtures. The receptors were made of thin-walled PVC tubes to provide only weak confinement. The thickness of the PVC tube walls varied from 8 mm to 3 mm for internal diameters ranging from 307 mm to 108 mm, and from 2 mm to 1 mm for internal diameters ranging from 55 mm to 12 mm. Charges less than 254 mm in diameter were installed vertically, whereas the larger charges were set up horizontally at a slight angle to facilitate charge filling. The detonation velocity was measured by contact gauges or piezoelectric shock pins mounted along the tube wall. For neat IPN in small diameter tubes, the end-on luminosity gauges were also installed at

the charge bottom to examine the quenching process. Selected experiments were performed as well with manganin gauges (Dynasen MN4-50-EK) mounted on a 19-mm thick PVC plate that was located at the charge bottom.
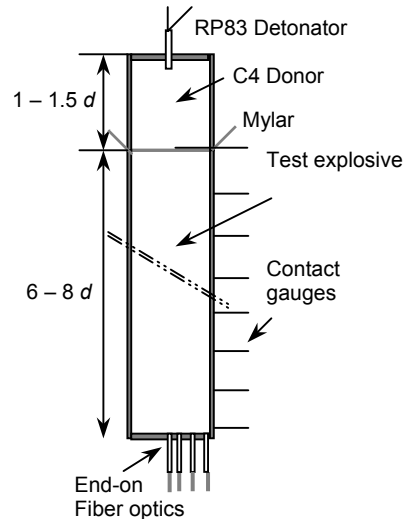

FIGURE 2. CRITICAL DIAMETER TEST SET-UP.

SHOCK INITIATION OF IPN

The IPN detonation velocity was first verified in a 51-mm diameter high impedance steel tube with a wall thickness of 3 mm. At 26 °C, the detonation velocity along the charge axis over a 0.5 m length (excluding the initial overdriven phase) is 5.6 mm/μs.

Shock initiation studies of IPN were carried out for four PVC attenuator thicknesses (6.24 mm, 12.48 mm, 18.72 mm, and 25 mm) for a temperature range between –5 °C and 5 °C. The results with a 6.24 mm attenuator that transmitted an incident shock pressure of 11.4 GPa are shown in Figs. 3 and 4. Note that the two figures represent two separate experiments. Time zero signifies the shock arrival at the donor/attenuator interface. The photodiode observing the charge via the central fiber optic detected a rapid rise in luminosity beginning at about 110 ns and 120 ns after shock entry, in the two respective shots. The details of the shock-detonation-transition cannot be distinguished given the current diagnostic resolution, and direct initiation can be considered to occur almost immediately after the strong shock enters the IPN. The initiated detonation is initially

in an overdriven mode, as indicated by the front peak luminosity, and decays to a C-J detonation after about 200 ns. The arrival of the detonation at the first shock pin, shown in Fig. 4, already occurs in the quasi-steady luminosity region, suggesting that the C-J detonation is established within the first 5 mm of the test liquid. The average detonation velocity over the subsequent distance from 5 mm to 29 mm is 5.56 mm/μs, as measured by four shock pins (not shown in Fig. 4). Note that no luminosity is observed at the luminosity gauge at a radius of 76.2 mm, suggesting that initiation was not promoted by wall interactions.
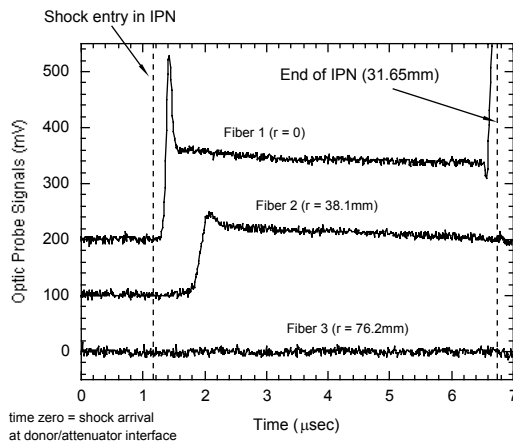
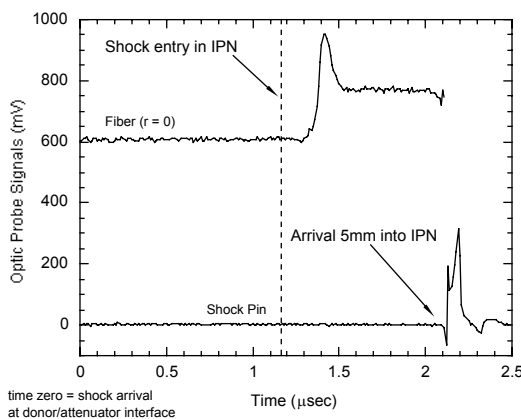FIGURE 3. END-ON LUMINOSITY SIGNALS AT THREE RADII FOR SHOCK INITIATION OF IPN AFTER A 6.24 MM PVC GAP.

When the thickness of the PVC attenuator is increased to 12.48 mm and 18.72 mm, the incident shock pressure is reduced to 9.9 GPa and 8.5 GPa, respectively. In this shock pressure range, unlike the case of direct initiation via a strong shock, a "hump" appears in the luminosity profiles detected by the central luminosity gauges, as shown in Figs. 5 and 6 for the case of 18.72 mm PVC attenuator. This hump is apparently associated with the onset of detonation that occurs at about 700 ns in Fig. 5 and at about 600 ns in Fig. 6. In Fig. 5, approximately 500 ns after the onset, a large spike in peak luminosity is observed, suggesting that the reaction propagating in the shock pre-compressed medium catches up to the leading shock front. Thereafter, the luminosity decays toward the quasi-steady value over more than 1 μs, indicating that the detonation propagates in an overdriven mode for a relatively long distance. In Fig. 6, the arrival of the shock front at the first shock pin occurs earlier than the catch-up event. This means that the catch-up takes place after the 5 mm position. The average shock velocity from 5 mm to 10 mm is 5.8 mm/μs, as measured by the first two shock pins, and reduces to 5.6 mm/μs between 10 mm and 29 mm, as measured by the last three shock pins. A distance-time diagram is displayed in Fig. 7 summarizing the information obtained from Figs. 5-6 and the impedance matching calculation based on a PVC shock attenuation experiment.

FIGURE 4. AN END-ON LUMINOSITY SIGNAL AND A SHOCK PIN SIGNAL AT 5 MM INTO IPN FOR SHOCK INITIATION AFTER A 6.24 MM PVC GAP.
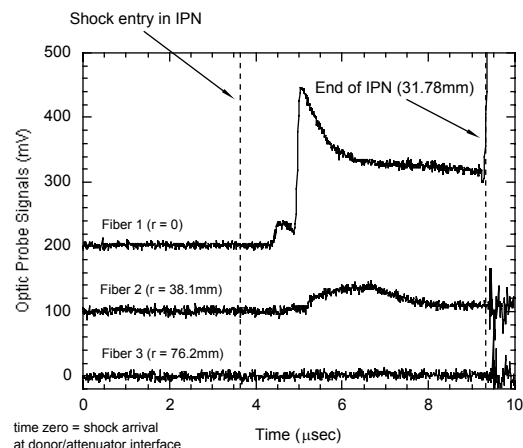
FIGURE 5. END-ON LUMINOSITY SIGNALS AT THREE RADII FOR SHOCK INITIATION OF IPN AFTER AN 18.72 MM PVC GAP.
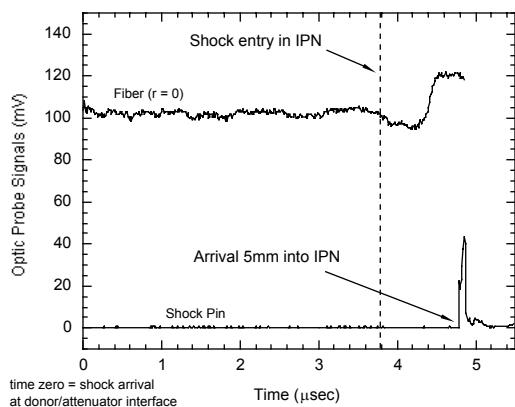
FIGURE 6. AN END-ON LUMINOSITY SIGNAL AND A SHOCK PIN SIGNAL AT 5 MM INTO IPN FOR SHOCK INITIATION AFTER A 18.72 MM PVC GAP.
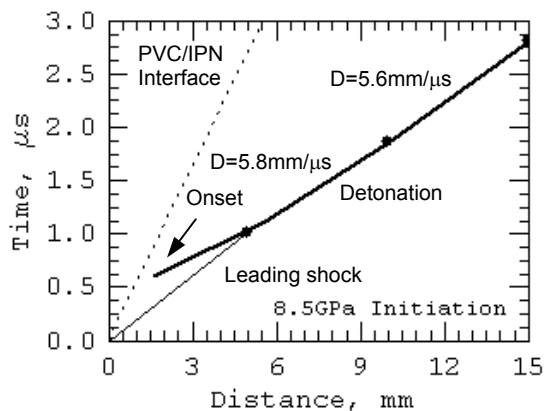

FIGURE 7. TIME-DISTANCE DIAGRAM BASED ON THE DATA PRESENTED IN FIGURES 5-6.

It is noted that the luminosity hump signifying the onset of detonation is considerably smaller in comparison with the peak luminosity corresponding to the catch-up event. Many studies of SDT in homogenous liquid NM have also suggested that there exists a detonation wave in the pre-compressed medium behind the shock (i.e., a "superdetonation").[12-15] The luminosity intensity for NM in the superdetonation stage was usually observed only mildly weaker than for the detonation of the uncompressed NM. Sheffield et al.[2] also observed the superdetonation stage in IPN using in-situ particle velocity measurements in gas gun experiments. To understand the low

luminosity hump observed in the present experiments, we performed equilibrium detonation calculations for precompressed IPN using the Cheetah code (version 2)[16] with the BKWS equation of state for the detonation products. For an initial shock pressure in the range of 7-12 GPa, the so-called superdetonation velocity, pressure and temperature vary in the range of 7.57-7.40 mm/μs, 25.0-26.1 GPa and 2641-2545 K, respectively. In comparison, the C-J detonation velocity, pressure and temperature for IPN at room conditions are calculated to be 5.85 mm/μs, 9.1 GPa and 2814 K, respectively. Hence, the chemical energy released in the superdetonation manifests more as mechanical energy of the detonation products rather than thermal energy, when compared with the C-J detonation states. If the detonation develops within the luminosity hump, its equilibrium limiting case would be the superdetonation with a temperature of 2600 K. While the low temperature of the precompressed IPN detonation will result in relatively weak luminosity, the small luminosity hump observed in the present experiments require more detailed quantitative measurements before firm conclusions can be drawn.
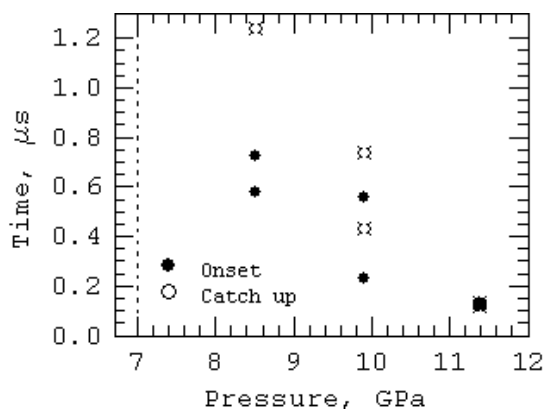

FIGURE 8. DEPENDENCE OF TIMES FOR THE ONSET OF DETONATION AND CATCH-UP ON SHOCK PRESSURES FOR SDT IN IPN.

When the attenuator thickness is further increased to 25 mm, which corresponds to a shock pressure of 7 GPa, no luminosity is observed at all and only a decaying shock propagates through the test charge. Thus, the critical gap thickness for shock initiation at 0 ± 5 °C must be in the range between 18.72 and 25 mm, and the critical shock

pressure is estimated to be between 7 and 8.5 GPa. The dependence on shock pressure of the characteristic times for the onset of detonation and the catch-up phase is summarized in Fig. 8.

The critical pressures and the dependence of the onset and catch-up times on shock pressure reported here refer only to the peak pressure. Since the use of point-initiated charges and attenuators involving lateral and rear-generated rarefactions will result in a shock followed by an expansion gradient, these values of critical pressure and the pressure-time relations cannot be directly compared to values for square-wave shock loading. Nonetheless, unambiguous shock initiation of detonation of IPN has been obtained on a large scale. This provides a means to examine the detonation initiation and propagation in relatively insensitive homogeneous explosives.

CRITICAL DIAMETERS FOR IPN

Critical diameter tests for IPN detonation were performed in low impedance PVC tubes with an internal diameter in the range between 100 and 307 mm. A detonation quenching process is displayed in Fig. 9 for a 100 mm tube with four equally spaced end-on luminosity probes positioned between the tube wall and axis of the charge, numbered 4 through 1, respectively. The detonation, which is initiated in IPN at 20 °C by a strong shock, is quenched by the expansion penetrating from the wall to the axis. It completely fails after 22 μs at a distance of approximately one tube diameter from the donor charge.

Figure 10 summarizes the detonation or shock velocities measured on the PVC tube wall for critical diameter tests at different temperatures. The detonation near the wall in a 250 mm diameter tube containing IPN at 5 °C fails at a distance of 1.5 charge diameters. In a similar test with IPN at 16° C in a 307 mm diameter tube, the detonation fails at a distance of 2.5 charge diameters. Thus, the critical diameter for IPN detonation in the low impedance PVC tube is larger than 307 mm. When compared with the critical diameter for NM detonation in a glass tube (20-25 mm),[4] the critical diameter for IPN is seen to be one order of magnitude larger than that for NM. This suggests that the reaction zone of the

IPN detonation is probably one order of magnitude larger than that for NM detonation as well.
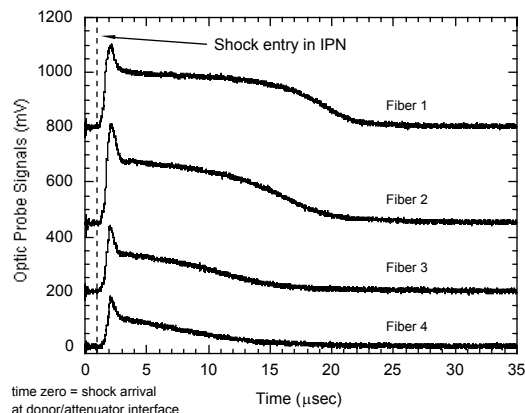

FIGRUE 9. AN IPN DETONATION QUENCHING PROCESS IN A 100 MM PVC TUBE WITH END-ON LUMINOSITY GAUGES EQUALLY SPACED ALONG THE CHARGE RADIUS.
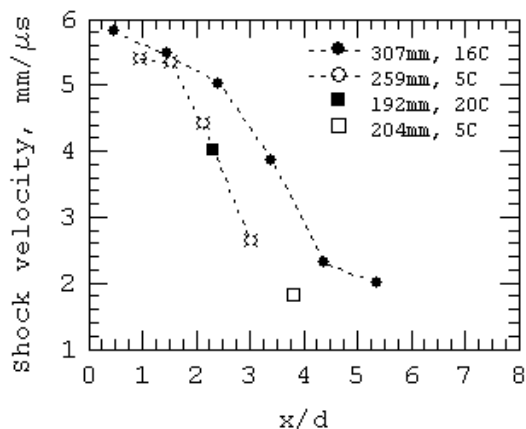

FIGURE 10. SHOCK VELOCITIES MEASURED ON THE TUBE WALL VERSUS DISTANCE (IN CHARGE DIAMETERS) FOR IPN CRITICAL DIAMETER TESTS.

IPN WITH ALUMINIUM ADDITIVES

Theoretical considerations

If the reaction length scales of combustible particles are larger than the detonation length scale of a homogeneous liquid explosive, the particles mixed in the liquid explosive have insufficient time to react within the reaction zone of the detonation and behave more like inert particles

that increase the detonation sensitivity but reduce the detonation velocity. While after-burning of the particles in the detonation products and surrounding air may enhance the near-field blast, the process involving particles is a complex multi length-scale problem. In the long-time limit, the effect of this after-burning can be described by the adiabatic equilibrium explosion pressure in a closed volume filled with air, assuming that heat losses to the wall are negligible. Sample calculations shown in Fig. 11 indicate that the explosion pressures for NM-Al or IPN-Al are generally higher than for TNT or RDX over an air-volume to charge-mass ratio ranging from 1 m^3/kg to more than 100 m^3/kg.
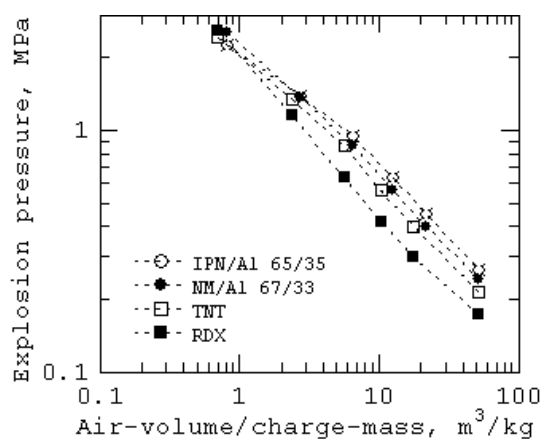

FIGURE 11. ADIABATIC EXPLOSION PRESSURES.

If the reaction length scales of the particles are smaller than or comparable to the detonation length scale of the liquid explosive, the particles will burn significantly within the reaction zone of detonation. In the equilibrium limiting case, the detonation properties can be obtained from equilibrium C-J detonation calculations. Since the critical diameter for IPN (> 310 mm in PVC tubes) is at least one order of magnitude larger than for NM, one might expect that the reaction zone of the IPN detonation would be close to 1 mm based on the reaction zone of 0.1 mm or less reported for NM.[17] Thus, it is possible that fine aluminum particles will have sufficient time to burn within the reaction zone of the IPN detonation.
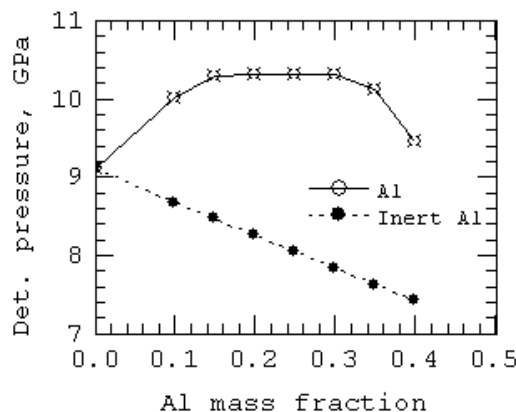

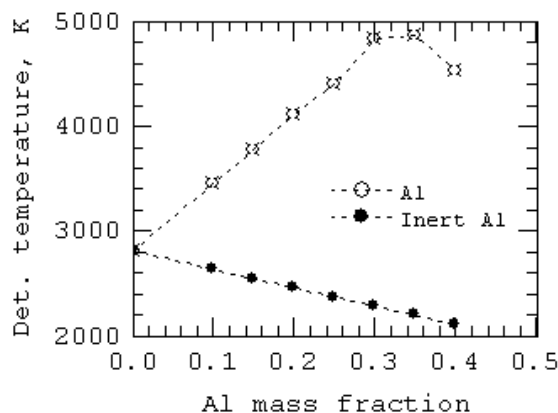FIGURE 12. EQUILIBRIUM C-J DETONATION PRESSURES.

FIGURE 13. EQUILIBRIUM C-J DETONATION TEMPERATURES.

Equilibrium C-J detonation calculations were carried out for a wide range of aluminum mass fraction in IPN using the Cheetah code with the BKWS equation of state.[16] Computed results for reactive aluminum and for aluminum assumed to be inert are displayed together in Figs. 12-14 for comparison. For both reactive and inert particles, the detonation velocity decreases monotonically with increasing particle mass fraction due to the negative oxygen balance. However, the detonation pressures and temperatures for reactive aluminum are significantly higher than for inert aluminum over the aluminum mass fraction range shown in Figs. 12-13. The pressure increases by 13% for a mass fraction of 0.2-0.3 and the temperature reaches its maximum increase of 73% for a mass fraction of about 0.3. Apparently, detonation velocity measurements alone do not provide sufficient information to evaluate the

effects of adding fine aluminum additives to a homogeneous explosive such as IPN. Figure 14 shows both the mechanical energy of detonation for reactive aluminum in IPN as well as the total detonation energy. The latter includes the mechanical energy as well as the thermal energy stored as heat in the detonation products at 1 atm. The mechanical energy of detonation is obtained at the 1 atm endpoint of the adiabat and represents the amount of energy available to do mechanical work. At an aluminum mass fraction of about 0.3, the mechanical energy achieves its maximum increase of 51%.
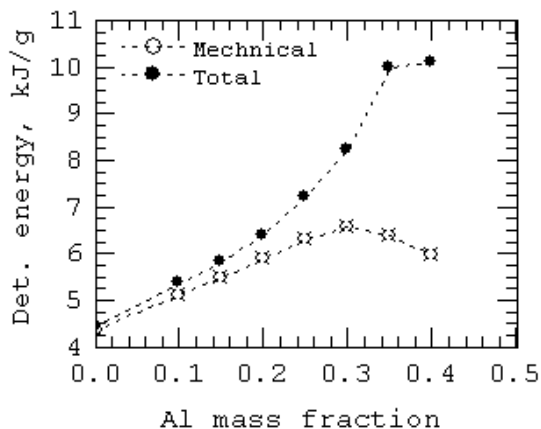

FIGURE 14. MECHANICAL ENERGIES OF DETONATION AND TOTAL DETONATION ENERGIES FOR REACTIVE AL IN IPN.

Unlike the equilibrium detonation parameters, the critical diameter is a dynamic parameter that depends on both the interaction of the leading shock with the particles and the competition between the surrounding expansion and the multiple time-scale processes of reaction and transport occurring between the IPN and aluminum particles within the reaction zone. Among the various sizes of aluminum particles available, the fine aluminum particles, which can combust and equilibrate within the reaction zone of the detonation, may be of most interest in studying the influence on the critical diameter of homogeneous liquid explosives. In this case, although the detonation velocity, pressure and temperature are completely determined by equilibrium considerations, the detonability must depend on the two-phase interactions during the process of the particles crossing the leading shock

front and the interaction between the surrounding expansion with the mass, momentum and heat transport occurring between the phases within the reaction zone.

Experimental results

A nanometric grade of aluminum known as "Alex" supplied by Argonide Corporation was employed to examine its influence on the detonation of IPN. This material is manufactured by an exploding wire process and has a mean particle diameter of approximately 100 nm. Alex was chosen for two reasons: (i) its mass fraction in a packed bed saturated with IPN approaches 0.24-0.32 which gives the best performance in terms of equilibrium detonation parameters, and (ii) these are the smallest aluminum particles commercially available and will likely burn significantly within the reaction zone of the IPN detonation.
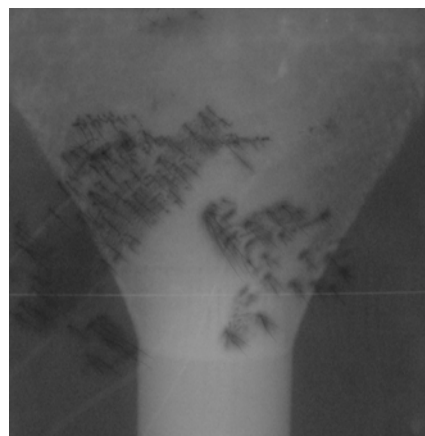

FIGURE 15. A FLASH X-RAY RADIOGRAPH FOR DETONATION IN IPN/ALEX IN AN 84-MM DIAMETER PVC TUBE. THE DARK CLUSTERS VISIBLE ARE PRODUCED BY THE DISCHARGE OF STATIC ELECTRICITY THAT BUILDS UP BETWEEN THE X-RAY FILM AND SCREEN DURING BLAST WAVE LOADING.

Figure 15 shows a flash radiograph for an experiment of a stable IPN/Alex detonation in an 84 mm diameter, thin-walled PVC tube. The detonation propagates quasi-steadily throughout the entire length of 6 diameters with an average velocity of 4.94 mm/μs. No significant afterburning of aluminum is distinguishable in the fairly uniform detonation products. Figure 16

displays a pressure history measured with the manganin gauge mounted on a 19-mm thick PVC plate that was located at the bottom of a 48-mm diameter charge. The transmitted pressure history also does not show significant afterburning of aluminum.
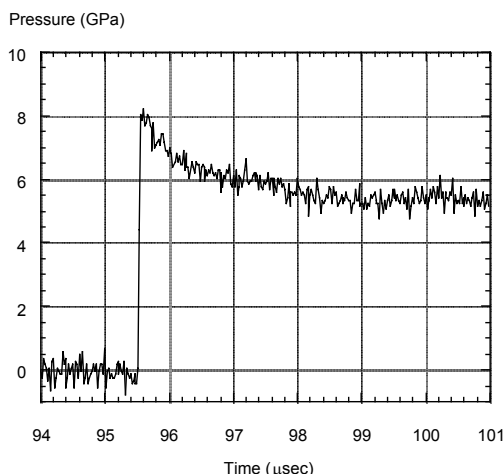

FIGURE 16. PRESSURE HISTORY ON A 19 MM PVC PLATE LOCATED AT A 48-MM DIAMETER IPN/ALEX CHARGE BOTTOM.
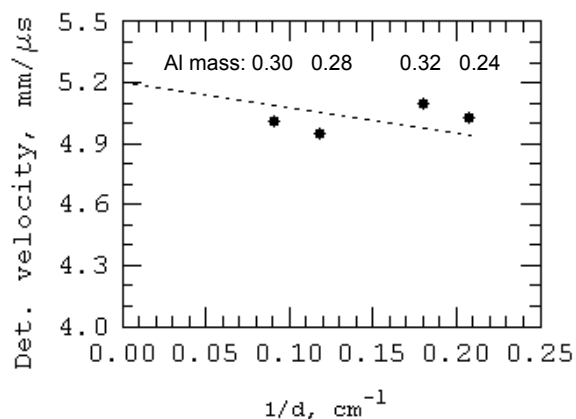

FIGURE 17. DETONATION VELOCITY VERSUS INVERSE CHARGE DIAMETER FOR IPN/ALEX IN PVC TUBES.

Figure 17 summarizes the detonation velocity variation with PVC tube diameter for the IPN/Alex with an aluminum mass fraction of 0.24-0.32. Tests were carried out down to a tube diameter of 48 mm and all tests exhibited stable detonations over the entire tube length (6-8 diameters). Detonation fails at a tube diameter of 29 mm. Thus, the critical diameter for IPN/Alex

detonations lies between 29-48 mm and is almost one order of the magnitude smaller than that for neat IPN. The detonation velocity extrapolated to infinite diameter is estimated to be ca. 5.2 mm/μs. This is close to the calculated equilibrium prediction of 5.28 mm/μs using the Cheetah code with the BKWS equation of state.

CONCLUSIONS

Nitromethane has been used for a long time as a model for the study of detonation physics in homogeneous liquid explosives. Although modern diagnostic technology is now allowing some of the details of the reaction zone of NM detonation to be resolved, the very small detonation length scale (ca. 6-15 ns[17]) still presents a challenge in terms of elucidating the mechanism of detonation. Liquid isopropyl nitrate (IPN) is considered a practical alternative for fundamental studies of detonation due to its larger detonation length scale which facilitates higher diagnostic resolution. In the present paper, the detonability of IPN has been investigated, including shock initiation of detonation and determination of the critical diameter in low impedance PVC tubes.

Two modes of shock initiation of detonation have been observed: direct initiation of detonation, and SDT with a build-up or onset process involving detonation in the shock precompressed medium. While the SDT process is similar to that for NM reported in the literature, the reaction thermal energy in the precompressed medium is much lower than that observed for NM. The SDT process, from the onset of detonation, through the overdriven mode, and finally to C-J detonation, occurs in a transition distance of about 10-15 mm near the critical shock pressure. Limited data for the initial shock pressure versus the time of onset of detonation and the catch-up time have been obtained. The critical shock initiation pressure is estimated to be in the range of 7-8.5 GPa at 0±5 °C. The critical diameter for neat IPN was found to exceed 310 mm at 16 °C in a low impedance PVC tube. Thus, the critical diameter for IPN is at least one order of magnitude larger than that for NM.

The large reaction zone of the IPN detonation provides an opportunity for nanometric grades of aluminum to burn toward equilibrium within the

detonation reaction zone when these particles are mixed with IPN. Experiments involving such mixtures can serve to enhance our understanding of detonation in homogeneous liquids versus heterogeneous particle-additive liquids.

Equilibrium calculations predict a maximum mechanical energy of detonation in IPN/Al mixtures 1.5 times greater than for neat IPN detonation for an aluminium mass fraction of 0.3. It is possible to achieve this mass fraction using 100 nm (average) aluminium particles known as Alex in the form of a packed bed saturated with IPN. The critical diameter for IPN/Alex mixtures was found to lie between 29 mm and 48 mm in a thin-walled PVC tube in the present experiments, a reduction in critical diameter of almost one order of magnitude in comparison with neat IPN.

ACKNOWLEDGEMENTS

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