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HUGONIOT AND SHOCK INITIATION STUDIES OF ISOPROPYL NITRATE[†]

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Isopropyl nitrate (IPN) is a liquid explosive of rather low energy. We have measured the sound speed and used it in the universal liquid Hugoniot to produce an estimated Hugoniot for this material. Gas-gun-driven, multiple-magnetic-gauge measurements were made to measure a Hugoniot state at 6 GPa; it was in good agreement with the prediction. Two similar experiments were conducted at higher pressure inputs to study the shock-to-detonation transition in IPN; the high inputs required for initiation necessitated the use of a two-stage gun. One experiment with an input of 9.0 GPa into the IPN produced a run to detonation of about 3 mm and the in-situ particle velocity profiles showed the expected homogeneous initiation behavior of a growing wave behind the shock front that overtakes the front and decays to a steady detonation. The reactive wave in the shocked IPN appears to have achieved a steady superdetonation in both of the initiation experiments. This is the first time a steady superdetonation has been measured with in-situ gauges.

INTRODUCTION

Liquid explosives have been of interest for many years because they offer the opportunity to experiment with a homogeneous material and determine the properties of the material without the influence of other phenomena that might lead to hot spots (heterogeneous behavior). Nitromethane (NM) has typically been the explosive of choice because it is easy to obtain and has been extensively studied. Most other studies of liquid explosives have concentrated on critical diameter, gap test sensitivity, detonation velocity, initial temperature effects, etc., and have not studied in detail the shock initiation properties. In this study we have concentrated on isopropyl nitrate to more carefully study its shock and reaction properties. This information has led to increased understanding of IPN as well as adding important new information on the homogeneous initiation process.

Liquid isopropyl nitrate [(CH₃)₂CHONO₂] (IPN) (see Fig. 1) is a rather low energy explosive because it does not have a good oxygen balance. It has been

used in propellants or as a monopropellant. A rather large study of the detonation (failure) properties of IPN in steel and glass tubes as a function of temperature was reported in Ref. 1. Unfortunately, the accuracy of the detonation velocity measurements was only 3 to 5%, less than one would hope because NM has been shown to have a velocity change (velocity deficit) of only 1% from infinite diameter to failure diameter in glass.² We have taken the liberty of replottting some of the room temperature data from Ref. 1 to get some idea of what the diameter effect curve is for IPN in steel tubes; the data are shown in Fig. 2. The detonation velocity in steel tubes is between 5.3 and 5.4 mm/μs, depending on the tube diameter. The line stops at roughly the critical

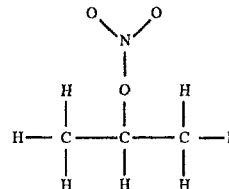


FIGURE 1. Chemical structure of isopropyl nitrate.

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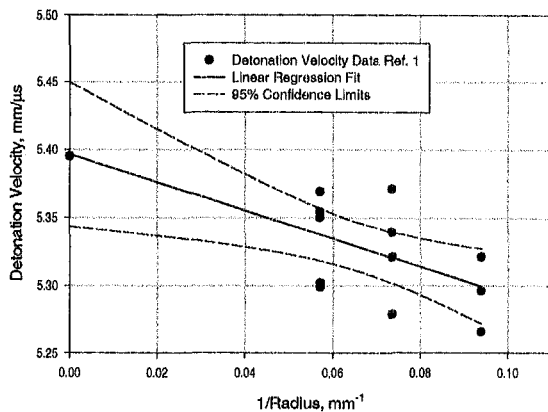


FIGURE 2. Diameter effect curve for IPN in steel tubes at room temperature. Data are from Brochet in Ref. 1.

diameter in steel, about 10 mm radius (20 mm diameter). The velocity deficit for IPN appears to be about 2%, somewhat larger than that for NM but comparable.

In Ref. 1, the IPN detonation velocity was measured to drop about 0.2 mm/μs when the temperature increased from room temperature to 350K; about -3.3 m/s/K. It was stated that the change was due to density changes as a function of temperature. NM detonation velocity drops with increased temperature at a rate of -3.7 m/s/K. (Ref. 3) This is thought to result mostly from density change but there is also a contribution due to internal energy changes with temperature.²

EXPERIMENTAL DETAILS AND DATA

Sound speed measurements at ambient conditions were made on liquid IPN to provide the information necessary to estimate the Hugoniot. Magnetic gauge experiments were completed using both our single-stage and two-stage guns to measure the Hugoniot state and provide shock initiation information. Information about and data from these experiments are presented below.

Material – Liquid IPN was obtained from Aldrich Chemicals with a purity of 99 wt% IPN. It was used as received. IPN has an initial density at room conditions of 1.036 g/cm³, a boiling point of 101–102 °C and a freezing point of 12 °C. IPN is a colorless liquid with a viscosity about like water.

Sound Speed and Estimated Hugoniot – An estimate of the Hugoniot for this material was obtained using the universal liquid Hugoniot.⁴ This

empirical equation has the form

$$U_s = C_0 \{1.37 - 0.37 \exp(-2u_p/C_0)\} + 1.62u_p$$

where U_s is the shock velocity, u_p is the particle velocity, and C_0 is the room condition sound speed – the only required parameter. It has been shown to provide reliable Hugoniot estimates for essentially all the liquids for which shock data are available. The room condition sound speed for IPN was measured (see Ref. 5) to be 1.10 mm/μs.

Gun Experiments/Liquid Cell Design – In the gun experiments of this study, the input shock was produced by impacting an impactor-faced projectile on a plastic cell containing the liquid IPN. The diameter and depth of the liquid sample was such that edge effects did not complicate the measurements, i.e., they were designed to be 1-D.

The cell body was made from two pieces of PMMA which were machined to fit together with the gauge membrane epoxied between them. The design was such that the suspended membrane was a plane inclined 30° with respect to the cell front (impact plane). The cell front was made from either PMMA or Kel-F, depending on the input desired into the IPN. An epoxy coating was put on the inside of the cell to isolate the IPN from the PMMA. The cell front was epoxied and screwed (with nylon screws) to the cell body. Fill holes were located on the side of the cell. Details of how these experiments were done are contained in Refs. 6 and 7.

In each experiment the gauge membrane included ten gauges and a “shock tracker.” Another gauge (called a “stirrup” gauge) was epoxied on the back of the cell front to measure the input to the IPN. This setup provided a total of eleven in-situ particle velocity gauges and the shock tracker, which provides data to use in constructing a distance vs. time (x-t) plot of the shock front as it moves through the IPN. The particle velocity gauges provide dynamic information relating to the state of the shocked IPN (which may be reacting) at specific Lagrangian positions.

There are some difficulties in using an inclined gauge membrane suspended in a liquid. The membrane is composed of FEP Teflon. It has been shown that if the liquid is the same shock impedance as the gauge, the gauges provide an accurate

measurement of the particle velocity. If the liquid is a higher impedance, the gauge measures high and if the liquid is lower impedance, it measures low.⁸ The errors can be up to $\pm 10\%$, depending on the impedance difference. IPN is a lower impedance than the membrane so the gauges read low in this material. These errors in the measurement are due to slippage at the gauge plane; this happens in experiments with liquids but not those involving solid materials.

IPN Gas Gun Experimental Data – Three multiple-magnetic-gauge gun experiments were completed. One was done on the single-stage gun (Shot 1129) below the condition where reaction was initiated to confirm that the estimated Hugoniot was correct. Two higher pressure input experiments were completed on the two-stage gun (Shots 2s-28 and 2s-29) to measure the details of the shock-to-detonation transition in IPN.

Shot 1129 provided Hugoniot a point to compare to the estimated Hugoniot. This shot involved a Z-cut single-crystal sapphire impactor hitting a Kel-F cell front at a velocity of 1.408 mm/ μ s. A stirrup gauge on the cell front, in contact with the IPN, measured a particle velocity input of 1.50 mm/ μ s to the IPN. The membrane gauges measured about 8.5 % lower than this. The shock tracker provided a value for the shock velocity of 3.896 mm/ μ s. Using these values, the IPN input pressure was 6.05 GPa. This Hugoniot point is plotted on Fig. 3 along with the universal liquid Hugoniot prediction. It is obvious that the predicted Hugoniot is accurate.

On Shot 2s-28 the input shock to the IPN was generated by a Kel-F impactor on the projectile hitting a PMMA cell front. Unfortunately, the projectile velocity measurement failed so we can only estimate the impact velocity to be between 2.75 and 2.85 mm/ μ s. Particle velocity measurements from this experiment include the stirrup gauge at the beginning of the IPN and ten gauges in the IPN. The particle velocity waveforms from all eleven gauges are shown in Fig. 4. In addition, a shock tracker measured the progress of the shock as it moved through the IPN. The initial shock velocity was 4.8 mm/ μ s, the particle velocity (as shown in Fig.4) was about 1.6 mm/ μ s, so the initial shock pressure was about 8 GPa. The waveforms in Fig. 4 show that the

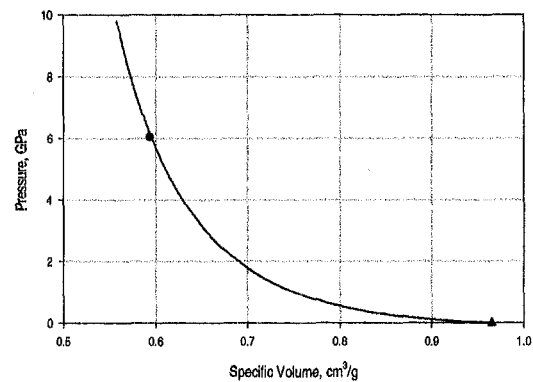


FIGURE 3. P-V Hugoniot plot for IPN. The curve is obtained from the universal liquid Hugoniot and the data point is that measured in Shot 1129. The initial specific volume is shown as a triangle.

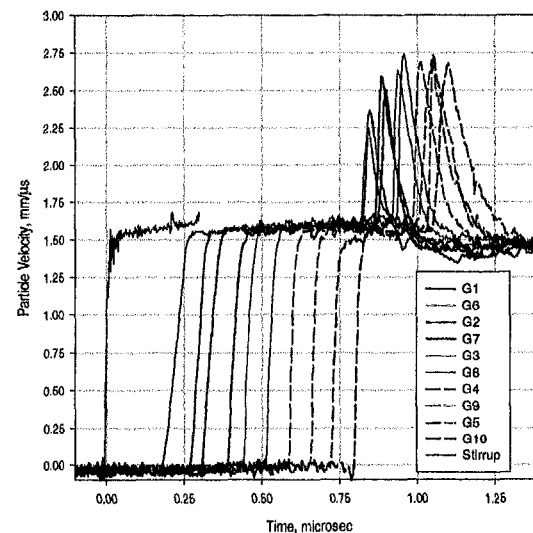


FIGURE 4. Particle velocity waveforms obtained from Shot 2s-28. Eleven gauges are shown – the first gauge is the stirrup gauge at the input face of the IPN and the other ten gauges are in-situ in the IPN at positions from 0.8 to 4 mm deep.

superdetonation achieved a steady state and, using the gauge positions and arrival times, a velocity of about 10 mm/ μ s was measured for this wave as it moved through the initial state. Data from the shock tracker was used to determine that the detonation immediately after overtake had a velocity of 6.9 mm/ μ s, which decreased to a steady 5.34 mm/ μ s by the end of the shock tracker data. The position of overtake was 5.7 mm into the IPN.

Shot 2s-29 produced the particle velocity waveforms shown in Fig. 5. The projectile velocity

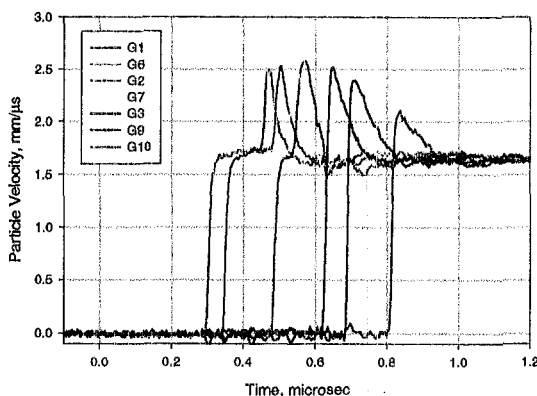


FIGURE 5. Particle velocity waveforms obtained from Shot 2s-29. Seven gauges are shown, all of them are in-situ in the IPN at positions from 1.3 to 4.5 mm deep. The stirrup gauge broke so the input to the IPN was not recorded.

was 2.97 mm/μs with a Kel-F impactor hitting a Kel-F cell front and providing a shock of 8.9 GPa into the IPN. Several gauges failed in this experiment so they are missing from the figure. However, the shock tracker provided information about the shock front propagation. The input particle velocity was measured at 1.73 mm/μs; it was calculated to be 1.89 mm/μs, about 8.3% low. The input shock velocity was 4.5 mm/μs. The waveforms show the superdetonation achieved a steady velocity of about 8 mm/μs. After overtake of the initial wave, the overdriven velocity was 6.3 mm/μs which decreased to a steady 5.34 mm/μs by the end of the shock tracker. The position of overtake was 2.7 mm into the IPN.

DISCUSSION OF RESULTS

From the low pressure experiment, it is obvious that the universal liquid Hugoniot is a good estimate of the IPN unreacted Hugoniot. The shock initiation experiments both showed the same initiation behavior. A reactive wave builds behind the initial input shock to a steady superdetonation (steady velocity 8 to 10 mm/μs) which overtakes the initial shock, producing an overdriven detonation (6.3 to 6.9 mm/μs) that decreases to a steady detonation of 5.34 mm/μs. This agrees with the process proposed earlier for NM.⁹ In this study of IPN, the superdetonation reached a steady velocity, the first time this has been measured with in-situ gauges. The steady detonation velocity of 5.34 mm/μs would be

expected to be the infinite diameter velocity because the experiments are 1-D. This value agrees reasonably well with the earlier data of Fig. 2.

When the three experiments are considered as a group, there are some inconsistencies. Shots 1129 and 2s-29 agree with each other in that the input particle velocities measured by the gauges are about 8 % low, when compared to what was expected; this is as it should be for liquid IPN. However, Shot 2s-28 was different. The particle velocity measured by the in-situ gauges agreed with the stirrup gauge as one would expect for a solid and the measured particle velocity and shock velocity were not what was expected. We have conjectured that this may be the result of freezing in the initial wave on Shot 2s-28. This means the initiation occurred in the solid state rather than the liquid state, producing a faster superdetonation. However, this is highly speculative and should be considered suspect until additional experiments are completed.

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REFERENCES

1. Brochet, C., *Astronautica Acta*, **15**, 419 (1970).
2. Engelke, R., *Phys. Fluids* **23**, 875 (1980).
3. Campbell, A. W., Malin, M. E., and Holland, T. E. *Proceedings of the Second Symposium on Detonation*, Naval Surface Warfare Center Report NSWC MP 87-194, pp. 454-471 (Reprinted in 1987, Short, J., Editor).
4. Woolfolk, R. W., Cowperthwaite, M., and Shaw, R., *Thermochimica Acta* **5**, 409 (1973).
5. Engelke, R., Sheffield, S. A., and Davis, L. L., *J. Phys. Chem. A* **104**, 6894 (2000).
6. Sheffield, S. A., Gustavsen, R. L., and Alcon, R. R., *Shock Compression of Condensed Matter - 1999*, Eds. Furnish, M. D., Chhabildas, L. C., and Hixson, R. S., AIP Proceedings 505, p. 1043 (2000).
7. Sheffield, S. A., and Alcon, R. R., *Shock Compression of Condensed Matter - 1989*, Eds. Schmidt, S. C., Johnson, J. N., Davison, L. W., North-Holland, Elsevier Science Publishers B. V., Amsterdam, The Netherlands, p. 683 (1990).
8. Gustavsen, R. L., Sheffield, S. A., and Alcon, R. R., *High Pressure Science and Technology - 1993*, Eds. Schmidt, S. C., Shaner, J. W., Samara, G. A., Ross, M., AIP Conference Proceedings 309, p. 1703 (1994).
9. Sheffield, S. A., Engelke, R. and Alcon, R. R., *Proceedings of the Ninth Symposium (Intl.) on Detonation*, Office of Naval Research OCNR 113291-7, pp. 39-49 (1989).