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We performed experimental studies on the stability of the detonation wave front in mixtures of the liquids tetranitromethane (TNM) and nitrobenzene (NB). Tetranitromethane is an oxygen-rich explosive and nitrobenzene was used as a solvent or dilutant. (NB is not classed as an explosive but as an explosive would be oxygen poor and fuel rich.) The primary diagnostic was a laser velocimetry method with high temporal resolution. Data obtained were compared with the detonation parameters of the TNM/NB mixtures.

In previous experimental work [1,2] it was shown that the detonation wave front in liquid explosives may be either smooth or rough. Rough detonation fronts have been reported in nitromethane, as well as nitromethane mixed with a solvent. Smooth detonation fronts have been reported in tetranitromethane. Previously, we conducted studies on the structure of the detonation wave front in liquid explosives containing tetranitromethane [3–5]. Smooth, stable fronts were recorded in pure tetranitromethane and in a 46/54 mixture of tetranitromethane and nitromethane. A

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pulsating, unstable detonation wave front was recorded in a 74/26 mixture of tetranitromethane and nitrobenzene. The goal of the present work is to extend our research on the structure of the detonation wave front in mixtures of tetranitromethane diluted with less energetic nitrobenzene. To this end, the following TNM/NB mixtures were studied: 95/5, 90/10, 85/15, 80/20, 74/26, and 50/50.

Keywords: detonation, initiation, liquid HE, nitrobenzenes, nitromethane, tetranitromethane, unstable detonation

Experimental Setup

Figure 1 presents a schematic of the experimental setup. An electric detonator initiates detonation in a layer of plasticized pentaerythritol tetranitrate (PETN). This, in turn, launches a detonation into the liquid explosive mixture. After traversing the liquid explosive sample, the detonation wave interacts with a window made of lithium fluoride (LiF). The velocity of the LiF/explosive interface, $U_{LiF}(t)$, is measured using the Fabry-Perot laser interferometry method [3–5]. The temporal resolution of our system is $\sim$1 ns. In all cases, a spherically diverging detonation wave propagates in the liquid explosive.

Further details of the experiment are as follows. The internal diameter of the liquid high explosive (HE)-containing cells

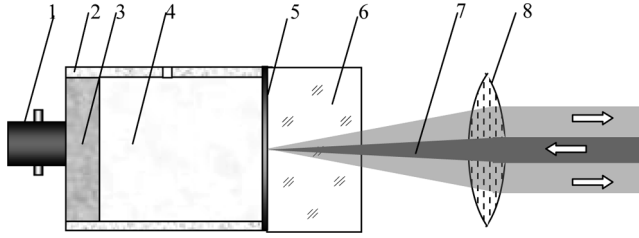

Figure 1. Schematic of the experimental setup. 1. electric detonator; 2. cylindrical cell; 3. plasticized PETN, 4. liquid explosive mixture; 5. aluminum coating, 0.5–1.5μm thick; 6. LiF single crystal; 7. laser beam, $\gamma = 694.3$ mm; 8. focusing lense.

varied from 6 to 15 mm. The liquid thickness was a minimum of 2 mm and a maximum of 70 mm. PETN thicknesses, liquid thicknesses, and the cell diameter were varied to try to get steady detonations. A 20-μm-thick aluminum foil was placed between the plasticized PETN and the liquid explosive. The reflector at the LiF/explosive interface was vapor-deposited aluminum with a thickness of ~1 μm.

Experimental Results

TNM/NB Mixtures with NB <20%

For tetranitromethane (TNM)/nitrobenzene (NB) mixtures with less than 20% nitrobenzene, a stable detonation front was recorded. For the 95/5, 90/10, 85/15, and 80/20 compositions, the ~17 GPa shock from the PETN pellet initiated detonation in the liquid explosive. The von Neumann spike pressure was estimated to be close to that measured in pure TNM: $P = 21.6$ GPa [3].

The critical shock wave pressure to initiate pure TNM is 8 GPa and the detonation pressure is $P_{C\text{-}J} = 16$ GPa [2,6]. With increasing NB content, the critical pressure to initiate detonation increases. Detonation velocity and pressure also increase with increasing NB content. This is in accordance with Eremenko et al. [7]. With NB <20%, the critical pressure of initiation does not exceed detonation pressure. The result is a smooth and stable detonation front. As will be shown below, if NB content is more than 20%, the critical pressure of initiation exceeds the detonation pressure and the shock wave front becomes unstable and pulsating.

The 74/26 TNM/NB Mixture

The 74% tetranitromethane, 26% nitrobenzene mixture is very close to the stoichiometric composition of 76.85/23.15. Among compositions of TNM/NB this has the maximum detonation velocity, $D = 7.5$ km/s. The density of this composition is $\rho = 1.51$ g/cm^3.

Figure 2 shows a detonation wave profile characteristic of a steady detonation with a smooth front. This result was

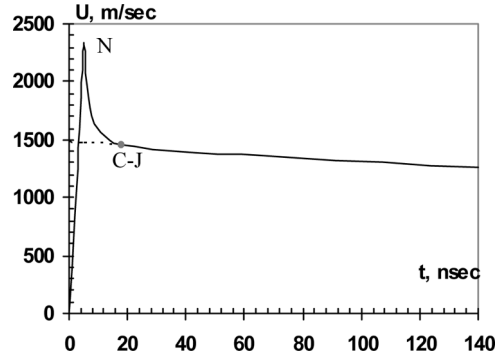

Figure 2. Detonation wave profile in 74/26 TNM/NB. This is believed to be a steady detonation with a smooth front. The result is not typical of this mixture.

obtained in one experiment on the 74/26 TNM/NB mixture. It is not typical of the mixture but is shown to orient the reader to what the profile from a steady, smooth detonation wave looks like. The configuration for this experiment was 11 mm of plasticized PETN and 50 mm of liquid explosive.

Detonation wave profiles are interpreted using pressure–particle velocity (P-U) diagrams. The P-U diagram for the 74/26 TNM/NB composition is presented in Fig. 3 and will be used for reference when explaining the experimental observations. Unreacted Hugoniots for the liquid explosive were calculated using mixtures of the Hugoniots for TNM and NB. Hugoniots for two of the compositions studied in detail are as follows:

$$\text{for 74/26 TNM/NB: } U_S = 1.897 + 1.767U_P$$

$$\text{for 50/50 TNM/NB: } U_S = 2.207 + 1.517U_P$$

The calculated value of the von Neumann peak was $P_N = 36$ GPa for the 74/26 composition and $P_N = 26$ GPa for the 50/50 composition. Note that all experimental observations must lie on the Hugoniot of the LiF window. States in the

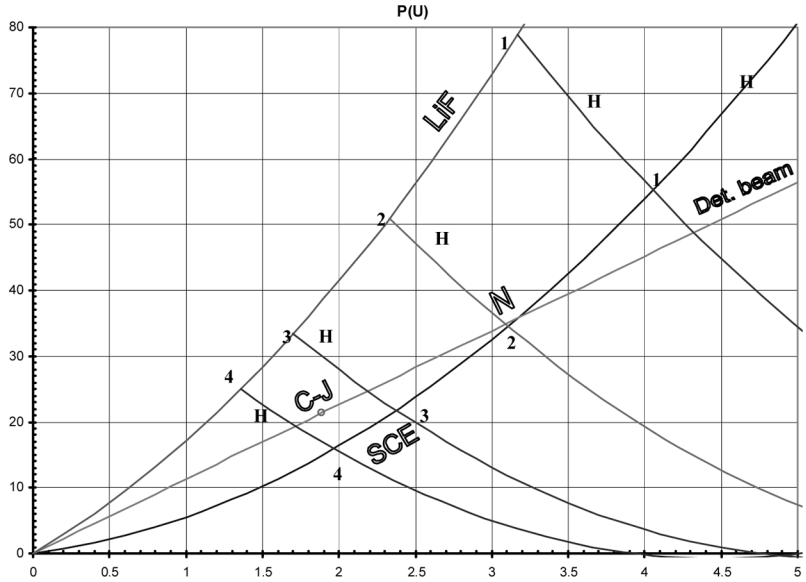

Figure 3. Pressure–particle velocity (P-U) diagram for the 74/26 TNM/NB mixture. Det. beam = detonation beam or Rayleigh line; H = unreacted Hugoniot of 74/26 TNM/NB; LiF = Hugoniot of LiF; C-J = Chapman-Jouguet state; N = von Neumann spike; SCE = range of shock-compressed explosive; 1, 2, 3, 4 = wave amplitudes in explosive; 1′, 2′, 3′, 4′ = states matched onto LiF window.

explosive before the wave interacts with the window are determined by impedance matching calculations.

In the wave profile shown in Fig. 2, we see a von Neumann spike (N) value of ∼2.3 km/s. The rise time of the shock front is $\delta \approx 5$ ns. Using the P-U diagram (Fig. 3) the von Neumann spike pressure in the explosive is ∼34 GPa. The von Neumann spike is followed by a drop in particle velocity to the Chapman Jouguet (C-J) state. The C-J state in the explosive is calculated to be 21.4 GPa, which impedance matches to ∼1.5 km/s on the LiF Hugoniot. The length of the chemical reaction zone from the von Neumann spike to the C-J state is therefore $\Delta t \approx 13$ ns. Taylor wave unloading follows the C-J state. (Note that most

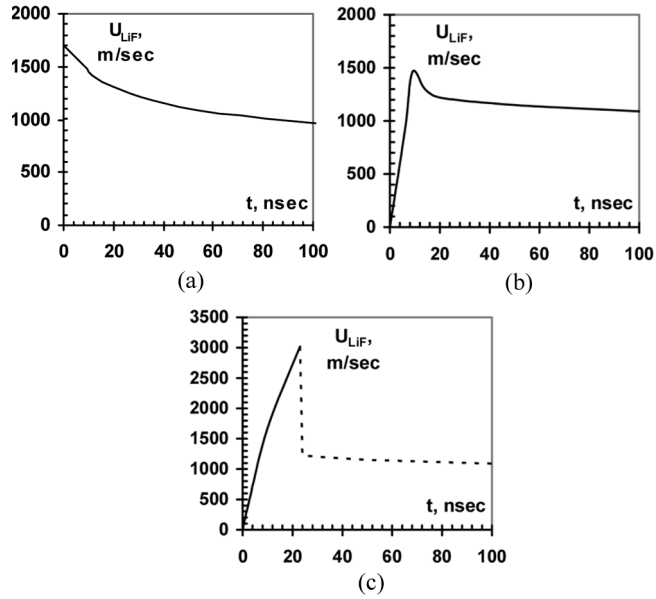

Figure 4. Characteristic types of particle velocity wave profiles in 74/26 TNM/NB. (a) Sharp front: These profiles are characterized by a very fast <1 ns rise followed by a smooth decrease in particle velocity. At first, they appear to be a classic von Neumann spike and Zeldovich-Von Neumann-Doering (ZND) reaction zone (in the picture of Sheffield et al. [5]) Comparison with wave profiles from Fig. 2 and the P-U diagram of Fig. 3 indicate that the peak particle velocity is too low to be the von Neumann spike. Peak particle velocities at the LiF/explosive interface ranged from 1.4 to 1.7 km/s and correspond to peak pressures of 16 to 22 GPa in the explosive. These particle velocities and pressures correspond to the region marked shock compression explosive (SCE) or C-J on the P-U diagram (Fig. 3) and indicate that the von Neumann spike state was not reached. (b) Rounded peak: These profiles have a 3–9 ns rise time; a smooth, rounded peak; and then a smoothly decreasing velocity. Peak particle velocities at the LiF/explosive interface range from 1.46 to 1.68 km/s. Peak pressure and particle velocities in the explosive are in the range marked C-J or SCE in Fig. 3. (c) Interrupted recording: The third type of profile is

researchers report a very short rise, <1 ns, to the von Neumann spike state. See, for example, Sheffield et al. [5]).

For all other experiments on the 74/26 TNM/NB mixture, unstable pulsating detonations were inferred. Wave profiles at the LiF/explosive interface can be grouped into three types, which are shown in Fig. 4. Appearance of a particular type of profile was unpredictable and not based on PETN thickness or liquid thickness.

The 50/50 TNM/NB Mixture

For the 50/50 mixture of tetranitromethane and nitrobenzene, wave profiles can be grouped into three types as above. Examples are shown in Fig. 5 and will be described below.

Discussion of Results

In explaining the variety of wave profiles observed, we start with the assumption that three-dimensional structures form in unstable pulsating detonations. We combine this with the idea that the laser beam from the Fabry-Perot interrupts a small portion of a structure as it collides with the explosive/LiF interface. Figure 6 shows a qualitative picture of several possible structures that could form in pulsating detonations [5,7]. The position where the laser beam interrupts the structure and what

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characterized by a long 5–25 ns rise followed by abrupt termination of recording. Peak particle velocities (at the point of interruption) ranged from 0.5 to 3.17 km/s. High interface velocities (>3.0 km/s) can be explained by propagation of oblique (or transverse) detonation waves moving through precompressed HE. Such waves are known as superdetonation waves driven by Mach stem wave interactions [6,8]. For the interrupted structure with greater than 3 km/s interface velocity, pressure in the explosive can exceed 49 GPa. Similar maximum pressures ($P \approx (50\,GPa)$ have been reported for superdetonations in shock precompressed nitroglycerine [2,6].

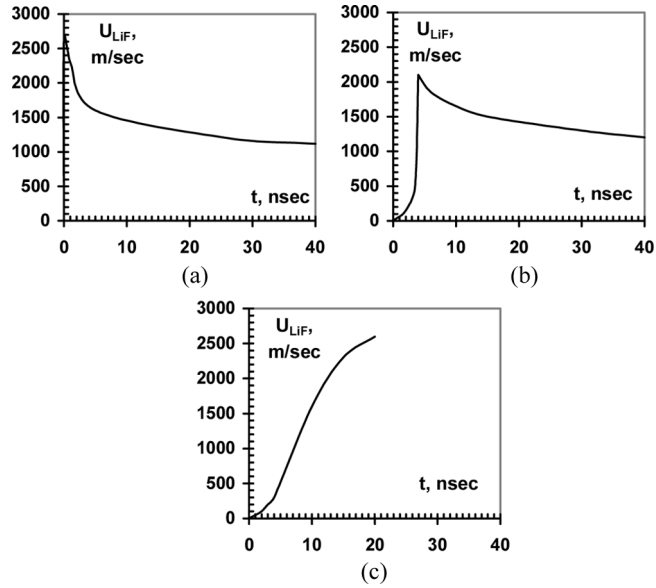

Figure 5. Characteristic types of particle velocity wave profiles in 50/50 TNM/NB. (a) Sharp front: This class of profile is characterized by a sharp <1 ns rise. The profile shown was from an experiment in which the liquid explosive was initiated with a 3-mm-thick layer of plasticized PETN. The peak interface velocity at the explosive/LiF interface was 2.62 km/s, corresponding to a pressure of ∼34 GPa in the explosive. An estimates of $P_{C\text{-}J}$ with a polytropic equation of state and index $n = 3$ gives $P_{C\text{-}J} =$ 14.6 GPa; thus, the peak pressure is clearly above the C-J pressure. Note that the ∼15 GPa C-J pressure for the 50/50 mixture is similar to the 16 GPa C-J pressure of pure TNM. In contrast, the C-J pressure for the 76/24 TNM/NB mixture is 21 GPa. The C-J pressure is expected to be maximum for a stoichiometric mixture and lower for other compositions. This is in line with these results. For the experiment shown here the peak pressure of ∼ 34 GPa is clearly above the C-J pressure. It is also above the von Neumann spike pressure of ∼26 GPa. Within 15 ns after the peak, the pressure decreases to near the C-J pressure. Following the C-J point, the velocity continues to decrease smoothly in a Taylor unloading wave. (b) 3–5 ns rise, shock, then smooth

kind of structure is interrupted determine the type of wave profile observed.

An explanation of the structures in Fig. 6 is as follows. In the lower part, a planar detonation wave has failed and a planar shock is propagating into the liquid explosive. As the shock front progresses, the thickness of the layer between the detonation products and the shock front increases. In the center picture, a thermal explosion at a point initiates a spherically divergent detonation wave. Because it is traveling through precompressed HE, this is a superdetonation wave. The top part of the picture shows a time just after the spherical superdetonation has reached the shock front. In the central region the detonation is again failing. Notice that the structure is quite flat in this region. Near the upper corners, there is an oblique detonation wave. At the sides, there is a superdetonation wave traveling sideways into the shock-compressed HE. Again, the interface velocity recorded by the Fabry-Perot would depend on which type of structure was striking the LiF window and on which part of the structure is registered by the laser beam.

decrease: This type of profile showed a ramp wave increase in velocity during the first 3–4 ns. Interface velocity at the end of this segment was 0.45–0.60 km/s. Following this, a shock takes the interface velocity up to 2.1 km/s. We believe this corresponds to the von Neumann spike; states in the explosive are calculated to be $P_N = 26$ GPa, $U_N = 2.85$ km/s. The increase in velocity during the first 3–4 ns represents an induction period before reaction starts. (c) Interrupted recording: The main characteristic of this wave profile is a particle velocity that rises over a period of about 20 ns to a maximum value of ~2.6 km/s. At this point, the recording is abruptly terminated. Similar records were obtained in which the velocity at interruption ranging from 0.5 to 2.6 km/s. The interface velocity of 2.6 km/s corresponds to a pressure of 34 GPa in the liquid explosive. This is clearly above both the C-J pressure and the von Neumann spike pressure.

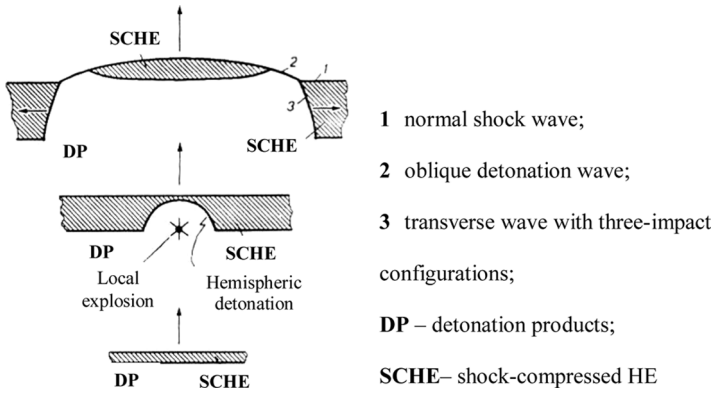

Figure 6. Qualitative picture of pulsating detonation and appearance of local explosions at the interface between overcompressed wave and shock-compressed HE [7]. 1, Electric detonator; 2, cylindrical cell; 3, plasticized PETN; 4, liquid explosive mixture; 5, aluminum coating, 0.5–1.5 (m thick; 6, LiF single crystal; 7, laser beam, $\gamma = 694.3$ nm; 8, focusing lens.

The very high particle velocities observed, well above CJ particle velocity and even the von Neumann spike velocity, we take as evidence of superdetonations. We know of no other way in which a pressure of 49 GPa in the 74/26 mixture or 34 GPa in the 50/50 mixture could arise spontaneously. The scheme shown in Fig. 6 shows one way in which superdetonations could be produced.

We believe that the 3–5 ns of slowly ramping velocity prior to the shock (Figs. 4b and 5b) is evidence of an induction time or delay before energy release begins. Such induction times are common with thermal explosions; induction times would be longer and energy release slower in tetranitromethane diluted with nitrobenzene. (Reviewer's note: ramping wave profiles could also be explained by a domed wave, such as Fig. 6 top. In this case, the laser beam would need to be off the center of the dome. High sound velocities in the LiF window would cause interface motion prior to arrival of the main shock.)

It is well known [6] that pulsating unstable detonation is typical for HE with a slow velocity of energy release. Such

explosives include all gaseous mixtures and some liquid explosives. Reaction breakdown and re-ignition in the shock-compressed HE take place in randomly located points. An initially one-dimensional shock front soon takes on a complex three-dimensional structure. It was demonstrated in the present article that the phenomenon of unstable, pulsating detonation in a homogeneous liquid explosive is not limited to weak explosives but can also occur in a powerful explosive, such as the 74/26 TNM/NB mixture. It is not known whether this behavior is due simply to slower reaction rates or is inherent in all liquid explosives consisting of fuel/oxidizer mixtures.

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