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Investigation of Detonation Wave in Tetranitromethane, Nitromethane, and Their Solutions with Methanol

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The experimental investigation of reaction zone structure was conducted for steady-state detonation in tetranitromethane (TNM), nitromethane (NM), TNM/methanol (TNM/M), and NM/methanol (NM/M) solutions. A VISAR laser interferometer was used. As the result of measurements, particle velocity profiles with clearly defined Von Neumann spikes were determined. For TNM and NM reaction time, parameters in Von Neumann spike and in Chapman-Jouguet point were determined.

Keywords: detonation, liquid HE, methanol, nitromethane, tetranitromethane

Introduction

The chemical reaction in shock-condensed homogeneous high explosive HE has thermal character, and initial rate of decomposition is a strong function of activation energy, which can change for liquid HE in a wide range by means of inert solvent

Address correspondence to A. V. Utkin, Institute of Problem of Chemical Physics, Acad. Semenov Avenue, 1, Chernogolovka, Moscow 142432, Russia. E-mail: utkin@icp.ac.ru addition to HE. Because the initial rate essentially affects detonation wave structure, stability, and limits of propagation, the experimental definition of the initial rate value gives important information for the prediction of liquid HE detonation. In this work the experimental investigation of reaction zone structure for steady-state detonation in tetranitromethane (TNM), nitromethane (NM), TNM/methanol (TNM/M), and NM/ methanol (NM/M) solutions was conducted.

Experimental Scheme

The scheme of experiments is shown in Fig. 1. An HE charge was placed into the polyethylene shell with an internal diameter of 36 mm and 2 mm thick wall (4). The length of the charge was 150 mm. Detonation in the samples was initiated by a shock wave with $\sim 15 \,\text{GPa}$ amplitude, created by the HE plane generator (1). Wave profiles were registered by the VISAR laser interferometer with 2 ns time resolution and $\pm 10 \,\mathrm{m/s}$ accuracy of velocity determination. A laser beam was reflected from a 7to 400- μ m aluminum foil (3) placed between the charge and the transparent window (2). As the result of the experiment, we have the velocity of the foil-window boundary, which represents all details of the reaction zone structure in the detonation wave. To determine the detonation velocity, an ionization gauge (5) was used. It was the first defined point. The second time point was detected by the VISAR signal when the detonation wave reached the boundary with a water window.



Figure 1. Scheme of the experiments.

TNM

TNM with $1.64 \,\mathrm{g/cm^3}$ initial density and $6.4 \,\mathrm{km/s}$ detonation velocity was used in the experiments [1,2]. Typical velocity profiles of aluminum foil-water boundary are presented in Fig. 2. The thicknesses of Al foils in micrometers are designated in the figure by numbers. After the shock jump the velocity decreases. Duration and amplitude of the spike are determined by a Von Neumann spike in TNM. The subsequent velocity increase is caused by circulation of compression and rarefaction waves in Al foil. The attenuation of the velocity spike is recorded during its propagation through the thick foils (100– $400\,\mu\mathrm{m}$). Therefore, to find the exact value of the spike amplitude it is necessary to use thin foils. TNM has a stable detonation front [1], and good registration of particle velocity is observed when the thickness of Al foil decreases to $7 \,\mu m$. It follows from the experiments that the velocity profile for 7- μ m Al foil is almost the same as for 25- μ m Al foil (Fig. 2). This means that the pressure becomes even along the foil thickness during the time of the first circulation of the shock wave inside the Al foil, and in experiments with 7-µm Al foil velocity



Figure 2. Velocity profiles of the aluminum foil–water window boundary for TNM at different thicknesses $(7-400 \,\mu\text{m})$ of Al foil.

profiles describe the state of the HE–water window boundary. The parameters of the Von Neumann spike were found by analysis of shock wave interactions in the pressure P–particle velocity u plane: $u_N = 2.86 \pm 0.06$ km/s, $P_N = 29.9 \pm 0.6$ GPa.

Chapman-Jouguet parameters were also determined from these experiments. The transition from the reaction zone to unloading wave is smooth, and it does not allow defining the position of Chapman-Jouguet point precisely. In Fig. 2 the discrepancy of velocity profiles is observed when the time exceeds 250 ns. Such behavior is usually produced by rarefaction waves. This means that the approximate value of response time is 250 ns. The calculated pressure P_{CJ} and velocity u_{CJ} in the Chapman-Jouguet point are 17.7 ± 0.3 GPa and $1.70 \pm$ 0.03 km/s, respectively. These values are in a good agreement with known data [1,3]. The P_N/P_{CJ} ratio is equal to 1.7 ± 0.1 ; that is, the Von Neumann spike has a great amplitude, which is not typical for liquid HE.

The maximum gradient of particle velocity is observed after the shock jump, and during 50 ns the amplitude of the Von Neumann spike (defined as difference $u_N - u_{CJ}$) decreases twice. If we suppose that about 50% of HE reacts during this time, the estimation of initial reaction rate can be received, which exceeds the value of ~10⁷ 1/s that is typical for powerful HE. Thus, although TNM has low Chapman-Jouguet parameters and large duration of the reaction zone, the high initial rate of decomposition provides existence of a steady-state detonation front in it.

NM

NM with the initial density of 1.14 g/cm^3 and detonation velocity of 6.3 km/s was used in experiments [1]. An example of measured velocity profiles is shown in Fig. 3. Two experiments, which are presented in Fig. 3, do not differ one from another, and a good reproducibility of measurements is observed when 7-µm Al foil is used. On obtained profiles, the velocity peak is registered distinctly in the reaction zone, which is smoothly changed by the rarefaction wave. The sharp velocity decrease takes place during the first 25 ns. As for TNM, it is impossible



Figure 3. Velocity profiles of the aluminum foil–water window boundary for NM in two of the same experiments (7- μ m foil thickness).

to define the position of Chapman-Jouguet point precisely; it is in 25- to 75-ns intervals. Chapman-Jouguet particle velocity and pressure are equal to 1.80 ± 0.05 km/s and 13.0 ± 0.4 GPa, respectively. The problem of C-J point location was noted by researchers who investigated NM. For example, the authors [4] estimated reaction time at 6 ns, whereas it was found in Sheffield et al. [5] that the maximum decrease of particle velocity really was observed during the first 10 ns, but total reaction time (C-J point) was 50 ns.

Parameters of Von Neumann spike were found as for TNM by analysis of shock waves interactions in the *P*-*u* plane: $u_N = 2.60 \pm 0.15 \text{ km/s}, P_N = 18.7 \pm 0.6 \text{ GPa}$. This value is in accordance with the results in Sheffield et al. [5].

It was determined in a previous investigation [1] that the detonation front in NM was not a plane; rather, it had a three-dimensional cellular structure. Nevertheless, in our experiments the reliable recording of particle velocity is produced over a long time ($\sim 1 \,\mu$ s), and reproducibility of experimental results is good enough (Fig. 3). This means that the

instability is feebly marked and the amplitude of small perturbations of the detonation front is significantly smaller than $7 \,\mu m$. Otherwise, the aluminum foil deformation caused a sharp decrease of the intensity of the reflected laser beam and it would be impossible to get any information about the motion of the HE–water window boundary. In addition, the detonation front oscillations are averaged over focal spot with ~100 μm diameter from which the experimental information is recorded. The amplitude of these small perturbations can be comparable with shock wave thickness. In this case the cellular structure is connected with the phenomena inside the shock front rather than inside the reaction zone, as it is assumed when the detonation wave instability is analyzed.

TNM/Methanol, NM/Methanol

The TNM/M solutions were prepared directly before the experiments. Methanol is not an inert solvent for TNM, and in TNM/M solutions tetranitromethane is considered as an oxidant and methanol as a fuel. Detonation properties of this solution do not change steadily with the increase of methanol concentration. In many similar systems the shift of detonation velocity maximum to negative oxygen balance is observed [2]. In Fig. 4 calculation dependence of D on methanol concentration for TNM/M is shown (line 1), which was calculated by scheme [6]. According to this scheme, the detonation velocity is estimated from molecular formula, heat formation, and initial density of HE. Experimental values received in the present work are marked with black spots. Detonation parameters reach a maximum when the concentration of methanol is about 20% and decrease at subsequent increase of solvent concentration. We are interested in the transition from a stable to an unstable detonation front, which is observed at high concentration of methanol.

Experiments were conducted with TNM/M 65/35 $(\rho_0 = 1.207 \text{ g/cm}^3)$ and 50/50 $(\rho_0 = 1.071 \text{ g/cm}^3)$ solutions; that is, at the concentration of methanol, which considerably exceeds the stoichiometric one. Measured rate profiles are



Figure 4. Dependence of detonation velocity on methanol concentration for TNM/methanol.

shown in Figs. 5 and 6. The qualitative difference in results for TNM and TNM/M solution 65/35 becomes the most strongly apparent in Von Neumann spike amplitude, which is nearly



Figure 5. Comparison of velocity profiles for TNM and TNM/ methanol 65/35. Profile of TNM/M is lowered by 250 m/s.



Figure 6. Velocity profiles for TNM and TNM/methanol 50/50.

half the amount in solution than in a pure TNM (profile of TNM/M in Fig. 5 is lowered by 250 m/s for clear comparison with TNM velocity). This leads in particular to approach the rate gradient in reaction zone to the value in unloading wave, and the problem of detection of the Chapman-Jouguet point becomes still more difficult than in TNM. The decrease of particle velocity gradient behind the shock wave front for TNM/M solution can be caused by an HE reaction directly in the shock wave front. An assumption about the reaction in the front for homogeneous HE is grounded on the work by Dremin [7] and explains the sharp decrease of Von Neumann spike amplitude and the low reaction rate behind the front.

The experimental results for TNM/M solution 50/50, which are shown in Fig. 6, significantly differ from the data, which were given above: the velocity profiles oscillate, which is evidence of detonation front instability. Velocity oscillations do not have a distinctly marked period; nevertheless, typical oscillations time is approximately 50-100 ns, which corresponds to the spatial heterogeneity size of about $50 \,\mu\text{m}$.



Figure 7. Velocity profiles for NM/methanol 95/5 and 90/10.

NM/M solutions preparation was analogous to TNM/M. As distinct from TNM, the methanol addition gradually decreases detonation parameters of NM/M solutions. At 5% methanol content, the velocity profile (Fig. 7) is practically the same as the one for NM (Fig. 3). Only weakly marked high-frequency oscillations appear. At methanol concentration increase up to 10% oscillation amplitude rises (Fig. 7). In solutions with 20% methanol, velocity disturbances are so considerable that the intensity of the reflected laser beam decreases to zero at the moment of shock wave coming on the Al foil–water window boundary. This means that the amplitude of oscillations is about 10 μ m.

Conclusions

The received results show that interferometer method is efficient for the study of hydrodynamic instability under shock wave loading of a chemically active medium. Flow instability is registered even if the disturbance amplitude is a value of about a micrometer.

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