Instability of detonation waves in FEFO–methanol mixtures

A V Utkin$^{1,2}$, V M Mochalova$^{1,2}$, S I Torunov$^1$ and S A Koldunov$^1$

$^1$ Institute of Problems of Chemical Physics of the Russian Academy of Sciences, Academician Semenov Avenue 1, Chernogolovka, Moscow Region 142432, Russia
$^2$ Tomsk State University, Lenina Avenue 36, Tomsk 634050, Russia

E-mail: roxete20000@hotmail.com

Abstract. The stability of one-dimensional flow, which occurs at the absence of influence of the boundaries, and instability at the edge of the charge has been investigated in liquid high explosive bis-(2-fluoro-2,2-dinitro-ethyl)-formal (FEFO) and in the mixtures of FEFO with methanol. The structure of the detonation wave was recorded by a VISAR interferometer and by high-speed streak camera. For FEFO it was found strong velocity oscillations in chemical reaction zone but the boundary at the edge of the charge was smooth. When the methanol concentration changes in the interval 10–20%, the stabilization of detonation front is observed and failure reaction waves are occurred at the edge of the charge. Further increase of the concentration of the inert diluent results in the instability of the detonation front at existing of the reaction failure waves. Obtained results show, therefore, that the failure reaction waves and stability of one-dimensional detonation front appear, in general, independently.

1. Introduction

In liquid high explosive (HE) detonation waves propagate both in steady and unsteady pulsating regimes [1–3]. At that it should be distinguished the loss of stability of one-dimensional flow [4,5] which occurs at the absence of influence of the boundaries, and instability at the edge of the charge (wave of reaction failure) [6]. Both types of instabilities are caused by the same factors: the mutual influence of the flow dynamics and the chemical reaction rate. Therefore, it may seem naturally that either the detonation front should be stable or the detonation wave instability and the wave of reaction failure should occur simultaneously. Numerous studies of various liquid HE would seem to confirm this fact. It is equally obvious, however, that the flows both at the center and at the edge of the charge are substantially different and the conditions of loss of stability are not required to be satisfied along the entire front. The purpose of this work is to prove that, in practice, the relationship between these two types of instability is not strictly determined. Bis-(2-fluoro-2,2-dinitro-ethyl)-formal (FEFO, $C_5H_6N_4O_{10}F_2$) and the mixtures of FEFO with methanol were investigated in this work. The structure of the detonation wave was recorded by a VISAR interferometer and by a high-speed streak camera.

2. Experimental scheme

Liquid high explosive FEFO with the initial density of 1.60 g/cm$^3$ [7–10] was investigated. Methanol was used as inert diluent (the initial density was equal to 0.79 g/cm$^3$). The mixtures of FEFO–methanol were prepared immediately before the experiment. The velocity profiles
Figure 1. The scheme of the experiment with VISAR: 1—initiating explosive charge; 2—water window; 3—Al foil; 4—shell; 5—ionization gauge (a). The scheme of the experiment with a high-speed streak camera for recording of the glow of the detonation front: 1—initiating explosive charge; 2—PMMA plate; 3—PMMA tube; 4—FEFO—methanol; 5—mirror (b).

Inside the reaction zone were recorded by a laser interferometer VISAR with a time resolution of about 2 ns. The dependence of the velocity on the time gives information about the instability of detonation wave. The scheme of the experiments is shown in figure 1a. A high explosive charge was placed in a shell 4 with an internal diameter of 26–50 mm. The shells were made from different materials, the compressibility of which was like higher (polypropylene, PMMA) and less (steel, brass) than compressibility of FEFO. The variation of the diameter and material of the shell made it possible to eliminate the influence of boundaries on the flow in the central part of the charge. The length of the charge was 150 mm. Detonation was initiated by pressed charges of RDX or TNT 1, in which a plane detonation front was produced by an explosive lens. The probing laser beam was reflected from 7 \( \mu \)m aluminum foil 3 placed between the end of the charge and a water window 2. In the experiment we recorded the velocity of motion of the foil/water window boundary. Compressibility of water higher than the compressibility of FEFO hence a rarefaction wave was reflected in the products of detonation. I.e. in the experiments in fact the information was received about the change of state of detonation products in the reflected rarefaction wave. This fact was taken into account in interpretation of experimental data. The interferometer constant is 305 m/s, which allows determining the particle velocity with an error of \( \pm 10 \) m/s. In each experiment simultaneously with the particle velocity the detonation velocity \( D \) was measured using the ionization gauge 5. The second time mark was the interferometer signal recording the arrival of the detonation wave at the interface with the window. The error of the detonation velocity determination was no more than 1%.

Investigation of stability of non-one-dimensional flow arising in the detonation wave at the edge of the charge and observation of waves of reaction failure were carried out by high-speed streak camera (figure 1b). Liquid high explosive 4 was placed in a PMMA tube 3 with an internal diameter of 36 mm and a wall thickness of 2 mm. The length of the charge was 150 mm. Detonation was initiated by pressed charges of RDX/TNT 50/50 with diameter of 40 mm 1. A glow of the detonation front as it propagated through the charge was recorded by streak camera. The view of the boundary of liquid high explosive depends on the absence or presence of waves of reaction failure: in the first case, the boundary is smooth, and in the second case it is zigzag shaped [6].
3. Experimental results
Particle velocity profiles for FEFO and FEFO–methanol solution measured by VISAR are given in figures 2—3. Mass concentration of methanol, material and diameter of the shell are indicated in the figures legends. It was found earlier [9, 10] that instability of detonation wave in FEFO was observed. For FEFO the instability is revealed on the dependence of the velocity on the time as strong velocity oscillations with character amplitude of 50 m/s in both the chemical reaction zone and the unloading wave. A distinct oscillations period is absent, but the order of magnitude is approximately 50 ns. This means that the detonation front is unstable and the size of inhomogeneities is comparable with the foil thickness, i.e. of the order of 10 µm. Furthermore, the instability pattern is not related to the influence of the charge boundary, which is confirmed by the unchanged qualitative shape of the velocity profiles at the change of the diameter and material of shells.

Dilution of FEFO by methanol reduces detonation parameters of mixtures, so one would expect greater development of the instability of detonation front. However, there is a much more complex nature of the influence of methanol on the detonation instability. The flow remains almost un-changed if the methanol concentration less than 10%: the nature of the oscillations, their frequency and amplitude are the same as in a neat FEFO (figure 2). Further increase of methanol concentration in the range of 10–15% instead of development of the instability results in unexpected stabilization of the detonation front, which is manifested in the disappearance of the oscillations on the velocity profiles (figure 3). And only when the concentration of methanol is 20% or more, the flow becomes unstable again. Moreover, the velocity oscillations are so high that one can only speak about reproducibility of the averaged detonation parameters. In particular, the detonation velocity for a specific composition of the mixture remains constant. This regime of propagation of detonation waves is remained up to 32% of methanol, which corresponds to a maximum concentration. When it is exceeded, the mixture does not detonate and on the dependence of the detonation velocity $D$ on the concentration of methanol a sharp drop of the measured velocity of decay detonation is observed with respect to

![Figure 2. Velocity profiles for unstable detonation waves: 1—0%, Al, internal diameter of 36 mm; 2—2%, PMMA, internal diameter of 36 mm; 3—8%, PMMA, internal diameter of 36 mm.](image)
Figure 3. Velocity profiles for high concentrations of methanol: 1—10%, Cu, internal diameter of 26 mm; 2—12%, Cu, internal diameter of 26 mm; 3—15%, Cu, internal diameter of 26 mm; 4—20%, steel, internal diameter of 50 mm.

Figure 4. The dependence of detonation velocity on methanol concentration for mixture of FEFO–methanol.

A linear approximation of the experimental data:

\[ D = 7.49 - 0.057C, \quad (1) \]

where \( D \) is measured in km/s, and concentration in mass % (figure 4). This is clearly seen when \( C = 35\% \) of methanol (the bright spot in figure 4).

The stability of one-dimensional detonation in a mixture of FEFO–methanol was investigated up to now. For record of the flow on the edge of the charge the high-speed camera was used (figure 1b). The streak images of light of the detonation wave front are shown in figures 5. Since the detonation front in the neat FEFO is unstable, then one would expect the formation of wave of reaction failure at the edge of the charge. However, contrary to received opinion, the
Figure 5. The streak images of light of the detonation wave front in FEFO–methanol. Vertical arrows indicate the time of detonation initiation and the time of the wave exit on the free end of charge, respectively. Concentration of methanol is equal to 0 (a), 10% (b), 12% (c), 15% (d).

lateral boundary of propagation of the detonation wave is smooth (figure 5a). Thus, FEFO is an example of a liquid high explosive with a pronounced cellular structure of the detonation front in which waves of reaction failure at the edge of the charge are absent. Possible reasons for this unusual flow pattern will be considered at discussion of the results.

The FEFO dilution by methanol does not result in waves of reaction failure, if the concentration of the diluent is less than 10%. The first signs of instability appear in a mixture of FEFO–methanol 90/10 (figure 5b). When the concentration of methanol is 12% the existence of reaction failure waves is becoming more pronounced (figure 5c). When the amount of the diluent reaches 15%, the reaction failure waves are propagated to the center of the charge, what results in the attenuation of the detonation. This means that the diameter of charge used in the experiments (36 mm) is critical for mixture 85/15. The streak image of light of the detonation front for this situation is shown in figure 5d, it is typical for liquid high explosives which critical diameter is determined by the waves of reaction failure [6].

4. Discussion of results
The results show, therefore, that FEFO dilution by methanol allows realization of different combinations of one-dimensional instability (pulsations of the detonation front) and instability at the edge of the charge (the waves of the reaction failure). When the concentration of methanol less than 10% the detonation front is unstable, but the waves of the reaction failure are absent. In the range of 10–15% the front is stabilized, but there are the waves of reaction failure. Further increase of methanol concentration results in a complete loss of detonation stability: pulsations of the detonation front and the waves of reaction failure are observed. If we add a well-known fact of the existence of absolutely stable detonation in liquid high explosives [6] to these results, we will come to the conclusion about the absence of a rigidly deterministic connection between the conditions of coexistence of considered two types of instability.

The most unexpected result is a flow without the waves of the failure of reaction in the presence of the pulsation structure of the detonation front, which is observed in FEFO. This phenomenon can be explained if we assume that HE can react directly in the front of the compression wave. In the neat FEFO the degree of decomposition can be high because of the high initial rate of reaction. This results in a decrease of the reaction rate behind the shock front, which is manifested in a relatively slow decrease of the velocity in the reaction zone. As a result the formal stability criteria for one-dimensional detonation [4] are violated. The addition of methanol decreases the initial reaction rate and hence the amount of HE, which reacts at the front. The consequence of this, the fast stage of the reaction moves into the reaction zone and stabilization of detonation waves is observed.
Decrease of the pressure and temperature at the approaching to the end of the charge results, in this case, in the same effect as dilution of FEFO with an inert diluent: the fraction of the decomposed HE in the shock-wave front decreases but the chemical reaction rate behind the shock jump may even increase. As a result, the conditions required for the formation of the waves of reaction failure are not established. Note also that the criteria for the detonation front instability in the form of cellular structure cannot be used in this case as they were obtained without considering the reaction in front [4, 5].

Obtained results show, therefore, that the waves of reaction failure and stability of one-dimensional detonation front appear, in general, independently.

Acknowledgments
This work was supported by Russian Foundation for Basic Research (project No. 15-03-07830).

References
[1] Schelkin K I and Troshin Ya K 1963 Gas Dynamics of Combustion (Moscow: Izd. Akad. Nauk SSSR)
[2] Voitsekhovskii B, Mitrofanov V and Topchiyan M 1963 Structure of the Detonation Front in Gases (Novosibirsk: Izd. Sib. Otd. Akad. Nauk SSSR)
[3] Zverev I and Smirnov N 1987 Gas Dynamics of Combustion (Moscow: Izd. Mosk. Univ.)
[4] Zaydel R 1961 Dokl. Akad. Nauk SSSR 136 1142–1145
[5] Aslanov S K, Budzirovskiy V N and Schelkin K I 1968 Dokl. Akad. Nauk SSSR 182 53–55
[6] Dremin A, Savrov S, Trofimov V and Shvedov K 1970 Detonation Waves in Condensed Matter (Moscow: Nauka)
[7] Zhukov B P 1999 Energetic Condensed Systems: A Short Encyclopedic Dictionary (Moscow: Yanus-K)
[8] Finger M, Lee E, Helm H, Hayes B, Hornig H, McGuire R, Kahara M and Guidry M 1976 Proc. Sixth Symp. (Int.) on Detonation 710–722
[9] Torunov S I, Utkin A V, Mochalova V M and Garanin V A 2010 Combust., Explos. Shock Waves 46 599–603
[10] Utkin A V, Mochalova V M, Torunov S I and Koldunov S A 2015 Combust., Explos. Shock Waves 51 476–481