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SELF-SUSTAINING DETONATION IN LIQUIDS WITH BUBBLES OF EXPLOSIVE GAS

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Compression waves in liquids with gas bubbles cause the most significant change in such basic parameters of the medium as bubble size and volumetric gas content β_0. When a bubble is compressed by a shock wave, the temperature of the gas in the bubble increases and may be high enough for ignition of the reactants. Compression ignition of liquid explosives when cavitation bubbles are collapsed near the walls of a tube has been widely studied and described, for example, in [1, 2]. It was experimentally confirmed in [3, 4] that it is possible for a single bubble of an explosive gas to undergo shock ignition in a nonreactive liquid. The ignition of systems of the oxidant bubble — liquid fuel type was observed in [5, 6].

In [7], the term "bubble detonation" was used for the process of the shock-wave ignition of a chain of vertical bubbles of explosive gas in a liquid over a length of 0.7 m. It is premature to introduce such a term since it has not been shown that an ignition wave can propagate independently via bubbles over a greater length without additional action by the initiating shock wave. Also, it follows from [8] that the process observed in this case was not self-sustaining — the wave attenuated as it propagated (pressure and velocity decreases). There has been almost no study of shock-wave phenomena and ignition conditions in reactive gas—liquid systems with gas bubbles uniformly distributed in the volume (systems of the nonreactive liquid—explosive gas and liquid fuel—gaseous oxidant types). The features of processes accompanying shock-wave motion have been explained only recently [5, 6]. The same is true of the experimental observation of a self-sustaining detonation wave [5, 9].

It is most worth noting that the amount of heat given off per unit mass in these systems as a result of the reaction is several orders smaller than, for example, in liquid and gaseous explosives, i.e., the Hugoniot and detonation curves nearly coincide. In light of this, the existence of a detonation wave in a bubble system [5, 9] is an important matter.

Here we have the goal of explaining the properties and conditions of excitation of detonation in relation to the value of β_0 and the physicochemical composition of the bubble medium.

The experiments were conducted on the unit shown in Fig. 1. The vertical shock tube, 35 mm in diameter, consisted of an initiation section 1 separated from a working section 2 by a breakaway membrane 3, two optical sections 4 and 5 with 8 × 240 mm organic glass windows, and a bubble generator 6. Piezoelectric transducers 7-13 were placed flush against the inside surface of the tube. The natural frequency of the transducers was about 300 kHz, while the diameter of the piezoelectric ceramic was 2 mm. The optical input 14 of a photomultiplier (FÉU-31) was located opposite either transducer 12 or 11.

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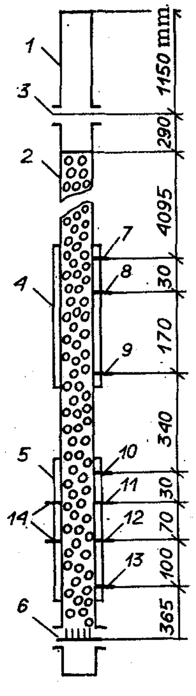

Fig. 1

Section 2 was filled with the liquid to the height h. The value of β_0 was determined from the rise of the liquid column by Δh to the control level $L_0 = h + \Delta h = 5.2$ m, so that $\beta_0 = \Delta h / L_0$. The tests were conducted with (series A) and without (series B) a membrane. A shock wave was generated in the system by the ignition of a mixture of $C_2H_2 + 2.5\ O_2$ with an initial pressure p_{0g} in Sec. 1. The intensity of the shock wave was varied by changing p_{0g} ($p_{0g} \leqslant 0.4$ MPa). The initial pressure behind the membrane p_0 was usually equal to atmospheric pressure in the series A tests and equal to p_{0g} in the series B tests.

The experiment entailed recording of the profiles of pressure and the velocity of the waves with four piezoelectric transducers and profiles of luminescence with the photomultiplier. The wave process of light generation was photographed with a drum-type photodetector. The signals from the pressure transducers and photomultiplier were recorded by two OK-33 oscillographs. The transducers were connected to the inputs of the oscillographs through a source follower which ensured a time constant greater than 1 sec.

We studied the following systems consisting of a liquid and bubbles of an explosive gas: 1) $L_{\alpha\gamma}$ — ($C_2H_2 + 2.5O_2$), $\gamma = 0, 0.15, 0.25, 0.5$; 2) $L_{\alpha\gamma}$ — ($2H_2 + O_2$), $\gamma = 0, 0.15, 0.5$; 3) 3) $L_{\alpha\gamma}$ — ($C_3H_8 + C_4H_{10} + nO_2$), n = 6, 12; $\gamma = 0, 0.5, 0.75$; we also studied the following nonreactive bubble systems: $L_{\alpha\gamma} - N_2$, $L_{\alpha\gamma}$ — ($2H_2 + N_2$), $L_{\alpha\gamma}$ — ($C_3H_8 + C_4H_{10} + nN_2$) with n = 6, 12; $\gamma = 0$, where $L_{\alpha\gamma} \to \alpha H_2O + \gamma(CH_2OH)_2CHOH$ was the liquid; $\alpha + \gamma = 1$ (α and γ are the volumetric concentrations of water and glycerine). The tests were conducted with bubbles of the size d = 2.5-4 mm, the exact size depending on p_0. In the tests, $0.5\% \lesssim \beta_0 \lesssim 10\%$ (in the determination of β_0, the relative error $\Delta\beta_0/\beta_0 \lesssim \pm 10\%$).

Figure 2 shows typical oscillograms illustrating the propagation in the bubble systems of shock waves V_2 and short solitary waves V_1 formed with the reaction (V_3 represents the wave reflected from the end). The upper rays represent pressure, while the lower rays represent luminosity (a, b) and pressure (c) in the waves. The period of the sinusoids is 100 µsec. Then the intensity of the shock wave is close to the critical value with a pressure p_1^*, the bubbles in the front of the wave are ignited (Fig. 2a, where $\beta_0 = 2\%$, $d \simeq 3.5$-4 mm, $p_0 = 0.1$ MPa, $p_{0g} = 0.25$ MPa, series A test, system 1, $\gamma = 0$). The value of p_1^* depends on β_0 and the physicochemical properties of the bubble medium, and it turns out to be close to the combustion pressure in a closed volume of a fuel mixture with an initial pressure p_{0g}^*. For example, with system 1 ($\gamma = 0$) when $\beta_0 \simeq 2\%$ $p_{0g}^* \simeq 0.25$ MPa ($p_1^* \simeq 2$-3 MPa), with increasing β_0 to 6% $p_{0g}^* \cong 0.35$ MPa ($p_1^* \simeq 4$-5 MPa).

Shock waves of an intensity below the critical value do not cause the resisting media to give off light. Nor was any luminescence seen ($p_{0g} \lesssim 0.4$ MPa) in any of the above nonreactive bubble systems or in the systems $L_{\alpha\gamma} - O_2$ ($\gamma = 0.25$ and 0.75). These results are

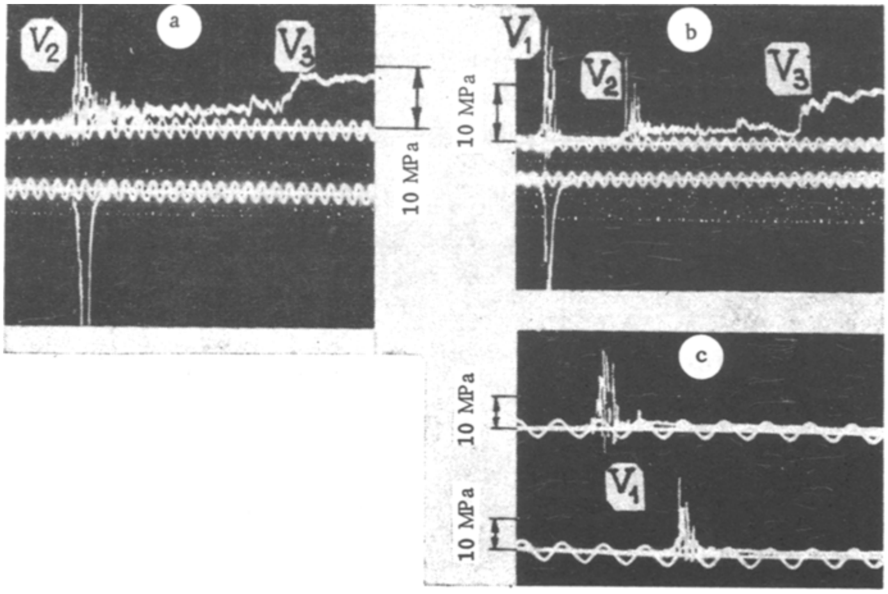

Fig. 2

evidence that light is given off as a result of the chemical reaction of the explosive gas rather than from the increase in temperature during compression of the bubbles by the shock wave or the reaction of the liquid $L_{\alpha\gamma}$ with oxygen. Under the conditions in our tests, the liquid $L_{\alpha\gamma}$ ($\gamma \leqslant 0.75$) can be considered nonreactive, i.e., it has no chemical effect on the initial stage of ignition of the bubbles.

It is apparent on the oscillograms that the pressure jump in the shock waves with ignition (see Fig. 2a) is preceded by a smooth increase in pressure (a precursor). Pulsations with a period of 150-300 μsec develop in the front and pressure attains an almost constant level. The pressure pulsations are the result of oscillations of gas bubbles; if the bubbles ignite, the amplitude of the pulsations increases.

Figure 2b, c shows later stages of evolution of a shock wave igniting bubbles. A short solitary wave V_1 of duration 50-150 μsec is formed under different conditions for systems 1-3 in the series A and B tests. The location of sections 4 and 5 by transducers 7-13 in the upper regions of the tube shows that the wave V_1 departs from the shock wave V_2 at a distance $L \simeq 1$-3.5 m from the control level of the bubble medium; the value of L decreases with an increase in p_{0g}. For example, with $\beta_0 = 2\%$, $p_{0g} = 0.3$ MPa, and $p_0 = 0.1$ MPa in system 1 ($\gamma = 0$), $L \simeq 3$ m, and the velocity of the wave before the moment of separation is about 400 m/sec; after the separation of wave V_1, it accelerates to the velocity $v_1 \simeq 700$-800 m/sec and moves farther away from the shock wave V_2 traveling behind it at a slower speed $v_2 \simeq 400$-450 m/sec.

The bubbles generate light only in wave V_1. There is no luminescence (chemical reaction) in the shock wave V_2. Characteristic oscillograms of pressure and luminosity are shown in Fig. 2b, where the process is occurring in system 1 ($\gamma = 0$) at $\beta_0 = 2\%$, $d \simeq 3.5$-4 mm, $p_0 = 0.1$ MPa, and $p_{0g} = 0.35$ MPa ($v_1 \simeq 750$ m/sec, $v_2 = 500$ m/sec). Such a two-wave process is absent from nonreactive system in similar tests, i.e., generation of the solitary wave V_1 is due to energy release in the reacting bubble medium.

Figure 2c shows an example of the case when the wave V_1 moves a significant distance away from shock wave V_2 (here, the wave propagates in system 2 ($\gamma = 0.5$), $\beta_0 = 2\%$, $d \simeq 2.5$-3.0 mm, series B test, $p_0 = p_{0g} = 0.25$ MPa).

Pressure pulsations due to the compression and ignition of bubbles are recorded in waves V_1; the pressure peaks on the tube wall reach 15-25 MPa. Pressure falls behind the wave V_1 and is close to p_0. Ignition of the initial bubbles is evident on the photographs, the duration of the luminescence of each bubble in the wave V_1 being 2-3 μsec.

Figure 3 shows measurements of the velocities v_1 (curves 1 and 2) and v_2 (curves 3 and 4) in relation to p_{0g} (series A, $p_0 = 0.1$ MPa) for system 1 ($\gamma = 0$) with $\beta_0 = 2\%$. Curves 3 and 4 were obtained from measurements in section 4, while curves 1 and 3 were obtained from section 5. It is evident that v_1 reaches a nearly constant value at a fixed distance and ceases to depend on the initiation pressure, i.e., the velocity of the shock wave. Thus, the process

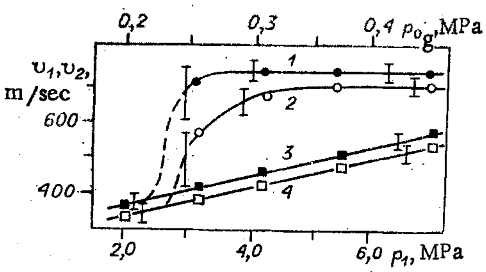

Fig. 3

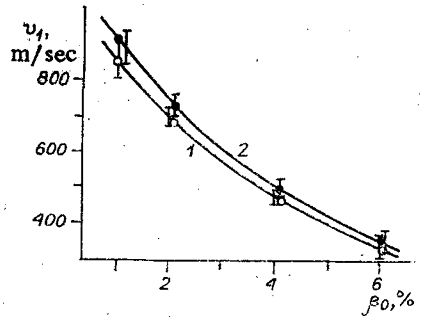

Fig. 4

of the propagation of wave V_1 is self-sustaining. The small difference in the velocity in sections 4 and 5 is due to the natural dependence of the wave velocity on β_0. A decrease in β_0 from top to bottom due to the effect of hydrostatic pressure leads to an increase in the velocity of the compression wave (for the shock wave, $v_2 = (p/\beta_0\rho_0)^{0.5}$, where p is the pressure in the wave and ρ_0 is the density of the gas–liquid medium [10]). The wave V_1 propagates in the medium at supersonic velocity – v_1 is several times greater than the speed of sound $c_0 = (p_0/\beta_0\rho_0)^{0.5}$.

It should be noted that in contrast to the solitary waves formed in nonreactive bubble systems due to their disperse and nonlinear properties (see [11, 12], for example), the solitary wave V_1 is formed only in the presence of energy release in the system, is self-sustaining, and has a more complicated internal structure; propagation of the wave V_1 in the bubble medium leads to irreversible changes in the medium.

Since chemical energy is released in wave V_1 and its steady-state supersonic velocity is independent of the conditions of initiation, such a process can be considered to be a detonation process. We will henceforth refer to solitary wave V_1 as a detonation wave (as in [5, 9]).

The two-wave process seen in our tests (the presence of wave V_2 along with V_1) is due to the use of a shock wave to initiate bubble detonation. This is the character of the oscillograms in test series A and B, i.e., it was not caused by the use of the membrane. Of course, only the detonation wave V_1 will propagate over long mean free paths since the pressure in the wave V_2 decreases with distance and the latter wave attenuates. If we increase p_{0g}, then v_2 may turn out to be equal to or greater than v_1 and supercompresssive detonation will occur – there will be a long wave with a pressure profile close to rectangular and luminescence in the front. However, such a detonation wave is not self-sustaining, and its existence requires a piston action by the medium behind the front.

Figure 4 shows the dependence of the velocity of steady-state detonation on β_0 in system 1 ($\gamma = 0$) at $p_0 = 0.1$ MPa (series A), where curves 1 and 2 show the velocities v_1 in sections 4 and 5, respectively. At $\beta_0 \gtrsim 8\%$ and $p_0 = 0.1$ MPa, the bubble medium does not ignite if $p_{0g} \lesssim 0.4$ MPa, i.e., losses increase with an increase in the concentration of the gas phase, and the initiation of detonation requires an increase in the intensity of the incident shock wave. With a decrease in β_0 to 0.5%, the detonation wave becomes unstable, the process is easily disrupted, and detonation dies out (the wave V_1 becomes diffuse).

Table 1 shows critical pressures p_{0g}^* and values of v_1. The critical pressures are the pressures at which detonation is or is not excited in systems 1-3 with $\beta_0 = 2\%$ and $p_{0g} \lesssim 0.4$ MPa. It is evident from the table that systems 1 have the best capability of being ignited and exciting detonation. The explosive gas mixture in these systems ($C_2H_2 + 2.5O_2$) has the shortest ignition delays τ_i. In every case for systems 1-3, an increase in the viscosity of the liquid with the addition of glycerine leads to a decrease in p_{0g}^*, i.e., facilitates ignition of the bubble systems. Ignition in propane–oxygen gas mixtures (τ_i is maximal in them) requires longer maintenance of high temperatures. These temperatures are reached in the final stages of compression of the bubbles. Longer maintenance of these temperatures is assured by increasing the viscosity of the liquid (in systems 3, ignition and detonation are excited only at $\gamma = 0.75$). An increase in the concentration of glycerine also leads to an increase in the speed of sound in the liquid, which explains the increase in v_1 seen here (see Table 1).

In our opinion, a detonation wave propagates in the above-examined bubble systems by the mechanism of microexplosions: Bubbles compressed in the pressure field explode and send out

TABLE 1

System	γ	Test series	p^*_{0g}, MPa	v_1, m/sec
1	0	A, $p_0 \doteq 0{,}1$ MPa	0,25	750
	0	B	—	—
	0,15	B	0,13	850
	0,25	B	0,1	940
	0,5	B	0,05	1050
2	0	A, $p_0 = 0{,}1$ MPa	0,4	500*
	0	B	—	—
	0,15	B	—	—
	0,5	B	0,23	940
$3(n = 6)$	0	A, $p_0 = 0{,}1$ MPa	—	—
	0	B	—	—
	0,5	A, $p_0 = 0{,}1$ MPa	—	—
	0,5	B	—	—
	0,75	A, $p_0 = 0{,}04$ MPa	$<0{,}3$	940
	0,75	B	$<0{,}37$	1200
$3(n=12)$	0	A, $p_0 = 0{,}1$ MPa	—	—
	0	B	—	—
	0,5	A, $p_0 = 0{,}1$ MPa	—	—
	0,5	B	—	—
	0,5	A, $p_0 = 0{,}075$ MPa	0,34	450 *
	0,75	B	$<0{,}26$	1050

*Ignition was recorded in section 5, but no detonation wave was formed under the given conditions. In the series B tests, a shock wave was excited by the ignition of a $2H_2 + O_2$ mixture in the tube in system 2, while section 1 was filled beforehand with a $C_2H_2 + 2.5O_2$ mixture to the pressure $p_{0g} \simeq p^*_{0g}$ in systems 3.

shock waves into the surrounding liquid. These shock waves in turn compress and ignite bubbles located upstream. A similar mechanism was examined in [2] to describe the excitation of low-velocity detonation in liquid explosives.

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