Dynamics of shock waves in bubble zones of finite size with a hydrate-forming gas

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Abstract. The purpose of this study is to study the dynamics of the wave field, which is realized in a channel with a liquid containing a rectangular zone with bubbles of the freon-12 hydrate-forming gas during the propagation of a pressure shock wave. In the initial state, the considered gas-liquid system is under pressure $P_0$. After a sudden increase in pressure to the value of $P_e$, a pressure wave of a stepped profile propagates in the system and, as a result of the presence of a bubble curtain, its amplitude increases, which in turn has a more favorable effect on the formation of hydrate in gas bubbles. In the initial state, the hydrate formation process was not taken into account. As a result, the dynamics of the pressure wave is shown during its propagation in a semi-infinite channel containing a gas curtain with a hydrate-forming gas. The mechanism of gas hydrate formation is described in this work on the basis of the theory of nonequilibrium phase transitions in vapor-liquid systems.

1. Introduction

Bubble liquid is unique in its acoustic properties. So, the addition of a small amount of gas in the form of bubbles to the liquid leads to anomalous compressibility, therefore, a curtain of a mixture of liquid with gas bubbles can be used as a protective layer for underwater objects from shock waves, for "masking" during sonar, and also as an underwater sound channel [1]. Also, during the passage of impulse signals in a liquid containing a bubble curtain or a cluster of finite sizes, a significant increase in the amplitude of the pressure wave can be observed [2-7].

Currently, gas hydrates are attracting increased attention both as a potentially new source of energy and as a new way of storing large volumes of gas [8-10]. One of the methods for the synthesis of gas hydrate is to obtain it in a bubbly liquid by means of exposure to a shock wave [11, 12]. Another area of application of such effects can be the use of such channels to achieve the necessary conditions for hydrate formation during the propagation of pressure waves of small amplitude. The hydrate formation process is rather slow and needs to be intensified [13-15]. Increasing the amplitude of the compression wave can facilitate this process. This can be achieved in the same way in the case of a change in the volumetric content of gas in a liquid during the propagation of a pressure wave in it.
2. The Basic equations

Let us suppose that in a semi-infinite channel filled with water, there is a rectangular zone with a hydrate-forming gas Freon-12 and a volume concentration of the gas phase $\alpha_0$. Gas bubbles in the cluster area have the same radius $a_0$. The bubble curtain itself is spaced from the plane $z=0$ at some distance $l_{cl}$ (figure 1). At a moment in time $t=0$ at the end of the channel ($z=0$) the pressure jump to the value $P_c$. When the time $t>0$, the pressure wave propagates from left to right. It is known that the speed of wave propagation in a "pure" and bubbly liquid is very different, therefore, a wave in a "pure" liquid will outstrip the wave in the bubble zone, focusing in it and generating a complex wave structure in it, which will depend on the parameters of the bubble region (dimensions bubble cluster, volumetric content and dispersion of bubbles in the cluster). Focusing a wave in a bubble cluster leads to the formation there of a quasi-stationary shock wave propagating along it along the symmetry axis. A numerical study of the formation and propagation of a wave in the bubble zone is carried out within the framework of the equations of the mechanics of multiphase media [16].

![Figure 1. Layout of the bubble curtain: $l_x$ and $l_y$ are the measurements of the bubble zone, $l_{cl}$ is the length from the front wall of the bubble curtain to the plane $z$.](image)

To describe the wave motion of bubble systems, assuming sticking or splitting of bubbles, we write down the linearized equations of masses, the number of bubbles, momenta and pressure in bubbles for the two-dimensional case [2]:

$$
\begin{align*}
\frac{d\rho_i}{dt} + \rho_i \left( \frac{\partial u}{\partial x} + \frac{\partial \vartheta}{\partial y} \right) &= 0 \quad (i = l, g), \\
\frac{dn}{dt} + n \left( \frac{\partial \alpha_l}{\partial x} + \frac{\partial \vartheta}{\partial y} \right) &= 0, \\
\rho \frac{du}{dt} + \frac{\partial p_l}{\partial x} &= 0, \\
\rho \frac{d\vartheta}{dt} + \frac{\partial p_l}{\partial x} &= 0, \\
w &= \frac{da}{dt} \left( \frac{d}{dt} = \frac{\partial u}{\partial t} + u \frac{\partial}{\partial x} + \vartheta \frac{\partial}{\partial y} \right), \\
\alpha_l + \alpha_g &= 1, \quad \alpha_g = \frac{4}{3} \pi a^3 n, \quad \rho_l = \rho_l^0 \alpha_l, \quad \rho = \rho_g + \rho_l
\end{align*}
$$

where $a$ is a bubble radius, $\gamma$ is a gas adiabatic exponent, $p_l$ is the fluid pressure, $\rho_l^0$ are the true phase density, $\alpha_l$ are the volumetric phase contents, $n$ is a number of bubbles per unit volume, $w$ is a radial bubble velocity, $u$ and $\vartheta$ are the velocity components along the $x$ and $y$ axes. The subscripts $i = l, g$ denote the parameters of the liquid and gas phases.
When describing the radial motion in accordance with the refinement proposed in [17], we will assume that \( w = w_R + w_A \), where \( w_R \) is determined from the Rayleigh–Lamb equation, and \( w_A \) is determined from the solution of the problem of spherical unloading on a sphere of radius \( a \) in the carrier fluid in the acoustic approximation:

\[
\frac{a}{\tau} \frac{dw_R}{dt} + \frac{3}{2} \frac{w_R}{a} + 4 \nu_l \frac{w_R}{a} = \frac{p_g - p_l}{\rho_l^2}, \quad w_A = \frac{p_g - p_l}{\rho_l^2 c_l a_g} \tag{2}
\]

where \( \nu_l \) is the fluid viscosity and \( C_l \) is the sound speed in a "clean" liquid.

To describe the change in pressure in gas bubbles, we will use the first law of thermodynamics. Taking into account the equations of conservation of masses (1), as well as the equation of state of the gas \( \varepsilon_g = c_{gv} T_g \) (\( c_{gv} \) is the specific heat capacity at constant volume and \( T_g \) is the average temperature of the gas in the bubble), we can write:

\[
m_g c_{gv} \frac{d p_g}{dt} = -4\pi a^2 (p_g w + j_g c_{gv} (T_g - T_a) + q(ga)). \tag{3}
\]

Here \( q(ga) \) is the heat flow from the phase of gas to the interface, \( T_{ga} \) is the interface temperature, \( p_g \) is the gas pressure, \( m_g \) is the mass of gas in a bubble.

The presented system of equations must be supplemented with expressions describing the kinetics of hydrate formation, the intensity of heat transfer at the interface and the heat effects of the phase transition.

The heat balance condition has the form:

\[-q(ga) + q(al) = j_h l_h, \tag{4}\]

where \( q(al) \) is the heat flux from the bubble surface into the liquid, \( l_h \) is the specific heat of hydrate formation per unit of its mass.

The hydrate formation process is quite complex. It can be accompanied by diffusion, nonequilibrium effects during phase transitions, and also depends on the degree of metastability [11, 18], which affects the heterogeneous centers of nucleation of hydrate crystals. Therefore, by analogy with the theory of nonequilibrium phase transitions in vapor-liquid systems, the intensity of mass transfer between bubbles and liquid can be represented as [16]:

\[ j_g = \frac{\beta (p_g - p_s (T_a))}{\sqrt{2 \pi a^3 \tau_a}}, \quad p_s (T_a) = p_{s0} \exp \left( \frac{T_a - T_0}{T_u} \right), \quad p_{s0} = p_s (T_0) \tag{5}\]

Here \( \beta \) is a unified empirical parameter (reduced accommodation coefficient) responsible for diffusion phenomena, as well as features of the formation and growth of hydrate crystals in a liquid, \( p_s (T_a) \) is is equilibrium pressure of hydrate formation corresponding to temperature \( T_a \).

On the basis of [16], the heat flux \( q(ga) \) is given by the approximate final relation:

\[ q(ga) = \text{Nu} \lambda_g \frac{T_a - T_0}{a}, \quad \text{Nu} = \begin{cases} \sqrt{\text{Pe}}, & \text{Pe} \geq 10^2 \\ 10, & \text{Pe} < 10^2 \end{cases}, \tag{6}\]

\[ \text{Pe} = 12(y - 1) \left( \frac{T_a}{T_g - T_0} \right)^{g(T)}, \]

where \( g(T) \) is the gas thermal diffusivity. We write the intensity of the heat flux from the interface into the liquid as:

\[ q(al) = -\lambda_l \left( \frac{\partial T}{\partial r} \right)_0 = -\lambda_l \left( \frac{a}{a_0} \right)^2 \left( \frac{\partial T}{\partial r} \right)_0. \tag{7}\]
Here $T'_0$ is the temperature distribution in liquid around bubbles, $r$ and $r_0$ are Euler and Lagrangian radial microcoordinates. In order to determine heat fluxes around bubbles in accordance with (7), it is necessary to solve the heat conduction equation written in Lagrangian variables:

$$\rho'_0 c_1 \frac{\partial T'_l}{\partial t} = \frac{1}{r'_0} \frac{\partial}{\partial r_0} \left( r'_0^2 \lambda'_1 \frac{\partial T'_l}{\partial r_0} \right), \quad (a_0 < r_0 < \infty)$$

(8)

by the boundary conditions:

$T'_l = T_a$ when $r_0 = a_0$ and $T'_l = T_0$ when $r_0 \to \infty$.

Theoretical estimates, as well as calculation in relation to the data from [11] for water-freon, allows for the interface to accept the equality between the temperature system and the temperature at the interface and to consider it constant ($T'_a = T_0$).

We will also assume that the liquid is linearly compressible, and the gas is calorically perfect:

$$p_i = p_0 + C_i^2 (\rho'_0 - \rho_0^0), \quad p_g = \rho'_0 R_g T_g.$$  

(9)

Here $R_g$ is the reduced gas constant. Here and in what follows, subscripts 0 at the bottom indicate the parameters related to the initial unperturbed state.

In the case $a_g = 0$, this mathematical model leads to the wave equation of a linearly compressible fluid, which makes it possible to use end-to-end calculation methods to describe the propagation of pressure waves in the system under consideration.

3. Discussion

For numerical analysis in the initial state, a bubbly liquid with a hydrate-forming gas has the following values: $p_0 = 10^5$ Pa, $a_0 = 1 \cdot 10^{-3}$ m, $\rho_0^0 = 5.3$ kg/m$^3$, $\alpha_\infty = 10^4$, $T_0 = 274$ K. Thermodynamic parameters for Freon-12 gas are $\lambda_g = 8.76 \cdot 10^{-3}$ W/m·K, $c_g = 590$ J/kg·K, $\gamma = 1.14$, $R_g = 68.76$ J/kg·K. When carrying out the calculations, the presence of surfactants in the liquid was not considered; therefore, the surface tension coefficient is $\sigma = 73 \cdot 10^{-3}$ N/m$^2$. For the process of hydrate formation, we take: $p_{\infty} = 0.42 \cdot 10^5$ Pa, $G = 0.3$, $l_h = 3 \cdot 10^5$ J/kg, $T_* = 5.2$ K, $\beta = 3 \cdot 10^5$. The value of the parameter $\beta$ was chosen in accordance with the agreement between the experimental and calculated data [19].

A bubble curtain in a semi-infinite channel, from the front wall of which to the $z$ plane, the distance is equal to $l_z = 5$ cm, has dimensions $l_x = 2$ cm, $l_y = 10$ cm. At the center of it there is a pressure and mass sensor of the gas hydrate. The liquid at the boundary $x_0$ is influenced by a pressure wave of a stepped profile with an amplitude $\Delta p_0 = 3.3 \cdot 10^5$ Pa. Figure 2 shows the pressure distribution in the liquid at the moment of time 0.16 ms, when the maximum value of the burst is observed, which is formed as a result of nonlinear effects due to the presence of a bubble zone [6]. In this case, the hydrate formation process was not taken into account and the pressure surge shown exceeds the initial signal of $3.3 \cdot 10^5$ Pa by about $2.5 \cdot 10^5$ Pa.
Figure 2. Liquid pressure in a channel with a bubble curtain.

Figure 3 shows the change in pressure in the bubble zone over time, depending on the presence of the process of hydrate formation (black line) and its absence (red line). Figure 4 shows the change in the mass of the formed hydrate.

Figure 3. The dynamics of the shock wave in the bubble curtain with the formation of hydrate (black line) and without it (red line).
Figure 4. The mass of the formed hydrate.

The presented results show that at the considered time intervals, the hydrate formation process does not have a noticeable effect on the shock wave dynamics. But the very presence of a bubble curtain contributes to the formation of pressure surges in its area, which can contribute to the process of intensifying the transition of gas to a hydrated state.

4. Conclusion

In this work, a mathematical model of the propagation of shock waves in a semi-infinite channel with a liquid containing a gas curtain with a hydrate-forming gas is constructed for a two-dimensional case. On the basis of which the dynamics of the pressure wave is shown during its propagation along the channel, accompanied by hydrate formation. It has been established that, at certain parameters of the gas curtain and the wave, it is possible for it to form significant pressure surges, which in turn can contribute to the intensification of hydrate formation.

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