Journal of Applied Mechanics and Technical Physics, Vol. 41, No. 2, 2000

POSSIBILITY OF REACHING HIGHER CONTENTS OF A GAS IN A LIQUID USING SHOCK WAVES

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UDC 532.529

The effect of shock waves on gas absorption by liquid in bubbly media with different degrees of gas solubility is studied. It is shown that a shock wave acting on a gas–liquid medium can significantly enhance the mass transfer between the gas and the liquid.

It is commonly known that intense mixing of the liquid in gas–liquid media enhances the rate of mass transfer between the gas and the liquid. A similar effect can be attained by using acoustic vibrations. In particular, the mass-transfer rate in water saturated with carbon dioxide was found to increase sharply when acoustic vibrations of frequencies 20 to 200 Hz were introduced into the gas–liquid system [1–3]. Acoustic vibrations gave rise to instability at the gas bubble/liquid interface, which ultimately enlarged the contact area between the phases and increased the mass-transfer coefficient. The volume mass-transfer coefficient was raised by a factor of 1.5–4 depending on the amplitude and frequency of acoustic vibrations.

Studies of wave dynamics in multiphase systems [4, 5] point to the possibility of considerable enhancement of mass transfer in bubbly liquids under the action of shock waves. This postulate can be inferred from the general laws of interaction of a shock wave (SW) and a gas–liquid system. Indeed, an SW generated in a liquid with gas bubbles is expected to cause their disintegration into smaller bubbles; hence, it will act to enlarge the contact area between the phases. In addition, due to some special features of the behavior of the medium behind the SW front, the latter sets the liquid in the interfacial region into intense small-scale motion, which is also expected to promote the diffusional flow of gas molecules into the liquid. The expected rise in the mass transfer depends on the amplitude of the pressure wave and on the duration of its action on the liquid.

This work was undertaken to experimentally verify the hypothesis on the significant intensification of gas absorption by the liquid in a gas–liquid medium under the action of shock waves.

The experiments were conducted in two-phase shock tubes, raised in a vertical position, with channel diameters 8 and 25 mm and maximum lengths of the working channels 3.5 and 2.5 m. The design of the tubes is described in [6]. The scheme of the experiment is shown in Fig. 1. The working medium was prepared by bubbling gas through a stationary liquid that occupied the test section of the shock tube. The gas was blown through spicular capillaries 0.1 mm in diameter located at the bottom of the test section. In the experiments, the atmospheric and static pressures, temperature, gas content in the liquid, and bubble dimensions were measured. The pressure fluctuations and variation of the total volume of the medium behind the SW front were registered. The pressure fluctuations were measured by piezoelectric strain gauges with a resonant frequency of 15 kHz. The variation of the gas content in the pressure wave and that of the free level of the medium were registered by the optical shadow scheme [6] and based on the difference in deflection of laser beams passing through the liquid or gas. In this case, the signal from the photodetector of the optical system

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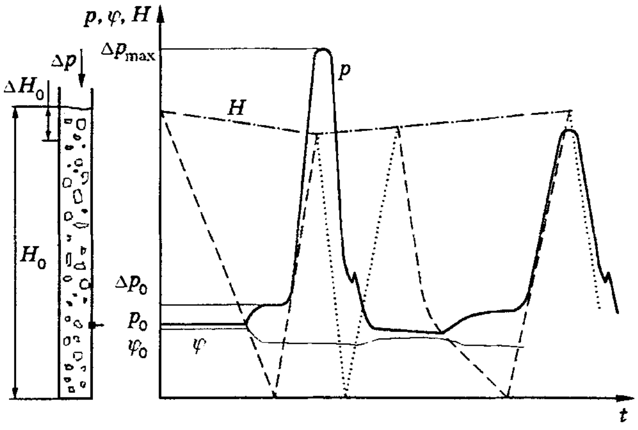

Fig. 1

was registered simultaneously with pressure-wave measurements, and it was used to reconstruct changes in the gas content in the liquid.

The measurement error in pressure fluctuations and gas content of the liquid depended predominantly on the parameters of the measuring system, and the magnitude of this error was determined from calibration tests. The measurement error in the gas content did not exceed 10%. The measurement error in pressure fluctuations, induced by the gauge-amplifier circuit, was also within 10%.

In the experiments, systems with different solubility of gas in liquid were used: water–carbon dioxide and water–ammonia. The equilibrium concentration of the gas as a function of pressure in these systems can be found in [7]. The system "water–air bubbles" was used as a test medium. In all experiments, the initial static pressure was $p_0 = 0.1$, and the amplitude of the disturbing shock wave Δp_0 varied from 0.02 to 2 MPa.

Prior to each experiment, the test section of the tube was filled with a pure liquid and, afterwards, a gas was bubbled through it. To reduce the initial content of the gas in the liquid, we restricted the duration of gas bubbling. The initial gas content in the medium was $\varphi_0 = 1$–25%. The lower values of φ_0 corresponded to the initial state of the medium in the form of a bubbly structure, and the gas content $\varphi_0 = 20$–25% to the missile structure of the medium. An SW of fixed amplitude was introduced into the liquid–gas medium. The SW profile and the variation of the gas content and the free level of the liquid were monitored in the course of wave propagation through the medium. Based on the measured pressure and gas content, the volume mass-transfer coefficient was calculated.

Since direct measurements of the concentration of the gas dissolved in the liquid behind the SW front were impossible because of the considerable time lag of gas analyzers, we used the measured parameters of the SW and gas content in our study.

The time dependences of the pressure $p(t)$ and gas content in the test section of the shock tube $\varphi(t)$ and a diagram illustrating the SW propagation through the medium are shown in Fig. 1. The free level of the liquid $H(t)$ is also shown by the dot-and-dashed curve. These dependences are typical of the media used, and the quantitative characteristics of these dependences differ only in sharpness of the leading front of the wave and its highest amplitude, all other parameters (the amplitude of the initial pressure wave and the gas content of the liquid) being equal.

We consider in more detail the wave evolution in the medium and the behavior of this medium. In Fig. 1, the dashed curve shows a disturbing SW with amplitude Δp_0. Entering the medium, this SW starts propagating in it with a velocity u_0 exceeding the velocity of sound $c_0 = \sqrt{\gamma p_0/(\rho_1 \varphi_0(1-\varphi_0))}$ (γ is the ratio of specific heats and ρ_1 is the density of the liquid) predicted by the homogeneous approximation. As the pressure increases, the two-phase medium becomes more and more disturbed, and a diffusional process begins, which levels out the concentrations of the components in accordance with the equilibrium parameters. For a system consisting of a liquid and bubbles of a soluble gas, this mean that, as the pressure increases,

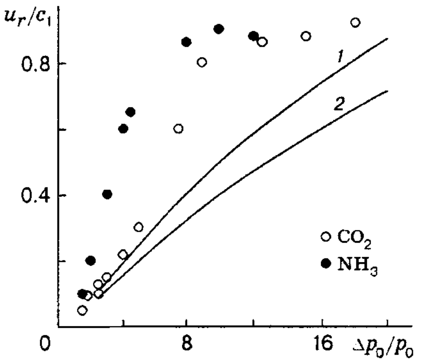

Fig. 2.

the gas content in the liquid diminishes due to gas dissolution in the liquid. When the amplitude of the disturbing pressure wave reaches a certain level, the gas is completely dissolved behind the SW front, and the density of the medium becomes equal to the density of the liquid.

Using integral conservation laws for an SW in a liquid of varying density, one can determine the wave and medium parameters: we have $p = p_0$, $\varphi = \varphi_0$, and $v = 0$ ahead of the wave front and $p = p_l = p_0 + \Delta p_0$, $\varphi = 0$, and $v = v_l$ behind it. Thus, when the gas phase behind the wave front totally vanishes, the liquid acquires a velocity given, in accordance with the conservation laws, by the relation $v_l = \sqrt{\Delta p_0 \varphi_0 / (\rho_l(1 - \varphi_0))}$. After reflection from the bottom of the shock tube, the shock wave starts propagating upwards (the dotted curve in Fig. 1). In the limiting case of total vanishing of the gas phase, the reflected pressure wave propagates with the velocity of sound in the liquid: $u_r \approx c_l$. From the law of conservation of momentum, it follows that the amplitude of the reflected wave is much greater than the amplitude of the pressure wave incident onto the tube bottom. This stage is manifested as an intense pressure overshoot of amplitude $\Delta p_{\max}$ in the measured time-dependent pressure (Fig. 1). Subsequent reflection of the pressure wave from the free surface of the liquid gives rise to an unloading wave (the dotted curve in Fig. 1), and its next reflection from the rigid bottom results in further reduction of pressure. Returning to the free surface of the liquid and being reflected from it, the unloading wave transforms into a compression wave, and the above process starts again. The time evolution of the gas content depends on the amplitude of the pressure wave. For a weak wave with an amplitude $(0.2\text{–}0.5)p_0$, the process has a pulsating character, and then it monotonically decays. For a strong wave, the dependence $\varphi(t)$ has an explicit stepwise shape with a zero asymptotic value behind the pressure-wave front. The duration of the decrease in the gas content from its initial magnitude to zero is in good agreement with the duration of the front of an strong pressure wave and characterizes the time required for complete dissolution of a gas bubble in the liquid.

A qualitative notion of the amount of the gas absorbed by the liquid behind the SW can be gained by considering parameters of the SW reflected from the bottom of the shock tube. The points in Fig. 2 show the measured velocity of the reflected wave u_r, and curves 1 and 2 are the dependences predicted by the model [5], respectively, for $\varphi_0 = 10$ and 20% (a medium with gas bubbles of a fixed mass). The higher the amplitude of the disturbing wave, the closer the velocity of the reflected wave u_r to the limiting velocity, i.e., the velocity of sound in a pure liquid. For amplitudes 0.8–1.0 MPa and higher, the velocity of the reflected wave is close to the velocity of sound in the liquid, which provides indirect evidence for complete dissolution of the gas behind the SW and a transition of the medium from the two- to single-phase state.

In view of the fact that all the gas transforms into the liquid behind the SW front, the amplitude of the reflected wave, according to the law of conservation of momentum, has the form $\Delta p_{\max} = \rho_l c_l v_l$. Substituting the value of v_l, we obtain the maximum amplitude of the reflected wave

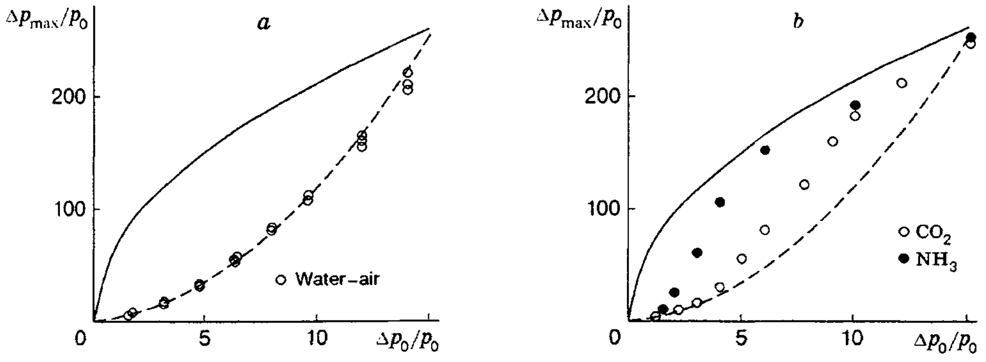

Fig. 3

$$\Delta p_{\max} = c_1 \sqrt{\rho_1/p_0}\,\sqrt{\Delta p_0 \varphi_0/p_0}. \tag{1}$$

In the opposite limiting case of complete conservation of the gas mass (i.e., in the absence of gas dissolution), the amplitude of the reflected wave depends on the degree of gas compression behind the front of the normal SW and, according to the model [5], it is described by the relation

$$\Delta p_{\max}/p_0 = (\Delta p_0/p_0 + 1)^2 - 1. \tag{2}$$

The limiting values for the wave amplitude calculated for $\varphi_0 = 20\%$ by relations (1) and (2) are shown in Fig. 3 by solid and dashed curves, respectively. The measured amplitudes of the reflected wave are expected to fall within the region bounded by these curves. Apparently, the higher the degree of conversion of the gas mass into the liquid state in the pressure wave, the closer the measured wave amplitude to the curve given by relation (1). Thus, from the measured amplitudes of the reflected wave, we can estimate the degree of SW-induced solubility of the gas in the gas–liquid medium. The points in Fig. 3 show experimental data. The measurement results for the test system water–air (Fig. 3a) agree with the values predicted by relation (2). For a gas soluble in the liquid, the situation is different. Figure 3b shows that the behavior of a medium with a soluble gas resembles the behavior of the test system only at small amplitudes of the disturbing pressure wave. When the medium is disturbed by a wave with an amplitude Δp_0 greater than 0.2 MPa for ammonia and 0.4 MPa for carbon dioxide, the value of $\Delta p_{\max}$ far exceeds the amplitude given by the shock adiabat of the bubbly medium. Thus, the higher the initial solubility of the gas in the liquid, the smaller perturbation of pressure needed to affect the process of gas dissolution in the liquid. From this viewpoint, the values indicated above should be considered as threshold values on exceeding which the effect of an SW on gas absorption by the liquid becomes distinctly pronounced.

On exceeding the threshold value and with further growth of the SW amplitude, the measured amplitudes of the reflected wave approach the values determined by the condition of complete conversion of the gas contained in bubbles into the liquid behind the wave front, i.e., by relation (1). From Fig. 3, it follows that complete dissolution of carbon dioxide in water behind the SW front occurs at $\Delta p_0 > 1$ MPa. For the water–ammonia system, the threshold SW amplitude for which full dissolution of the gas is observed is lower than in the previous case and amounts to about 0.6 MPa.

The threshold SW amplitude at which complete dissolution of the gas behind the SW front occurs depends not only on the gas solubility in the liquid but also on the behavior of gas bubbles. It is known [5, 8] that the bubbles contained in the liquid suffer intense destruction into smaller bubbles when the threshold SW amplitude is attained. The latter is determined by the type of the gas and the conditions of interaction between the bubbles and the shock wave. The disintegration into smaller bubbles results in a larger interfacial area between the gas and the liquid. As a result of disintegration, the specific area of the interface between the phases increases by a factor D_0/d, where D_0 is the initial diameter of the bubble and d is the typical diameter of the newly formed smaller bubbles. In addition, the high-velocity cumulative jets arising after the destruction of bubbles cause additional small-scale turbulization of the liquid, which promotes a rise in the

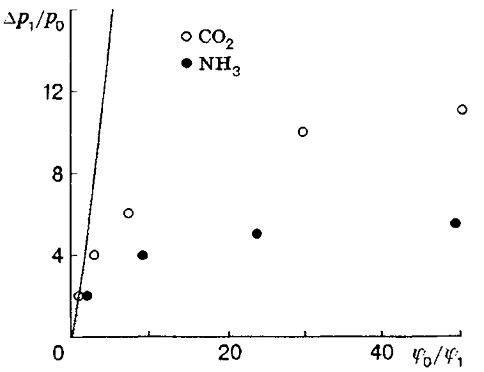

Fig. 4

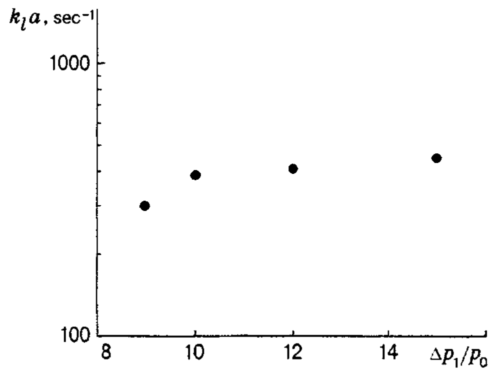

Fig. 5

mass-transfer rate at the interface. The critical SW amplitude at which intense destruction of the bubbles begins depends on the particular physical model chosen to describe the interfacial instability. The model [5] gives the following condition for the beginning of disintegration: $\mathrm{We} = D_0 \rho_g v_1^2/\sigma > \mathrm{We}_{cr} \approx 2\pi$. Here We is the Weber number, ρ_g is the gas density, and σ is the surface tension. The predicted SW amplitudes are 0.2–0.4 MPa, which is in line with the above threshold SW amplitude on exceeding which the effect of the SW on intensification of gas dissolution in the liquid becomes clearly pronounced.

Based on the measured gas content behind the SW front and the pressure in it, one can determine the amount of the gas dissolved in the liquid during the time of action of the SW. Figure 4 shows the lowest values of the gas content φ_1 in the medium attainable in an SW of amplitude Δp_0. The dependence $\varphi_0/\varphi_1 = F(\Delta p_1/p_0)$ is the equation of state for the gas–liquid medium. In a medium without mass transfer, the evolution of the gas content behind the SW front would depend only on the degree of gas compression in the SW of an amplitude Δp_1. In a system with gas bubbles of constant mass, the gas content behind the SW front can be found from the following relation given by the homogeneous model of a gas–liquid medium, which is a shock adiabat [5]:

$$\varphi_0/\varphi_1 = \varphi_0 + (1 - \varphi_0)(1 + \Delta p_1/p_0)^{\gamma}. \tag{3}$$

The gas content behind the SW front in a wave of amplitude Δp_1 calculated by the above relation is shown in Fig. 4 by the solid curve. As follows from Fig. 4, the measured values of φ_1 follow the shock adiabat only for small SW amplitudes. As the wave amplitude increases, these values clearly deviate from adiabatic relation (3). For example, a shock wave of 0.6 MPa amplitude in a water–carbon dioxide medium results in a decrease in the initial gas content $\varphi_0 = 20\%$ down to $\varphi_1 = 2\%$ behind the wave front, whereas isothermic compression of the gas in such a wave yields a value greater by a factor of 2.5. This difference increases markedly with increasing SW amplitude. Owing to small values of the gas content behind the wave front ($\varphi_1 \to 0$), the measurement error appreciably increases. For $\Delta p_1 > 1$ MPa, we can only say that the ratio φ_0/φ_1 is within the interval 50–1000. The values of Δp_1 for which a dramatic deviation of the gas content from the values predicted by the shock adiabat for a bubbly liquid is observed can be considered as threshold values above which complete dissolution of the gas in water behind the SW front occurs. The values $\Delta p_1 = 0.4$–0.6 MPa for ammonia and $\Delta p_1 = 0.8$–1.2 MPa for carbon dioxide (Fig. 4) agree with the data obtained from the measured velocities and amplitudes of the reflected pressure wave.

The characteristic time of complete dissolution of carbon dioxide for $\Delta p_1 = 1.5$ MPa is $t \approx 2$–3 msec. This time becomes shorter with increasing Δp_1. It can be noted that complete dissolution of the gas may also occur at a smaller amplitude of the disturbing wave; the time of complete dissolution (after SW passage) in this case, however, exceeds the time required for the SW to propagate through the entire height of the bubble–liquid column. Although it is not an easy task to derive a relation between the time of complete dissolution and the wave amplitude because of the influence of the reflected wave, the latter value may be considered as the lower estimate for this parameter. For instance, complete dissolution of carbon dioxide in water under the action of a wave of amplitude 0.05 MPa proceeds in 2–4 sec after the wave passage. It should also be noted that the characteristic time of dissolution of a carbon-dioxide bubble 5 mm in diameter which freely ascends in water is 20–30 sec [9].

Finally, we estimate the volume mass-transfer coefficient in water with dissolved carbon dioxide under conditions of an SW with amplitude above the threshold value. We assume here that the bubbles contain pure carbon dioxide, the concentration of carbon dioxide dissolved in water is close to zero prior to SW action, the diffusional resistance is concentrated in the liquid, the liquid temperature is constant, and the total pressure in the system is equal to the static pressure behind the SW. Because of the small volume fraction of the gas, the change in the concentration of the solution formed can be ignored. The equation of material balance is written as $dV/dt = -k_l S$, where V is the volume of the gas, S is its contact area with the liquid, and k_l is the mass-transfer coefficient. The area of the bubble can significantly increase due to SW-induced destruction. To eliminate this unknown quantity, we find the value $k_l a = (1/\Delta t_*) \ln (V_0/V_*)$, where Δt_* is the interval between the moments at which the initial (V_0) and final (V_*) volumes of the gas in the medium are determined and a is the specific interfacial area between the phases. Figure 5 shows the volume mass-transfer coefficient for carbon dioxide in water versus the amplitude of an SW passing through the bubbly medium. As is seen, rather high values of the mass-transfer coefficient are attained. For carbon dioxide bubbled through water, the mass-transfer coefficient reaches only 0.1–0.2 sec^{-1} [9], whereas acoustic vibrations raise it to 0.8–1.0 sec^{-1} [1, 2]. Thus, an SW can considerably increase both the mass-transfer coefficient and the rate of gas absorption by the liquid.

This work was partially supported by the Russian Foundation for Fundamental Research (grant No. 96-05-65956).

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