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STABILITY OF FORM AND CONCENTRATION OF ENERGY AT COMPRESSION OF GAS-VAPOR BUBBLES FORMED IN PULSED ACOUSTIC FIELDS

Pyotr Golubnichy, Denis Reshetnyak, Yuri Krutov, Eugenie Nikitin

Volodymyr Dal East-Ukrainian National University, Lugansk, Ukraine

Summary. We investigated the Rayleigh-Taylor stability of a single cavitation bubble in water, formed and pulsating in the field of a bipolar acoustic pulse of varying amplitude, duration and polarity. For the pulse parameters under which the shape of the bubble at the final stage of collapse is nearly spherical, we calculated the thermodynamic conditions in the bubble at the time of the first collapse.

Key words: cavitation, acoustic pulse, bubble, collapse, stability**.**

INTRODUCTION

A number of studies [1-3] have shown that the use of acoustic cavitation to achieve extreme thermodynamic parameters (temperature, pressure, density) in collapsing bubbles in a liquid looks very promising. This method of obtaining high energy densities is proposed for use in applications of plasma physics, nuclear physics (see eg. [4,5]) and high-energy chemistry [6]. The practical importance is the stability of the spherical shape of the collapsing bubbles, because concentration of energy is more efficient in the case of spherical collapse, and there will be achieved a higher peak temperature.

To increase the energy density at the stage of the collapse of bubbles in the work [7] the authors proposed to create cavitation in liquids using pulsed acoustic fields. In these fields, the volume concentration of bubbles in the zone of cavitation should be significantly less than in the case of a continuous wave, and the acoustic permeability of the zone above. This leads to a weakening of the interaction of bubbles and more effective absorption of acoustic energy of each bubble in the cavitation zone [8]. Due to weakening in interaction of bubbles in the slow phase ripple there is no distortion in their shape. This helps to preserve the sphericity at the stage of rapid compression.

Using single acoustic pulse (AP) also allows one to ignore the impact on the shape of the bubble in the vicinity of the first collapse of parametric instability, which develops only after several of its pulsations. This imposes fewer restrictions on the conditions of the experiments than in the study of single-bubble sonoluminescence [9]. Thus, in the field of AP mainly Rayleigh-Taylor instability will result in distortion of the bubble shape, which occurs at the accelerated collapse of the bubble.

In the present work we wanted to investigate the stability of Rayleigh-Taylor a single cavitation bubble in water, which was formed and was pulsating in the field of bipolar AP of varying amplitude, duration and polarity. Then we calculated thermodynamic conditions in the bubble at the time of the first collapse. Consideration was given to the bubble, which contained non-condensable gas – helium. In this case at the time of collapse of the cavity there is a comparably little amount of water vapor in it [7], this results in a stronger compression and enables to achieve higher temperature and pressure.

[i]MODEL OF GAS-VAPOR BUBBLE

Law of change in the radius of a spherical bubble in a compressible viscous fluid was determined by the Rayleigh-Plesset equation in the form [10]:

$$\left(1-\frac{U}{c_{lR}}\right)R\frac{dU}{dt}+\frac{3}{2}\left(1-\frac{U}{3c_{lR}}\right)U^2= \\ =\left(1+\frac{U}{c_{lR}}\right)\left[H_R-\frac{1}{\rho_{l0}}P_s\left(t+\frac{R}{c_{l0}}\right)\right]+\frac{R}{c_{lR}}\frac{dH_R}{dt}, \quad (1)$$

$$c_{lR}=\sqrt{\left.\frac{dP}{d\rho_l}\right|_R}=c_{l0}\left(\frac{P_{lR}+B}{P_0+B}\right)^{\frac{m-1}{2m}},\ H_R=\int_{P_0}^{P_l}\frac{dP}{\rho_l}=\frac{m}{m-1}\left(\frac{P_{lR}+B}{\rho_{lR}}-\frac{P_0+B}{\rho_{lR}}\right),$$

$$\rho_{lR}=\rho_{l0}\left(\frac{P_{lR}+B}{P_0+B}\right)^{\frac{1}{m}},\ P_{lR}=P_R-\frac{2\sigma_l}{R}-\frac{4\mu_l U}{R},$$

where: $U=dR/dt$, R − variable speed wall and the bubble radius; c_{lR}, H_R – the speed of sound in water and the enthalpy at the bubble surface; ρ_{l0}, c_{l0} – water density and velocity of sound in it away from the bubble; $P_s(t)$ – variable acoustic pressure in fluid; P_{lR}, ρ_{lR} − pressure in the liquid and the density of water at the surface of the bubble; P_0 = 1 bar − hydrostatic pressure, B = 3049.13 bar, m = 7.15; σ_l, μ_l – coefficient of surface tension and dynamic viscosity of water; P_R − pressure of gas-vapor mixture (GVP) on the bubble wall.

Pressure of GVP on the wall of the bubble, if we take into account the inertial correction proposed in [11] will be: $P_R=P_c-1/2\,\rho_m R\,dU/dt$, where ρ_m − average density of GVP in the bubble; P_c − pressure of GVP in the central region of the bubble. Pressure P_c in this work is defined by the Redlich-Kwong (RK) equation:

$$P_c=\frac{\nu_m R_u T}{V-\nu_m b_m}-\frac{\nu_m^2 a_m}{\sqrt{T}V\left(V-\nu_m b_m\right)}, \quad (2)$$

where: ν_m, b_m, a_m − the number of moles of the GVP and coefficients of the RK mixture; R_u = 8.314 J/(mol·K) − the universal gas constant; T, V − the temperature of the contents and volume of the bubble.

Compression of the bubble is accompanied by a rapid increase in temperature of its contents. The analysis showed that for such a process the second term in (2) can be neglected. Then, after simple transformations the equation of state GVP can be written as:

$$P_c = \frac{\rho_m R_{gm} T}{1 - b_m^* \rho_m}. \tag{3}$$

In equation (3) notation: $R_{gm} = \sum_i y_i R_u / M_i$, $b_m^* = \sum_i y_i b_i / M_i$, where: y_i, M_i, b_i − mass fractions, molar mass and the constant of the RK equation for components of the mixture. Although in the work we've taken into account the influence of thermal dissociation of water vapor on the energy in the cavity (see below), in our model for the dissociated phase we used equation of state of nondissociated water vapor ("frozen" shock adiabatic process). This assumption for the state of the dissociated phase, was suggested in [1] and justified by the velocity at the final stage of collapse, where dissociation of molecules is possible. Thus, as in (3): $R_{gm} = R_u \left(y_g / M_g + y_v / M_v \right)$, $b_m^* = y_g b_g / M_g + y_v b_v / M_v$; $b_g = 16.37 \cdot 10^{-6}$ m^3/mol, $b_v = 21.09 \cdot 10^{-6}$ m^3/mol, $M_g = 4 \cdot 10^{-3}$ kg/mol, $M_v = 18 \cdot 10^{-3}$ kg/mol.

External various pressure was set at a flat sinusoidal AP of different polarity: $P_s(t) = \mp P_a \sin(2\pi t/\tau) F(t)$, where P_a, τ − amplitude and pulse duration; $F(t) = \left\{ 1 + \exp\left[\frac{(t-\tau)}{0.01\tau} \right] \right\}^{-1}$ − the Fermi function. Sign (−) corresponds to the pulse expansion-compression (E-C), (+) − pulse-compression-expansion (C-E).

The processes of evaporation, condensation and heat transfer in this work were approximated by the model of the boundary diffusion layer, which is in thermal equilibrium with the surrounding fluid [12]. The equations of the flow of vapor molecules and the thermal conductivity through the surface of the bubble in this approximation are defined as:

$$\frac{dN_v^d}{dt} = 4\pi R^2 D \frac{n_{v0} - n_v}{l_d}, \quad l_d = \min\left(\sqrt{\frac{RD}{|U|}}, \frac{R}{\pi} \right), \tag{4}$$

$$\frac{dQ}{dt} = 4\pi R^2 \kappa_m \frac{T_0 - T}{l_{th}}, \quad l_{th} = \min\left(\sqrt{\frac{R\chi_m}{|U|}}, \frac{R}{\pi} \right), \tag{5}$$

where: D − coefficient of binary diffusion; n_{v0}, n_v – equilibrium and current concentration of vapor in the bubble; l_d – the thickness of diffusion layer; κ_m, χ_m – thermal conductivity and thermal diffusivity of the mixture in a layer; T_0 − temperature of the water; l_{th} – the thickness of the thermal diffusion layer.

Since it is assumed that the boundary layer is in thermal equilibrium with the liquid, the transport coefficients in (4) and (5) are set at T_0. For this reason, we assume that in the boundary layer water vapor is not dissociated. Thus, in our case, the GVS in the boundary layer will be a two-component one (water vapor and helium). Included in the equation (4) the coefficient of binary diffusion, depending on the concentration of the mixture is defined as [13]: $D = D_0 \left(n_0 / n_m \right)$, where $D_0 = 7.38 \cdot 10^{-5}$ m^2/s − coefficient of binary diffusion at normal pressure, which is determined by the method of Fuller, Shettler and Griddings [13]; n_0, n_m – the concentration of gas at normal pressure and the current concentration of the mixture in the layer. Thermal conductivity of a binary mixture κm, appearing in (5) was calculated by the method of Lindsay and Bromley [13].

Thermodynamic conditions inside the bubble at the stage of collapse also depend on the chemical transformations of components of the mixture. At this stage (heating) the influence on the peak temperature in the bubble will mainly due to the thermal dissociation reaction: $H_2O+M \leftrightarrow H+OH+M$, $OH+M \leftrightarrow O+H+M$, where M − any particle. The speed of forward and backward reaction is described by kinetic equations:

$$r_{f,j} = k_{f,j} n_{tot} n_A, \; r_{b,j} = \beta k_{b,j} n_{tot} n_B n_C, \; \beta = \frac{\exp\left(\dfrac{b_m^* \rho_m}{1 - b_m^* \rho_m} \right)}{1 - b_m^* \rho_m}, \tag{6}$$

where: $k_{f,j}$, $k_{b,j}$ − the rates of direct and reverse reactions; n_{tot} − the total concentration of particles in the cavity; n_A, n_B, n_C − the concentration of particles of type A, B and C, that take part in the reaction. β − the factor describing the acceleration of the backward reaction due to high density of contents of the bubble at collapse [12].

The parameters of the reaction rates were determined using modified Arrhenius equation: $k_{f,j} = A_{f,j} T^{c_{f,j}} \exp\left(-E_{f,j} / kT \right)$, $k_{b,j} = A_{b,j} T^{c_{b,j}} \exp\left(-E_{b,j} / kT \right)$, where $A_{f,j}$, $A_{b,j}$, $c_{f,j}$, $c_{b,j}$ − parameters of the Arrhenius equation; $E_{f,j}$, $E_{b,j}$ − activation energy of forward and backward reaction; $k = 1.38 \cdot 10^{-23}$ J/K − Boltzmann constant. Arrhenius equation parameter values are taken from [14] (see Table 1).

Table 1. **Arrhenius equation parameters**

j	reaction	$A_{f,j}$, m^3/(mol·s)	$c_{f,j}$	$E_{f,j}/k$, K	$A_{b,j}$, m^6/(mol^2·s)	$c_{b,j}$	$E_{b,j}/k$, K	ΔE_j, kJ/mol
1	$H_2O+M \leftrightarrow H+OH+M$	$1 \cdot 10^{18}$	-2.2	59000	$2.2 \cdot 10^{10}$	-2	0	-498
2	$OH+M \leftrightarrow O+H+M$	$8.5 \cdot 10^{12}$	-1	50830	$7.1 \cdot 10^{6}$	-1	0	-428

The equation for calculating the temperature in the model we used is as follows:

$$\frac{dT}{dt} = \frac{1}{C_v} \left\{ \frac{dQ}{dt} - P_R \frac{dV}{dt} + \left[4T_0 - 3T - \sum_i \left(\frac{\theta_{vi}}{\exp\left(\theta_{vi} / T \right) - 1} \right) \right] k \frac{dN_v}{dt} + V \sum_j r_j \Delta E_j \right\}, \tag{7}$$

where: C_v – specific heat of GVS in the bubble; θ_{vi} – characteristic vibration temperature of the water molecule (5262.4 K, 5404.6 K and 2294.9 K); $r_j = r_{f,j} - r_{b,j}$ and ΔE_j – speed and power (thermal effects) of chemical reactions. Heat capacity of the mixture in the cavity is defined by the expression:

$$C_v = \left[\left(3+S_v\right)n_v + \left(5/2+S_{OH}\right)n_{OH} + 3/2\left(n_H + n_O\right)\right]kV, \tag{8}$$

where: n_{OH}, n_H, n_O – concentration of the components of the mixture. Also in (8) notation for the vibrational components of the specific heat:

$$S_v = \sum_i \frac{\left(\theta_{vi}/T\right)^2 \exp\left(\theta_{vi}/T\right)}{\left(\exp\left(\theta_{vi}/T\right)-1\right)^2},\; S_{OH} = \frac{\left(\theta_{OH}/T\right)^2 \exp\left(\theta_{OH}/T\right)}{\left(\exp\left(\theta_{OH}/T\right)-1\right)^2},$$

where: θ_{OH} = 5378.8 K – characteristic vibration temperature of the radical OH.

THE STABILITY OF THE BUBBLE'S SHAPE

At a rapid compression of the bubble's surface some disturbances may appear that may lead to nonspherical collapse or fragmentation of the bubble. Development of distortion in spherical shape, which correspond to Rayleigh-Taylor instability, was investigated using the equation [15]:

$$\frac{d^2 a_n}{dt^2} + B_n\left(t\right)\frac{da_n}{dt} - A_n\left(t\right)a_n = 0, \tag{9}$$

$$B_n\left(t\right) = \frac{\rho_{lR}/\left(n+1\right)}{\rho_m/n + \rho_{lR}/\left(n+1\right)}\left\{\frac{3U}{R} + \frac{2\mu_l}{\rho_{lR}R^2}\left[-\left(n-1\right)\left(n+1\right)\left(n+2\right) + \frac{n\left(n+2\right)^2}{1+2\delta/R}\right]\right\},$$

$$A_n\left(t\right) = \frac{\rho_{lR}/\left(n+1\right)}{\rho_m/n + \rho_{lR}/\left(n+1\right)}\left(n-1\right)\times$$

$$\times\left\{\frac{1}{R}\frac{dU}{dt} - \left(n+1\right)\left(n+2\right)\frac{\sigma_l}{\rho_{lR}R^3} - \frac{2\mu_l U}{\rho_{lR}R^3}\left[\left(n+1\right)\left(n+2\right) - \frac{n\left(n+2\right)}{1+2\delta/R}\right]\right\} - \frac{\rho_m/n}{\rho_m/n + \rho_{lR}/\left(n+1\right)},$$

where: n – number of spherical harmonics; $\delta = \min\left[\sqrt{\mu_l \tau/\left(2\pi\rho_{lR}\right)}, R/\left(2n\right)\right]$ – the characteristic thickness of the layer in which perturbations are localized. Note that equation (9) takes into account the influence of varying density and water content of the bubble at its surface on the development of perturbations in the cavity surface.

RESULTS AND DISCUSSION

For the analysis of disturbances of the bubble's shape in the vicinity of the first collapse we've used numerical integration of equations (1), (4), (7), (9) by Rosenbrock's method. In calculations we used the following values of parameters of the AP: P_a = 1.5 – 20 bar, τ = 5, 10 and 15 μs, the polarity of the E-C and C-E; constants of water and vapor: T_0 = 277 K, ρ_{l0} = 1000 kg/m^3, c_{l0} = 1444.5 m/s, $\sigma_l = 7.5\cdot10^{-2}$ N/m, $\mu_l = 1.57\cdot10^{-3}$ Pa·s, $n_{v0} = 2.17\cdot10^{23}$ m^{-3}. The initial radius of the gas microbubble kernel in the water was taken R_0 = 1.5 μm [16], under the given conditions such a bubble would contain $7.4\cdot10^8$ of helium molecules. The instabilities were investigated for n = 2 (the lowest harmonic, leading to a loss of sphericity), the magnitude of the initial perturbation was taken as $a_2^0 = 0.01R_0$. Test calculations showed that the dissociation of steam has little effect on the amplitude of the perturbation: the relative change $\left|\left(a_2^{diss} - a_2\right)/a_2^{diss}\right|\cdot100\%$ did not exceed several percent. Therefore, when calculating the perturbation effect of dissociation of water vapor is not taken into account. The typical pattern of disturbances in the vicinity of the first collapse of a bubble pulsating in the field momentum E-C, is shown in Fig. 1a. Fig. 1b. shows the dependence of the relative amplitudes of the perturbations on the amplitude of the E-C. These results show that if the pulsations in the field of AP E-C have low amplitude, than the collapse is nearly spherical. An increase in the amplitude of the AP disturbances may develop both outside and inside of the bubble or have their value close to zero, which is also confirmed by the results of [17]. The resulting value of the perturbation is obviously determined by the combined influence on its speed and acceleration of the bubble wall during compression and increasing the density of the contents of the bubble and the water at its surface with increasing acoustic pressure. Thus, there are relatively high values of the AP amplitude, at which the collapse of the bubbles will also be spherical. The existence of such "stability islets" can be used for carrying out research of cavitation in a pulse field. Increasing the duration of the AP, in general, stabilizes the collapse of bubbles.

Calculations of surface disturbances of the bubble, pulsating in the field of pulse C-E showed that at the time of collapse, they are damped for all considered values of τ and P_a. Fig. 2. shows the evolution of the perturbation pulse C-E for the same duration and amplitude of the pulse as for the perturbation in Fig. 1a.

Equation (1) describes the dynamics of a spherical cavity, so the calculation of the thermodynamic conditions in the collapsing bubble will be adequate only for such pulsed fields in which its form in the final stage of compression differs only slightly from spherical. The possible formation of a converging shock wave and shock-wave heating of the contents of the bubble is also implemented for spherical collapse. Using the data obtained for the AP, which leads to the spherical collapse, we have identified peak values of temperature and pressure at the time of the first collapse of the bubble. Calculations were made as those described above, but instead of equation (9) we examined the kinetic equations, which were written with the use of (6) and determined the amount of H_2O, OH, O and H in a bubble: $dn_H/dt = r_{f,1} - r_{b,1} + r_{f,2} - r_{b,2}$, $dn_O/dt = r_{f,2} - r_{b,2}$, $dn_{OH}/dt = r_{f,1} - r_{b,1} - r_{f,2} + r_{b,2}$, $dn_v/dt = r_{b,1} - r_{f,1} + 1/V\, dN_v^d/dt$.

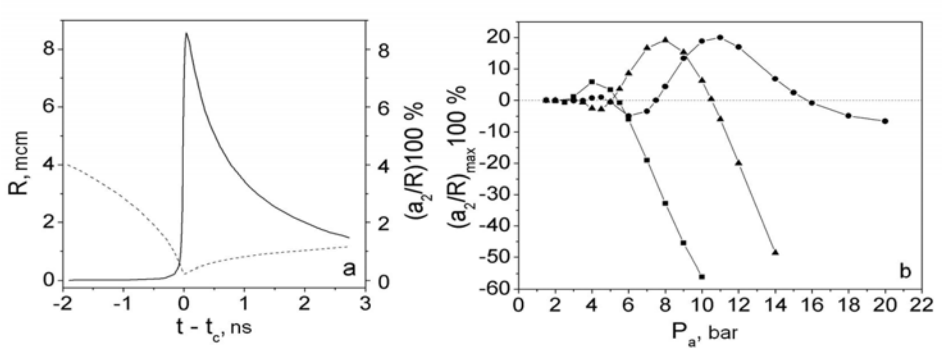

Fig. 1. (a) The development of perturbations in the vicinity of the collapse of the bubble when τ = 10 μs, P_a = 6 bar, the pulse E-C (t_c = 9849.9 ns) and (b) the amplitude of the perturbations of the amplitude of the AP E-C, with the duration τ: 5 μs (■), 10 μs (▲), 15 μs (●)

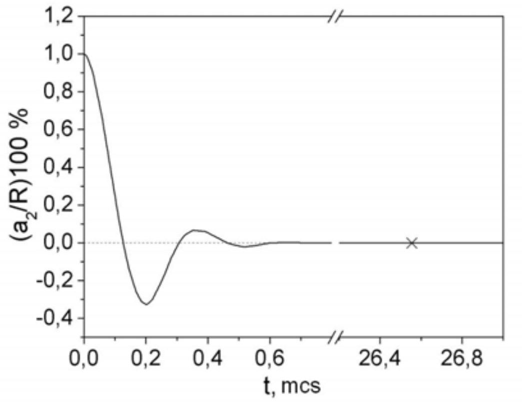

Fig. 2. Dynamics of perturbations in the AP C-E: τ = 10 μs, P_a = 6 bar, the moment of collapse indicated (×)

The results of calculations of thermodynamic conditions in the bubble are shown in Fig. 3 and 4. Calculations showed that with the increase in the pulse amplitude to some value, the peak temperature in the bubble grows, and then begins to decline. The critical value Pa decreases with increase in the pulse duration τ. This effect is quite obvious in the case of AP C-E (Fig. 3b). Explanation of it can be as follows: an increase in tensile acoustic pressure and pulse duration increases the maximum size of the bubble, i.e. its potential energy, which should lead to an increase in temperature of the collapse. On the other hand with an increase in cavity size, increases the amount of water vapor, which does not have time to condense at the time of collapse. This lowers the peak temperature. Thus, the maximum temperature is determined by the competing action of these two factors and decreases from a certain value P_a.

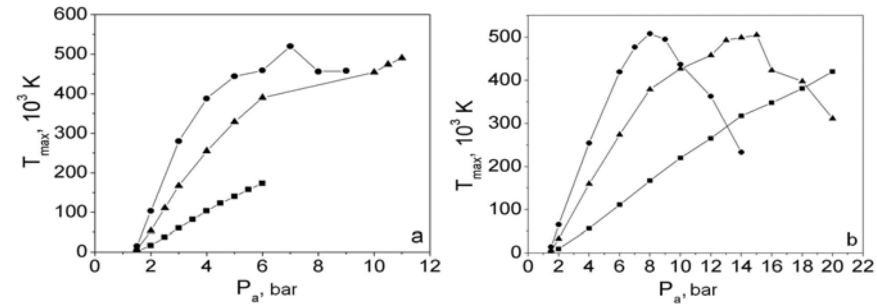

Fig. 3. Dependence of peak temperature in the bubble on the amplitude and duration of the AP E-C (a) and C-E (b); τ: 5 μs (■), 10 μs (▲), 15 μs (●)

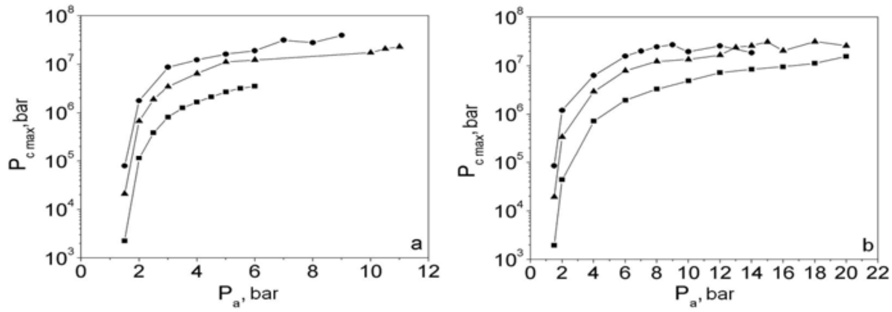

Fig. 4. Dependence of the peak pressure in the bubble on the amplitude and duration of the AP E-C (a) and C-E (b); τ: 5 μs (■), 10 μs (▲), 15 μs (●)

With an increase in P_a and τ peak pressure in the bubble increases after exit to a smooth part of the curve. It is determined not only by temperature but also by the density of GVS, which increases with P_a (Fig. 5).

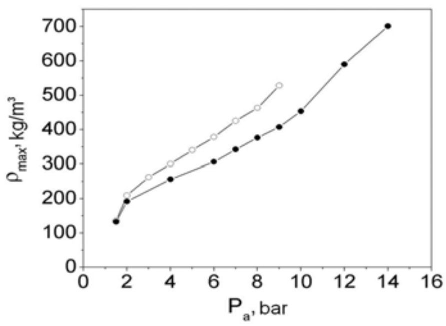

Fig. 5. Dependence of the maximum density of the amplitude GVS AP E-C (○) and C-E (●) for τ = 15 μs

Even with the loss of energy due to dissociation of steam, calculations show that the peak temperature is $T_{max} \approx 5\cdot10^5$ K. Naturally, in such circumstances there will be a development of ionization components of the mixture, which will lead to a decrease in temperature. This reduction in temperature can be estimated as: $\Delta T_{\max} = \Delta E_{ion} / C_v^*$, where: ΔE_{ion} − energy loss by ionization; C_v^* − the heat of the ionized mixture. Assuming that all vapor molecules disintegrate into atoms, heat of the mixture will be: $C_v^* = 3/2\left(n_g + 3n_v + n_e\right)kV,$ where n_g − concentration of gas (helium); n_e − electron concentration. Losses due to ionization were as follows: $\Delta E_{ion} = V\sum_p \sum_q n_{q,p} I_{q-1,p}$, where: $n_{q,p}$ − concentration of ions of species p with charge q; $I_{q-1,p}$ − ionization potential source of an atom or ion. Concentrations of ions $n_{q,p}$ and electrons n_e determined by the Saha formula:

$$\frac{n_e n_{q,p}}{n_{q-1,p}} = 2\frac{g_{q,p}}{g_{q-1,p}}\left(\frac{2\pi m_e kT}{h^2}\right)^{3/2} \exp\left(-\frac{I_{q-1,p}}{kT}\right),$$

where: $g_{q,p}$ and $g_{q-1,p}$ − statistical weight of the ion and the initial particle; m_e − mass of the electron; $h = 6.63\cdot10^{-34}$ J·s − Planck's constant. We assumed formation of such ions as He^+, He^{2+}, H^+, O^+, O^{2+} and O^{3+}. Such an estimate of ΔT_{max} for the case of AP C-E with $\tau = 15$ μs, and $P_a = 8$ bar, which without taking into account the ionization, leads to a peak temperature of $T_{max} = 5.08\cdot10^5$ K, showed a value lower than $\Delta T_{max} = 1.65\cdot10^5$ K. Note that our estimate of ΔT_{max}, apparently, was somewhat over-estimated due to the effect of reducing the ionization potentials in nonideal plasma. This question requires further analysis.

CONCLUSION

In the work we've studied influence of amplitude, duration and polarity of bipolar AP on stability of cavitation bubbles' formation at the final stage of their compression. For pulses that lead to collapse of sphericity we've calculated the maximum achievable values of temperature, pressure and density of the contents of the bubble. We've established extreme dependence of peak temperature on the magnitude of the AP. The observed strict thermodynamic conditions indicate prospects of using pulsed acoustic fields in sonophysic and sonochemistry which often require high energy density. The efficiency of such techniques can increase the concentration of energy, impact on the environment by the periodic sequence of pulses. Note that the resulting simulation of the maximum temperature (~10^5 K) have an order of magnitude higher than in experiments on single-bubble sonoluminescence (~10^4 K) [9].

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УСТОЙЧИВОСТЬ ФОРМЫ И КОНЦЕНТРИРОВАНИЕ ЭНЕРГИИ ПРИ СЖАТИИ ПАРОГАЗОВЫХ ПУЗЫРЬКОВ, ОБРАЗОВАННЫХ В ИМПУЛЬСНЫХ АКУСТИЧЕСКИХ ПОЛЯХ

Голубничий П.И., Решетняк Д.В., Крутов Ю.М., Никитин Е.В.

Аннотация. В работе исследован на устойчивость Релея-Тейлора одиночный кавитационный пузырек в воде, образованный и пульсирующий в поле биполярного акустического импульса различной амплитуды, длительности и полярности. Для параметров импульса, при которых форма пузырька на конечной стадии схлопывания близка к сферической, были рассчитаны термодинамические условия в пузырьке в момент первого коллапса.

Ключевые слова: импульсные акустические поля, акустическая кавитация, высокие плотности энергии, методики концентрирования энергии.