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LOCALIZATION AND INTERACTION OF SMALL PHASE INCLUSIONS IN THE SOUND FIELD IN THE PRESENCE OF PHASE TRANSFORMATIONS

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Phase inclusions (PI) occupy various positions in a sound field depending on a relation between density and compressibility both of inclusions and fluids. Such pattern becomes complicated in the presence of phase transformations and a size effect originates, namely the essential dependence on the sizes PI. It is shown that with reduction of the size PI a mass transfer related to phase transformations starts to import the increasing contribution to compressibility PI which result is occurrence of additional proper compressibility which can exceed the adiabatic compressibility PI considerably. As a result this circumstance can lead to change of a place of PI entrapment by ultrasonic field depending on their size and sound frequency. The last is revealed most strongly for small sizes of crystallization centers. Essential dependence of a localization place on the size of crystallization centers can be taken as a base of guidance for decomposition of structure of solid crystallizing in an ultrasonic field.

It is known, that phase inclusions (PI) occupy various positions in a sound field depending on a relation between density and effective compressibility of inclusions and fluids [1-3]. The most known case is a behaviour of bubbles in the field of a standing wave [3]. For bubbles it is important the relation between sound frequency and resonant frequency of bubbles, so a small bubbles with a resonant frequency of more then a sound frequency of a standing wave are localized in pressure crests while large bubbles are thrown out from fields with large pressure and they are agglomerated in pressure knots. Solid particles with a density more then a fluid density and with a small compressibility in comparison with a fluid compressibility are localized in pressure knots, i.e. behave like large above resonance bubbles. Such pattern sharply becomes complicated in the presence of phase transformations. The matter is that in the presence of phase transformations along with the contribution to effective compressibility of purely mechanical effects it is important as well a mass transfer leading to additional compressibility which sharply changes a habitual pattern of PI behaviour in external field of pressure, especially at the small sizes PI [4].

The parameters determining a place of localization PI in a standing wave. Localization of foreign particles in a sound field depends on a sign of z in the expression for force F acting on a particle of radius R in the field. In the case of a standing sound wave its spatial part can be written down in the form of [4]:

$$F(R,\omega;x) = 4\pi R^2 (k_1 R)\frac{P_m{}^2}{2\rho c_1{}^2}\sin(2k_1 x)\zeta(R,\omega), \qquad \zeta(R,\omega) = \frac{\rho' + 2\Delta\rho/3}{2\rho' + \rho} - \frac{\mathrm{K}\gamma}{3\beta}. \quad (1)$$

Here P_m and $k_1 = w/c_1$ are an amplitude of pressure and a sound wave number in a fluid, r and b are a density and an isothermal compressibility of a fluid, $\Delta\rho = \rho' - \rho$, γ is an adiabatic coefficient, accents concern to PI, K is a compressibility PI, x is co-ordinate along which the standing wave propagates. If $z > 0$, then particles settle down in pressure knots. As quantity K usually has in view the adiabatic compressibility β'/γ' [1-4]. For solid particles $\beta'/\gamma' < \beta/\gamma$, therefore we obtain

$$\zeta \cong \left(\rho' + 2\Delta\rho/3\right)/\left(2\rho' + \rho\right) > 0\,. \quad (2)$$

The plus sign of quantity ζ means, that the particle occupies a standing in pressure knots. For gas bubbles the situation becomes complicated owing to presence of resonant properties. For them compressibility $\mathrm{K} = \mathrm{K}(R,\omega)$ and essentially depends on a relation between frequency of a sound and resonant Minnaert frequency $\omega_0 = (1/R)\sqrt{3\gamma'/\beta'}$. It is possible to write the following approximate formulas for $\mathrm{K}(R,\omega)$ [3]:

$$\mathrm{K}(R,\omega) \cong (\beta'/\gamma')/3Q(R,\omega), \qquad Q(R,\omega) = 1 - (\omega/\omega_0)^2(1 + i\delta). \quad (3)$$

Considering, that quantity of the adiabatic compressibility $\beta'/\gamma' \approx 1/\gamma' P_0$ essentially exceeds compressibility of a fluid from (1) follows [3]:

$$\zeta(R,\omega) \cong -(\beta'/\gamma')/(\beta/\gamma)(1/Q(R,\omega)) = -(\rho c_1{}^2/\rho' c_1'^2)\left[1 - (\omega/\omega_0)^2(1 + i\delta)\right]^{-1}. \quad (4)$$

The formula (4) shows that in low-frequency field at $w = \ w_0$ (or the same for small bubbles with a size less resonant) the quantity z <0, therefore bubbles settle down in pressure crests. On the contrary, in high-frequency field at $\omega > \omega_0$ it is had $z > 0$ and then large bubbles are thrown out from fields with large pressure and they are agglomerated in pressure knots.

In the presence of phase transformations the pattern becomes complicated: even for vapor bubbles it is necessary to consider not only purely resonant properties, but also sharp increase of natural compressibility of the vapor bubbles related to a mass transfer which relative contribution sharply amplifies at the small sizes [4]. The similar effect should be considered and for crystallization centers (c.c.) in a crystallizing fluid in a sound field. For them the sharp increase of natural compressibility related to a mass transfer is observed. Here relative contribution of mass transfer is sharply amplified at the small sizes [4]. Using the theory presented in chapter 4 of the book [4], it is easy to make generalization on a case of any compressibility $\mathrm{K}(R,\omega)$. Partially it is shown in chapter 5 of the book [5] (generalization for non-linear parameter in chapter 8 see also). In this case quantity z has the complicated form depending on behaviour of a real part of a complex function $\mathrm{K}(R,\omega)$ of sound frequency and the size PI:

$$\zeta(R,\omega) = \frac{\rho' + 2\Delta\rho/3}{2\rho' + \rho} + \frac{1}{3}\Delta\rho c_1^{\ 2}\ \mathrm{Re}\left[\mathrm{K}(R,\omega)\left(1 - \frac{\beta_0}{\mathrm{K}}\frac{\rho'}{\Delta\rho}\right)\right] \tag{5}$$

Suppose $\mathrm{K} \cong \beta_0 \cong \beta'/\gamma'$, from the Eq. (5) it is possible to obtain Eq. (1). Guessing $\rho' < \rho$ it is obtained the simple expression for bubbles of any sizes (including vapor bubbles):

$$\zeta(R,\omega) = -(1/3)\rho c_1^{\ 2}\ \mathrm{Re}\left[\mathrm{K}(R,\omega)\right]. \tag{6}$$

In a case c.c. it is necessary to consider in addition an inequality $\Delta\rho < \rho$ then ζ will be equal [5]:

$$\zeta(R,\omega) = \frac{1}{3}\left\{1 + \Delta\rho c_1^{\ 2}\ \mathrm{Re}\left[\mathrm{K}(R,\omega)\left(1 - \frac{\beta_0}{\mathrm{K}}\frac{\rho'}{\Delta\rho}\right)\right]\right\}. \tag{7}$$

Thus, the sign quantity z essentially depends on compressibility $\mathrm{K}(R,\omega)$ which generally is a composite function of radius PI and frequencies of a sound field.

Additional compressibility in the presence of phase transformations. For the further analysis of localization PI it is necessary to analyze compressibility function. Most simply to understand the parent of the mechanism of additional compressibility in the presence of phase transformations it is possible in case of the crystallizing fluid containing c.c. Here it is important to consider competing magnification of volume of crystallization centers due to mass transfer (additional crystallization of a fluid on surface PI) and its obvious reduction at magnification of pressure due to pure mechanical compressibility. It appears that with reduction of the size PI when surface influence is more appreciably the mass transfer specified above starts to import also the increasing contribution to compressibility PI, and in which connection with an inverse sign. Features of compressibility taking into account phase transformations are presented by following equations:

$$K(R,\omega) = \beta_0 - \frac{C'_\sigma}{L}(\varphi - \Phi)d_\sigma\xi\ ,\ Q(R,\omega) = 1 - \frac{K}{3}\frac{\rho\omega^2R^2}{1 - k_1R} - \frac{G}{3}\left(\frac{2\sigma}{R} + 4i\eta\omega\right), \tag{8}$$

$$\varphi(k'_2R) = 3\frac{(k'_2R)\,\mathrm{cth}(k'_2R) - 1}{(k'_2R)^2},\qquad \Phi = 3\frac{\rho C_P}{\rho' L}\frac{1 - ik_2R}{(k_2R)^2},\qquad k_2 = \sqrt{i\omega/\chi}\ , \tag{9}$$

$$G = \frac{\beta'}{\gamma'} + \frac{C'_P}{L}\frac{\rho}{\Delta\rho}\Phi d_\sigma\xi\ ,\qquad \beta_0 = \frac{\beta'}{\gamma'} + \alpha'(d_S - d'_S)\frac{\varphi\Phi}{\varphi - \Phi} \tag{10}$$

$$x = (1 - iwt)^{1},\qquad \tau = \frac{C'_P}{L}\frac{R}{3\Lambda}(\varphi - \Phi),\qquad \xi = \xi(R,\omega) \Rightarrow \begin{cases} \underset{\Lambda\to 0}{\to} 0 \\ \underset{\Lambda\to\infty}{\to} 1 \end{cases} \tag{11}$$

Here C_P and C_s are heats capacity at constant pressure and along a curve of phase equilibrium, L is a heat of phase transformations (heat of crystallization or heat of vaporization), $d_\sigma = T\Delta\rho/\rho\rho' L$ and $d_S = aT / rC_P$ a declination of a curve of phase equilibrium and an adiabatic curve, accordingly,

$\alpha = -(1/\rho)(\partial\rho/\partial T)_P$ is an isobaric coefficient of thermal expansion, $\mathrm{K} = K/3Q$ is the generalized compressibility PI, $k_2 = \sqrt{iw/c}$ is a wave number of a thermal wave, c is a thermal diffusivity, L is a kinetic coefficient governing kinetics of phase transformations, for c.c. $\mathrm{L} = aLr/hT_s$, a is a constant of a crystalline lattice, h is a dynamic viscosity coefficient [4,5].

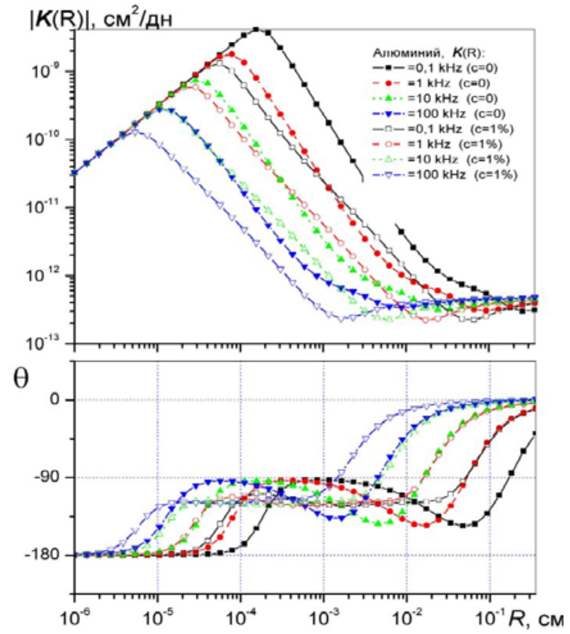

Fig. 1. Dependence of compressibility c.c. aluminium from radius for various sound frequencies and various concentration of an impurity (c=0 and c=1 %)

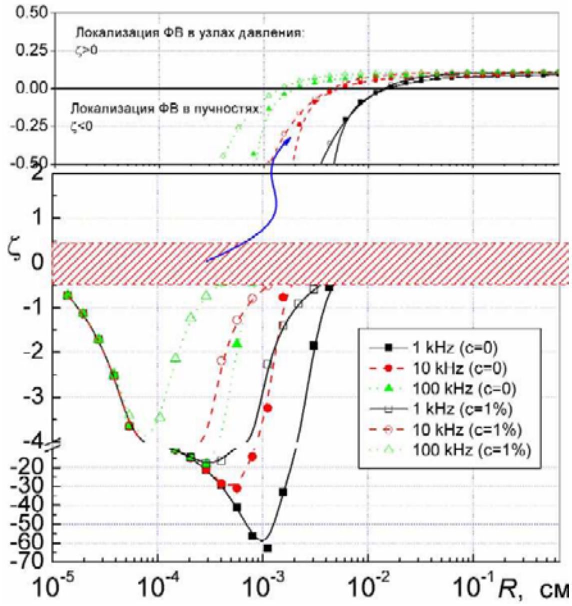

Fig. 2. Dependence of quantity ζ on radius for c.c. aluminium for various sound frequencies and various concentration of an impurity (c=0 and c=1 %)

At Fig. 1 the dependence of compressibility of a crystallization centre on radius is presented for various frequencies of a sound. One can see that as a whole at the small sizes there is the considerable amplification of volume pulsations of crystallization centers.

Localization PI in a standing wave. As with reduction of the size the compressibility c.c. sharply increases, it is possible to come to nontrivial deduction that at the small sizes c.c., despite its small mechanical compressibility, monopole oscillations turn to more essential. Especially it is essential owing to the known fact that near to a point of crystallization contrasts of density of a liquid and solid phase become small, i.e. $\Delta\rho/\rho = (\rho' - \rho)/\rho < 1$, and consequently the dipole component appointed space oscillations c.c. sharply decreases. So, in some range of the sizes the sign of the quantity z characterizing arrangement PI in the presence of phase transformations will be appointed by behaviour of function $\mathrm{K}(R,\omega)$. Especially it is necessary to pay attention to the range of R where the real part ofcompressibility K becomes negative – here quantity ζ also aspires to the negative value. In this case c.c. change a place of the usual arrangement in knots of a pressure wave and move in a pressure crest. Interval of radiuses and frequencies at which this phenomenon is observed it is possible to find out from Fig. 2. It is necessary to underline once again that solid particles without phase transformations always take up position in knots of a wave of pressure. The last is evidence of positivity of a real part of compressibility of such particles and thus always quantity $\zeta > 0$. It should be noted that location change c.c. in a sound field depending on their radius and frequency reminds behaviour of gas bubbles. However, the nature of these phenomena is various. In case of gas bubbles the parent of such behaviour is explained by resonant character of their oscillations, and in a case c.c. the main reason is the combination both magnification of compressibility module and also mechanisms of sign changing for a real part of compressibility due to mass transfer as a result of phase transformations. The important feature of dynamics c.c. under the influence of a sound is their sensitivity to concentration of foreign impurity in a melt. This circumstance is related to mass transfer depression on a surface c.c. at the expense of foreign impurity [4,5] and as a result – with a damping of monopole oscillations c.c. (see Fig. 1). The specified property can be applied to practical applications for the purpose of separation of the pure substance containing in c.c. from foreign particles which will be always localized in knots of pressure of an ultrasonic field. In turn essential dependence of localization place on the size c.c. can be taken as a base of guidance for decomposition of structure of solid crystallizing in an ultrasonic field.

Forces of interaction between PI. The considerable amplification of oscillations c.c. can give both increase of interaction forces and change of a sign of these forces. The most essential forces of interaction between various inclusions in a sound field are Bjerkness forces and Konig forces [1-3,4]. The first are related to a pulsation of inclusions, the last ones with dipole oscillations as whole. Bjerkness forces are characteristic in the core for squeezed inclusions , such as bubbles. In case of solid particles more considerable usually are Konig forces [1,2]. However in a case c.c. (for which $\Delta\rho << \rho$) their oscillations as whole relatively a fluid can be neglected. Therefore for c.c. unlike solid particles without phase transformations the basic forces of interaction are Bjerkness forces which it is possible to write by analogy with [4] in the form:

$$F = \frac{8\pi\rho}{r^2} R_1^{\,2} R_2^{\,2} \langle \dot{R}_1 \dot{R}_2 \rangle = \frac{4\pi\rho\omega^2 R_1^{\,3} R_2^{\,3}}{9r^2} \mathrm{Re}\left(\mathrm{K}_1 \mathrm{K}_2^{\,*}\right) P_m^{\,2} . \quad (12)$$

Here K_1 and K_2 are compressibility of two various PI. In case of identical PI Bjerkness force is equal:

$$F = 4\pi\rho\omega^2 R^6 |\mathrm{K}|^2 P_m^{\,2} / 9r^2 \quad (13)$$

From the given formulas and the effects presented on Fig. 1 it follows that Bjerkness forces for c.c. sharply increase in comparison with a case of solid particles without phase transformations. The greatest relative increase takes place at the sizes corresponding to a maximum of compressibility $\mathrm{K}(R)$. It is significant, that occurrence of the negative proper compressibility leads to occurrence of repulsion forces between PI of various sizes instead of usual attractive forces characteristic for PI without mass transfer. Incase of PI with identical sizes there will be attractive forces.

Conclusion. The solution of a problem on localization PI in a sound field in the presence of phase transformations is obtained. It is shown, that change of a localization place in ultrasonic wave depends on radius PI and frequency of a sound. Essential dependence of localization place on the size c.c. can be taken as a base of guidance for decomposition of structure of solid crystallizing in an ultrasonic field. It is shown, that forces of interaction c.c. (Bjerkness forces) sharply increase in comparison with a case of solid particles without phase transformations. It is necessary to pay attention that occurrence of the negative proper compressibility leads to occurrence of repulsion forces between PI of various sizes instead of usual attractive forces characteristic for PI without mass transfer.

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