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NATURE OF ACOUSTIC EMISSION DURING PHASE TRANSFORMATIONS AND ADEQUACY OF THE STEFAN CONDITION

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The kinetics of phase transformations of water and associated acoustic effects have been considered. The contribution of the energy of acoustic emission is estimated in relation to the latent energy of crystallization.

Problems of crystallization and melting lie at the junction of several fields of science. As is known, in the late 19th century Stefan laid the foundation for the methods of solution of these very complicated problems of mathematical physics, which have been developed intensely at present. Moreover, in recent decades some researchers have recorded acoustic emission during crystallization and melting [1, 2]. Because of this it is possible to suggest that the latent energy of crystallization can dissipate to a great extent (to 80%) by generation of acoustic waves [3].

It should be noted that crystal growth is a manifestly nonequilibrium process. In a system, passage from a nonequilibrium to an equilibrium state can proceed in different ways; however, waves are usually the most rapid mechanism of energy transfer. In view of this, the considerations expressed in [3] are formally reasonable. However, in this case, the heat evolved at the phase interface during crystallization of unit mass must be substantially lower than the specific heat of crystallization. In this case, in accordance with [3], the relative energy of acoustic emission can be neglected in the formulation of the boundary conditions for the Stefan problem only in the particular case of absolutely acoustically opaque media.

It should be noted that because of absorption of sound by matter, at a large distance from the phase interface the energy of acoustic emission will be transformed into thermal energy. Thus, when the relative amount of the energy of acoustic emission generated in crystallization processes is large, it is necessary to include such parameters as the sound absorption spectrum, the acoustic emission intensity, etc., which will make heat transfer problems extremely complicated.

It should be noted that formally similar problems arise when it is necessary to take account of heat transfer by IR radiation, which is always present along with ordinary heat conduction in various media. As is indicated in [4], for particular hydrocarbons the radiative contribution reaches 50%. In this connection it is stressed in [4] that experimental data on the heat conduction of many liquids can contain a noticeable contribution of the radiative component, which should be isolated, as a rule.

It should be noted that the authors of [3] suggest that an acoustic wave is induced by volume changes at the phase interface caused by the difference in the densities of the crystal and the melt. A similar problem was considered earlier in [5], where the appearance of the initial hydrodynamic section was assigned to this difference. Meanwhile, because of the smallness of the volume of a structural unit, even substantial changes in the volume, characteristic, for example, of ice, must only result in hypersonic oscillations, while acoustic emission recorded during crystallization and melting is found even in the region of sound frequencies.

In order to judge the adequacy of the Stefan condition, apart from experimental determination of the acoustic emission energy, it is necessary to proceed from particular considerations concerning the physical nature of the acoustic effects that develop during crystallization and melting of materials. This problem was solved to a first approximation in the study of the kinetics of phase transformations of water described in [6]. In order to

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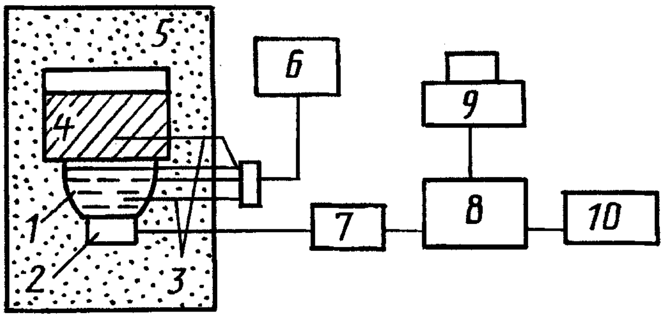

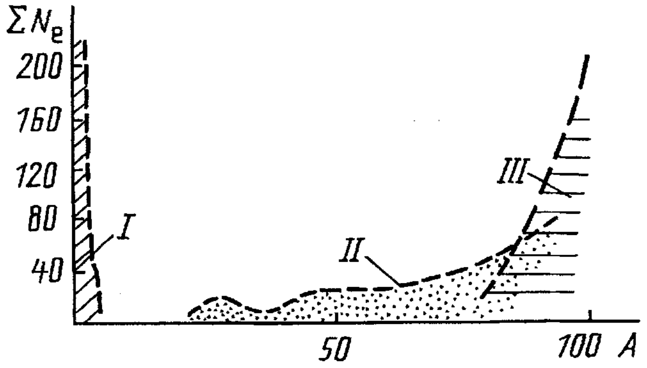

Fig. 1. Block diagram of experiments to investigate acoustic effects of crystallization and melting: 1) rubber cells with test materials; 2) piezosensor; 3) thermoresistors; 4) vessel with a cooling mixture; 5) heat insulation; 6) temperature indicator; 7) preamplifier; 8) AF-15 acoustic emission device; 9) PC; 10) plotter.

Fig. 2. Histogram of the amplitude distribution of signals in nucleation (I), growth (II), and melting (III) of ice: I) frequencies 0.02–0.2 MHz, II, III) 0.2–0.5 MHz. The amplitude is expressed in relative units.

analyze acoustic emission signals, a PC was included in the measuring system, because of which it was possible to carry out statistical processing of the data. A block diagram of the experiments carried out to investigate acoustic effects appearing in crystallization and melting is shown in Fig. 1.

In order to obtain information about the nature of the acoustic effects during nucleation, growth, and melting of ice, an analysis of the amplitude distribution of the signals was conducted, which was compared in some cases with visual monitoring. As an example, in Fig. 2 a combined hystogram of the amplitude distribution of events recorded during formation and melting of ice in the course of smooth (a 4^{o}C/h gradient) increase in the temperature above the surface of a specimen from -20^{o}C to 0^{o}C. The presence of three groups of signals apparently indicates different mechanisms responsible for a particular process of generation of waves in the course of crystallization and melting.

We now will suggest possible reasons for the generation of signals of all three groups. Signals of group I are recorded from the moment of appearance of individual crystals and, as a rule, are not found after the water surface is completely covered by an ice layer. This initial layer is formed by fine dendrite crystals, whose horizontal growth rate is known to depend on the subcooling temperature and to be several millimeters per second. With this in view, in this case the most probable reasons for the acoustic effects are collisions and fractures of the dendrites. These effects should be enhanced as the temperature gradient and the confinement of the space increase, which is confirmed experimentally [6].

Signals of group III are recorded upon a change in the sign of the temperature gradient in the solid phase and at positive temperatures. These signals are caused by generation and growth of microcracks under the action of temperature stresses. As one would expect, the activity of acoustic emission in melting increases with increase in the temperature gradient.

To explain the physical reasons for generation of signals of group II it is necessary to take into consideration that an increase in the volume of a polycrystalline body is always accompanied by generation of a field of internal stresses in it. This is facilitated by differences in the orientation of the axes of individual crystals, instability of the front, nonuniformity of heat fluxes into regions with a lower temperature, etc. As the thickness of the ice lens increases and its temperature falls, its rigidity increases, which leads to an increase in the density of collective defects and their elastic fields. With this in view, it is evident that acoustic emission due to crystallization may be treated as removal of the stress fields in the solid-phase mass. Moreover, because the boundary amplitude zones

TABLE 1. Energy of Acoustic Emission in the Frequency Range 0.2–0.5 MHz and Latent Energy of Crystallization of Water

t, min	$\dot{N}_0$	p, Pa	$E_a \cdot 10^{-2}$, J	Δh, cm	ΔV_1, cm^3	E_c, J	E_a/E_c, %
10	2408	16	3.08	0.038	0.266	79.8	0.078
20	2622	16	3.36	0.016	0.112	33.6	0.200
30	2605	21	4.38	0.012	0.084	25.2	0.348
40	2832	17	3.85	0.010	0.070	21.0	0.368
50	2983	19	4.53	0.009	0.063	18.9	0.478
60	3056	22	5.38	0.008	0.056	16.8	0.640
70	3050	21	5.12	0.007	0.049	14.7	0.696
80	3100	25	6.20	0.007	0.048	14.4	0.862

Note: In calculation of the actual ratio E_a/E_c, the numerator was doubled since the energy of acoustic emission determined in the experiments was found for a hemisphere.

of signals of groups II and III are joined together, it can be concluded that the growth of crystals is accompanied by generation and growth of microcracks, although the intensity of these processes is lower than in melting.

It should be noted that the reasons for generation of acoustic waves in crystallization given above do not contradict, in principle, the experimental conclusions [1] that the activity of emission grows with increase in the difference in the densities of the crystal and the melt and in the temperature gradient. It is evident that these factors lead to an increase in the spatial inhomogeneity of the growing crystal mass, which should be accompanied by more intense removal of stresses generated in the bulk of the solid phase.

In order to estimate the latent energy of crystallization the thickness of the growing layer of ice and its volume were calculated from the relation $h = \alpha\sqrt{t}$. The coefficient α was calculated after the experiment was completed and the actual thickness of the ice lens was measured. Since the parameters of the acoustic emission signals $\dot{N}_0$, $\dot{N}_e$, and A were recorded continuously, it was possible to calculate the acoustic emission energy for periods for which the corresponding increase in the volume of ice was determined. The acoustic emission energy was calculated from the expression $E_a = \rho_m V \dot{N}_0$. A sample of numerical data of one typical experiment is given in Table 1.

As can be seen from Table 1, as time passes, the growth of the ice layer and the release of the latent energy of crystallization decrease and the energy of acoustic emission increases. Nevertheless, its value relative to the latent energy of crystallization is very small. Measurements and corresponding calculations show that inclusion of components of acoustic energy gives only a slight addition to the obtained results in the entire kHz region.

Thus, the relative energy transferred by elastic waves in the kHz frequency range during crystallization is insignificant compared to the latent energy of phase transformations propagated by the thermal mechanism. Therefore, acoustic emission should not be included in the formulation of the boundary conditions for the Stefan problem.

NOTATION

h, thickness of the layer of ice; α, coefficient; t, time; $\dot{N}_0$, count rate of signal oscillations; $\dot{N}_e$, count rate of events; A, amplitude of the signals; E_a, energy of acoustic emission; ρ_m, sound pressurer in the middle hemispherical cross section of the cell; V, volume of the cell; p, sound pressure recorded by the sensor; V_1, volume of ice; E_c, latent energy of crystallization.

REFERENCES

1. S. N. Zadumkin, Kh. B. Khokonov, and Kh. B. Shokarov, Zh. Éksp. Teor. Fiz., **69**, No. 4, 1315-1319 (1975).
2. A. N. Smirnov and A. N. Dement'ev, Zh. Fiz. Khim., **59**, No. 7, 1791-1792 (1985).
3. O. G. Nalbandyan and S. T. Ovsepyan, in: Abstracts of Reports of the 2nd All-Union Conf. "Simulation of Crystal Growth," Riga (1987), Vol. 1, pp. 196-198.
4. N. B. Vargaftik, L. P. Filippov, A. A. Tarzimanov, and E. E. Totskii, Handbook of Thermal Conductivities of Liquids and Gases [in Russian], Moscow (1990).
5. R. Lodis and R. Parker, Growth of Single Crystals [Russian translation], Moscow (1974).
6. I. I. Sakharov, I. Yu. Golubev, I. V. Pavlov, and A. I. Potapov, Zh. Fiz. Khim., **66**, No. 2, 555-558 (1992).