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Abstract

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ON THE STUDY OF THE NONISOTHERM- MALITY OF THE FACES OF GROWING AND DISSOLVING ROCHELLE-SALT CRYSTALS GROWN FROM SOLUTIONS

(Presented by Academician A. V. Shubnikov, October 16, 1967)

Crystal growth is accompanied by the release of heat of crystallization, and this leads to heating of the near-surface layer of the growing crystal and of the nutrient solution. If the shape of the growing crystal is close to perfect, then under conditions of a static growth regime there is nonuniform release and dissipation of the heat of crystallization by its various parts (vertices, edges, and different regions of the faces), which is reflected in the temperature distribution over the surface of the growing face—nonisothermality of the surfaces of the crystal faces arises. The development of nonisothermality of the faces is influenced by the heat and concentration fluxes that arise around the growing or dissolving crystal, as well as by the shape of its faces.

A theoretical calculation of the growth of a cubic crystal from a supercooled stationary melt, carried out by G. P. Ivantsov (¹), showed that the surface of a growing crystal is nonisothermal; in this case the temperature at the centers of the faces should be higher than the temperature at the vertices. However, these propositions have not been confirmed experimentally.

Determination of the nonisothermality of the faces of growing crystals is important for establishing optimal regimes in the growth of single crystals; therefore we carried out experiments to determine the nonisothermality of the faces of growing and dissolving Rochelle-salt crystals grown from aqueous solutions. For these purposes an apparatus was constructed, consisting of a crystallizer with a thermoregulator and corresponding measuring equipment, making it possible to measure the temperature difference with an accuracy up to $\pm 0.038^\circ$. For the experiments we took finished Rochelle-salt crystals ranging in size from 5 to 50 mm, with well-developed faces c , p , a , and b (Fig. 1).

Measurements of nonisothermality were made on the best-developed faces c and p . The experiments were conducted at a constant temperature of 26° and at various supersaturations of the nutrient solution (-0.5 ; 0 ; $+1$; $+1.5$; $+2$; $+2.5$ and $+3^\circ$). The crystal under investigation, fixed in the holder of the measuring

Fig. 1. Rochelle-salt crystal

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apparatus, was immersed in the crystallizer with the nutrient solution. The face under investigation was positioned horizontally. The solution was not stirred. After one hour, measurement of the nonisothermality of the face was begun. The temperature difference was measured between the edge and points on the face along the y axis, at every millimeter from one edge of the face to the other. The entire process of measuring the nonisothermality of one face took from 3 to 5 min.

The data obtained made it possible to construct graphs characterizing the nonisothermality of the faces of growing and dissolving Rochelle-salt crystals. Figure 2 presents graphs of the nonisothermality of face c , obtained as a result of measurements at various supersaturations of the nutrient solution. Curve 1 shows the nonisothermality of face c during the process of dissolution of the crystal for the case of an undersaturated solution ($\Delta P =$

$= -0.5^\circ$). From this curve it is clear that the central part of the face has a lower temperature than at the edges. The temperature change here is smooth, without jumps. Curve 2 characterizes a supersaturation close to zero; it has a wavy character; apparently, this indicates an unstable crystallization regime; in the rising portions of the curve, a more intense deposition of substance probably occurs, and in the depressions—a lesser one.

Curves 3, 4, and 5 characterize the nonisothermality of the crystal face already in the process of its growth at supersaturations of the nutrient solution of $+1$, $+1.5$, and $+2^\circ$. These curves are characterized by smoothness and by an increase of the maxima with increasing supersaturation. Both these facts indicate that, with increasing supersaturation within the limits considered, the process proceeds calmly and, consequently, the crystal grows normally, with the greatest rate at $\Delta P = +2^\circ$.

Fig. 1. Rochelle-salt crystal

On curve 6, corresponding to a supersaturation $\Delta P = +2.5^\circ$, secondary maxima appeared which, judging from curves 3, 4, and 5, can hardly be attributed to measurement errors. Here, apparently, the excess supersaturation of the nutrient solution begins to play a role, leading to a new qualitative change in the growing crystal. This transition is most clearly expressed on curve 7, obtained at a supersaturation of $+3^\circ$. Here sharply expressed additional maxima and minima occur, although in principle the character of the curves remains the same as in the preceding ones. The presence of symmetrically arranged steps on curve 6 and of secondary maxima on curve 7 is possibly associated with the increased stepwise growth of the face, caused by the excess supersaturation of the nutrient solution.

Fig. 2. Graphs of nonisothermality of the c face of a Rochelle-salt crystal during its growth and dissolution.

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1—at a supersaturation of the nutrient solution $\Delta P = -0.5^\circ$; 2— $\Delta P \simeq 0^\circ$; 3— $\Delta P = +1^\circ$; 4— $\Delta P = +1.5^\circ$; 5— $\Delta P = +2^\circ$; 6— $\Delta P = +2.5^\circ$; 7— $\Delta P = +3^\circ$.

The nonisothermality graphs for the p face are analogous to the graphs for the c face, and therefore we shall not dwell on their discussion. We shall note only that at a supersaturation of $+3^\circ$ the curve behaves more calmly than for the c face at the same supersaturation. This is probably connected with the fact that the p face is less sensitive to supersaturations. In addition, the difference in the nonisothermality curves for the c and p faces is evidently connected with the anisotropy of the growth rate and of the thermal conductivity of the crystal.

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1. G. P. Ivantsov, in: *Growth of Crystals*, 1, Publishing House of the Academy of Sciences of the USSR, 1957.

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