Integrated studies of convection and ice layer formation during cooling of the bottom of a rectangular cavity

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Abstract. Experimental studies of the processes of crystallization of water as a liquid — simulator of melts with an inverse dependence of the density of melts on temperature — were carried out on two flat models. With the rapid cooling of the bottom of the cavity, substantial undercooling of the bottom layer of the liquid can be observed. The hydrodynamics in the process of growth of the ice layer at the bottom of the cavity was investigated. The effect of convective flow on the rate of crystallization and on the forms of the crystallization front has been studied. The evolution of the temperature field in the process of crystallization of water at the lower cooled boundary of the cavity is studied. It is shown that the evolution of temperature fields tracks the development stages of instability of the bottom layer of a liquid and the development of Rayleigh-Benard convection observed in a hydrodynamic experiment.

1. Introduction

Single crystals of various substances are the starting materials for the creation of a large number of modern technology and equipment in the fields of microelectronics, optoelectronics, nonlinear optics, etc. For example, cadmium-mercury telluride (CMT) to date occupy a leading position among the materials used to create instruments for recording infrared radiation, especially thermal imagers [1]. Among the methods for producing single crystals, the Bridgman method occupies an important place [2]. When implementing this method, the melt located in the ampoule or crucible and superheated relative to the crystallization temperature of the starting material is lowered at a given speed into the cold zone of the furnace. From the cooled bottom upwards after the nucleation of a crystal, the crystallization front propagates. Some melts have an inverse dependence of density on temperature. A similar dependence has a melt density of cadmium-mercury telluride [3, 4]. Among the many problems that need to be solved when creating the technology of growing high-quality and sufficiently large-sized single crystals of CMT in the mentioned works and many others, problems and features of hydrodynamics and heat exchange in the presence of inverse density versus temperature are not indicated. The solution of this question requires a deep understanding of the features of convective heat transfer in the presence of an inverse dependence of density on temperature and with various combinations of thermal conductivities of melts and crucible materials. This is what the research in this paper is directed to. Experimental studies of the effect of conjugate convective heat transfer on the rate of crystallization and on the forms of the crystallization fronts were carried out primarily on water, as a liquid — a melt simulator. Water, as is known, has an inverse dependence of density on temperature [5]. Above a flat crystallization front of melts with an inverse dependence of density on...
temperature, hydrodynamic and heat transfer features may arise, similar to those studied in this work. They can affect the local form of the crystallization front and the quality of the crystals.

2. Experimental stands and research results

The working section of the stand “Crystallizer 1” is a rectangular cavity with two copper parallel horizontal walls and two plexiglass parallel vertical walls (Fig. 1). Internal dimensions of the cavity filled with liquid: width × height × length = 30×105×105 mm³. The work area has optical quality plexiglass windows with a working part size of 105×105 mm² and allows video recording of the flow of the visualized liquid without optical distortion and allows studying the hydrodynamics and crystallization process on a quantitative level. Computer processing of video films allows to obtain quantitative data on the speed fields.

Stand “Crystallizer 2” has a similar design, but is designed to investigate crystallization processes using the methods of Hilbert optics and interferometry. Internal dimensions of the cavity filled with liquid: width × height × length = 30×84×136 mm³. In this case, the working area has optical-quality glass windows with the dimensions of the working section 84×136 mm².

Experimental studies of hydrodynamics during cooling of the lower boundary of a rectangular cavity were carried out at the “Crystallizer 1” installation, the circuit of the working section of which is shown in Figure 1. Here 1 is a fluid layer enclosed between two heat exchangers having copper walls that are maintained at specified temperatures by pumping thermostatic water in the cavity of the upper heat exchanger 3 and the coolant from the cryostat in the cavity of the lower heat exchanger 3. The flow of coolants through the nozzle is made from a thermostat a and cryostat so as to eliminate temperature gradients in the working walls. Water as a working fluid is visualized by polyamide tracer particles with a diameter of 20 microns. The flow of the visualized fluid is recorded by digital video cameras through transparent side walls (in the plane of Figure 1) when illuminated through transparent side walls with flat light streams through transparent end walls 5.

Figure 1. Diagram of the working section:
1 – liquid layer, 2 – copper walls,
3 – heat exchanger cavities, 4 – nozzles,
5 – transparent end walls of the cavity.

Figure 2 shows the temperature dependences of the temperature of the horizontal boundaries in the working section “Crystallizer 1”. The temperature of the coolant was set at –26 °C. After a prolonged holding of the working section at a temperature of +9°C by pumping thermostatted water through the upper heat exchanger, at the initial moment the cold heat carrier was pumped through the lower heat exchanger. Practically implemented mode of sudden cooling of the bottom of the cavity. In

Figure 2. Temperature dependences of the boundaries on time: 1 – on the upper wall of TW1; 2 – on the bottom wall of TW2. At the initial moment of time, a cryostat with a predetermined temperature value of −26°C is turned on.
the process of lowering the temperature of the lower boundary, the following features of hydrodynamics are observed.

Figures 3 and 4 show the fluid trajectories obtained by adding 200 video frames at those different times when the characteristic spatial flow pattern (PPT) is formed and when the transition to another PFT occurs as a result of lowering the temperature of the lower boundary.

In Figure 3a, in the central part of the downward flow of the fluid, and in the near-bottom region a stably stratified layer of fluid was formed. In Figure 3b, the bottom temperature is 0°C, the isotherm +4°C is located above the bottom at such a distance that low-intensity conveying of Rayleigh-Benard nature occurs in the bottom part. In Figure 3c, this type of convection already occupies a layer of greater thickness. Before the water starts to crystallize at the lower boundary, it is supercooled to −6.8°C (Figure 3d). The convective flow on this period of time has an irregular nonstationary character. The isotherm +4°C is on average above the bottom at a distance approximately equal to half

Figure 3. Fluid motion trajectories at different points in time.
The height of the fluid layer. At the time of the onset of crystallization, heat release of the phase transition occurs and a sharp rise in temperature, clearly visible in Figure 2 (curve 2).

After the onset of crystallization and with an increase in the thickness of the ice layer over the crystallization front, the Rayleigh-Benard nature convection takes on a more regular appearance. A cellular spatial flow pattern is formed. This is seen in Figure 4. In the upper part of the liquid layer, the flow becomes increasingly weak, as a layer of stably stratified fluid forms between the +4°C isotherm and the upper heated boundary, which is at a temperature of +9–8°C.

An idea of the characteristic values of the flow velocity is given in Figure 5.

In fig. 6-8 presents data on the shape of the crystallization front and the growth rate of the ice layer thickness. It can be seen that the shape of the FC depends on the direction of flow normal to the FC. In the upflow region, the local form is convex into a liquid. Thus, the inverse dependence of the density of water on temperature is affected.
Figure 7. The horizontal distribution of ice thickness at different times from the onset of crystallization: 1 – 600 s, 2 – 1960 s, 3 – 3330 s, 4 – 4620 s.

Figure 8. Dependence of the thickness of the ice layer on the time in the central section along the length of the working volume. Countdown from the onset of crystallization.

Figure 9. Fluid motion trajectories at different points in time in the monotonous mode of lowering the temperature of the lower boundary of the water layer.

Figure 9 shows steady-state spatial forms of the flow and the shape of the crystallization front obtained in the monotonous lowering of the temperature of the lower boundary with a low cooling rate. After the temperature in the cryostat was gradually lowered to a predetermined temperature level, the shutter speed at this level was followed for at least 1 hour. In this mode, the effect of deep supercooling of the near-wall layer of water near the bottom wall was not observed.

Figure 10 shows the interferograms obtained on the stand “Crystallizer 2”. Experimental studies of the evolution of temperature fields in a rectangular cavity in the modes of cooling the lower boundary of the cavity and the formation of the water crystallization front are carried out using the methods of interferometry and Hilbert optics. The optical measuring complex is implemented on the basis of the IAB-463M device with a modernized lighting module, filtering units for the optical signal and image recording. It is an optical system adapted to the task, described in [6, 7].
Figure 10 shows the evolution of the fields of isotherms in the monotonous mode of lowering the temperature of the lower boundary of the water layer before the onset of crystallization and during the growth of the ice layer. These data complement the information on the CFT, shown in Figures 3 and 4, with data on the evolution of temperature fields. Data obtained in cooling mode at a rate of 0.2 K / min. The main trends in the emergence and evolution of cellular flow, the transition to non-stationary flow regimes and the effect on instantaneous local forms of FCs will agree with those described above. The dark band in the lower part in Figure 10 corresponds to the solidified part of the substance.

\[ a - t = 00:26:07. \quad b - t = 00:27:53. \quad c - t = 00:34:14. \]

Figure 10. Isotherm fields, times are indicated from the beginning of the lower boundary.

Conclusion

Experimentally investigated the processes of crystallization of water in flat vertical layers with bottom cooling. The influence of thermogravitational convection and conjugate convective heat transfer on the rate of crystallization and on the forms of crystallization fronts is studied. Studies were carried out on stands with transparent working sections, which made it possible to observe and conduct digital video filming of spatial flow patterns in central sections of cavities in planes normal to the vertical walls and normal to the bottom. Digital video recording of the currents of the fluid visualized by tracer particles allowed to obtain data on the development of the spatial shape of the flow over time and velocity profiles in different sections along the layer height before the onset of crystallization and in the process of advancing the crystallization front. Using the stand created on the basis of the IAB-463M instrument, the evolution of the temperature field in the process of water crystallization at the lower cooled boundary of the cavity was studied in the interferometer mode. It is shown that the evolution of temperature fields tracks the development stages of instability of the bottom layer of a liquid and the development of Rayleigh-Benard convection observed in a hydrodynamic experiment.

Acknowledgments

This research was realized under the project III.18.2.5, number of state registration AAAA-A17-117022850021-3, and under RFBR project number 18-38-00790 мол_а.

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