Study of influence of volumetric radiating overcooling of a melt on the form of front of crystallization by means of numerical modeling processes of heat transfer at growth of sapphire crystals from the melt

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Abstract. It is lead numerical modeling of processes of heat exchange at growth of crystals of sapphire from a melt by Stepanov and Czochralski methods. The influences of volumetric radiating overcooling of a melt and of growth speed on the form of front of crystallization and distribution of temperature near to it is studied. It is shown, that the account of absorption of thermal radiation in a melt can lead to formation of volumetric supercooled areas near to crystallization front and to appreciable change of its shape.

1. Introduction
Last years there were many works on numerical modeling processes of growth of single crystals of the sapphire from a melt by various methods. It is connected with wide application of sapphire in modern techniques as optical windows and substrates of light-emitting diodes. Modern powerful program computing packages allow to model processes of growth and by that to improve technologies of growth, and quality of crystals and to raise productivity of process. For example for process of growth of sapphire by Stepanov method (EFG) in [1] by means of computing package COMSOL Multiphysics software numerical modeling the form of front of crystallization is lead at growth of rods and ribbons. In works [2-4] the form of front of crystallization and influence of various technology factors for process of growth of sapphire by Czochralski method were modeled. There are also many works on modeling process of growth of greater volumetric crystals of sapphire by Kyropolos method [5] and HEM method [6].

Now at numerical modeling processes of heat exchange at growth of semitransparent crystals from a melt for the account of influence of internal radiation in case of strongly absorbing melts Rosseland approach is widely used. Its essence is reduced to introduction of the additive of the effective thermal conductivity of participating medium to usual conductive component. However such approach at all we shall not apply in case of the decision of Stephan problems at the front crystallization. First, thus it is necessary to consider a melt opaque and to consider radiating losses of heat directly from the front of crystallization, and the opportunity of the account in model of volumetric overcooling a melt because of ablation of heat by radiation in a translucent crystal directly through front of crystallization, secondly, is cut. The correct account of these factors essentially influences the shape of front of crystallization, and in some cases can result in its instability. This phenomenon for the first time has been predicted in [7, 8].
Therefore the purpose of the present work was the study of influence of volumetric radiating overcooling of a melt on the shape of front of crystallization by means of numerical modeling processes of heat exchange at growth of crystals of sapphire from a melt by Stepanov and Czochralski methods.

2. Model description

The algorithm of the numerical decision has been developed for the decision of a problem of radiating carry of heat by a combination of a method of discrete ordinates and a method of trace of beams [9, 10]. Its advantages consist in two factors: 1) the method demands the moderate computing expenses both on time and on memory and practically does not suffer from numerical diffusion; 2) correctly works with diffuse and specular reflection boundaries, and it is capable to consider dispersion, absorption and emission of radiation by substance.

Calculations were spent for round cylindrical rods in diameter of $10\,\text{mm}$ with diffuse and smooth specular (Fresnel) reflection crystal-gas boundary. The absorption coefficient of a crystal was considered to constants on all its length equal to $17\,\text{m}^{-1}$ [11]. The absorption coefficient of thermal radiation of a melt varied from $200\,\text{m}^{-1}$ up to a case of its full opacity, but the majority of calculations has been made at value $1400\,\text{m}^{-1}$ [12]. Speeds of crystals growth varied from 30 up to $130\,\text{mm/h}$.

3. Result and discussion

3.1 Stepanov method (EFG)

In the present work the variant of Stepanov method from a flat top edge shaper is considered only. The threefold point is considered rigidly set on height $0.5\,\text{mm}$ above an edge of the shaper. The volumetric thermal emission in a resistive heater is defined as a result of the iterative decision of a full problem of heat exchange in quasi-stationary statement for maintenance of the set growth rate of a crystal. Thus also there is a search of the form of front of crystallization. Convection in volume of a melt it was neglected, as estimations show very small contribution of convectional stream of heat from a feeding capillary in comparison with other processes of heat exchange. The temperature at the front crystallization was assumed equal $T_{m}=2313\,\text{K}$.

Figure 1 shows the results of calculations of fields of temperatures of a melt and a crystal near to front of crystallization for a variant of the absorption coefficient of thermal radiation in a crystal – $a_{\text{crys}}=17\,\text{m}^{-1}$ and a melt - $a_{\text{melt}}=1400\,\text{m}^{-1}$ are resulted at growth rates of a crystal 30 (Figure1a), and $120\,\text{mm/h}$ (Figure1b). On Figure1c the result of calculation for speed of $120\,\text{mm/h}$ in case of an opaque melt is showed. These calculations have been executed in the assumptions diffuse reflection lateral surface of a crystal, as well as in works [1, 13]. On figure 1d calculation is made in the assumption of specular reflecting surface of a crystal. Comparison of the calculations executed for diffuse and specular reflecting lateral surfaces of a crystal, shows, that in the latter case instability of front is displaced to an axis of a crystal and leads to local rise of a meniscus of a melt. Such phenomenon is sometimes observed of crystals when there is a periodic capture of super cooled melt to the subsequent crystallization and formation of large cavities.

Figure 2 shows the distribution of temperature in a crystal and a melt along growth axis for different settlement variants. It is visible, that without taking into account absorption in a melt overcooling in a meniscus of a melt is absent. At the account of absorption in a melt there is an overcooling which grows from 0.6 up to 7K with increase in rate of growth. As a result at rate of $120\,\text{mm/h}$ there is the instability of front of crystallization. The reason of instability is in fact that the density of conductive flux of heat allocated from front of crystallization in a melt starts to be compared or even to exceed density of a similar flux in a crystal.

This instability [7,8] is caused by ablation of heat from volume of a melt by radiation and to possible decrease in temperature below temperature of crystallization. Such instability arising at high rate of crystallization, can lead to formation of cellular front and formation of gas inclusions in volume of a crystal. This phenomenon is often observed in practice at growth of crystals of sapphire [12].
Figure 1. Temperature field and the form of crystallization front at the growth of sapphire crystal by Stepanov method in diameter 10mm. Vertical scale is twice more than horizontal. Height of a threefold point above the top edge of the shaper is 0.5mm. a-Growth rate is $V=30\text{mm/h}$, b-$V=120\text{mm/h}$, c-$V=120\text{mm/h}$, calculation per the assumption of an opaque melt, d-$V=120\text{mm/h}$, calculation in the assumption of specular reflection of a stream of radiation from a lateral surface of a crystal. a, b, c- calculation for diffuse reflection of a crystal-gas boundaries.

Figure 2. Temperature distribution of in a melt and a crystal near to front of crystallization along an axis of growth by Stepanov method: 1 and 2 - for an opaque melt, growth rate $V=30$ and $120\text{mm/h}$ accordingly, 3,4 and 5 in view of the absorption coefficient in a melt $1400\text{m}^{-1}$, $V = 30, 60$ and $120\text{mm/h}$ accordingly, 6 - calculation in the assumption of specular reflection of radiation from a surface of a crystal-gas, $V=120\text{mm/h}$. Variants 1-5 for a case of diffuse reflection of a light stream.
3.2 Czochralski method
Similar calculations of a temperature field and the form of front of crystallization at the same values of factors of absorption have been lead for a case of growth of crystals of sapphire by Czochralski method in the same thermal zone. In this case a hydrodynamics of a melt was calculated too. In calculations it was supposed, that the lateral surface of a crystal is of specular reflection.

Figure 3 shows results of calculations of front of crystallization and a field of speeds of current of a melt are shown at growth of a crystal of sapphire with a speed of 80 mm/h at the account of absorption in a melt (figure 3a) and without taking into account (Figure 3b). At figure 4 distribution of temperature in a melt and a crystal for these cases is shown.

One can see that the front of crystallization is strongly curved in a melt and for a case of an opaque melt has characteristic bends near to a threefold point. A direction and speeds of a stream of a melt for two cases are similar, that is the melt flows from edges of a crucible along a surface of a melt and goes downwards, bending around front of crystallization. Such behaviour of a melt and the form of front of crystallization is characteristic for this method of growth of crystals of sapphire. It proves to be true calculations and experiments at growth of greater crystals of sapphire, 100 mm in diameter [3].

The effect of radiation supercooling for an opaque melt is absent as well as for growing by the Stepanov method. At the account of absorption in a melt calculations show occurrence of supercooled area under front of crystallization which increases with growth of growth rate.

4. Conclusions
As a result of numerical modelling it is shown, that the account of big, but final value of factor of absorption of thermal radiation in a melt leads to formation near to front of crystallization volumetric areas of supercooled melt and to appreciable change of the form of front. At greater growth rates and enough long crystal it can lead to instability of front of crystallization that proves to be true experimental supervision.

![Figure 3](image)

**Figure 3.** Temperature distribution (left side) and velocity field (right side) at the growth of a sapphire crystal of 10mm in diameter by Czochralski method with a rate 80mm/h: a - the absorption coefficient of a melt of 1400m⁻¹, b - case an opaque melt.
Figure 4. Temperature distribution of in a melt and a crystal near the front of crystallization along an axis of growth by Czochralski method: 1-for an opaque melt, growth rate of 80 mm/h, 2 and 3 in view of the absorption coefficient in a melt (1400 m$^{-1}$), growth rate 80 and 130 mm/h accordingly.

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