Study of instability of sapphire tubes growth by Stepanov method

M G Vasil'ev, S I Bakholdin and V M Krymov
Ioffe Institute, Politekhnicheskaya 26, St. Petersburg, 194021, Russia
E-mail: vasiliev.mg@yandex.ru

Abstract. The paper considers the experimentally observed instability of capillary shaping during the growth of thick-walled sapphire tubes by the Stepanov method. The explanation of this phenomenon is based on the theoretical model of radiative-conductive heat transfer in a crystal. An algorithm is developed for the asymptotic expansion of the problem based on the presence of two small parameters. It is shown that the spatial density of the radiation is inhomogeneous along the cross section of the tube and is maximum near its inner walls. This leads to their overheating and the meniscus separation from the inner edges of the shaper.

Experimental observations of the shaped sapphire crystals growth show that, under certain growth conditions, there is a loss of stability of the crystallization front. In the case of growing tubular profiles, the inner meniscus is torn off from the edge of the former and the shape of the tube is disturbed (see Figure 1). These phenomena are observed for thick-walled tubes grown from a shaper with a flat upper surface. In addition, this occurs during strong overheating of the melt and at a crystal length comparable with the height of the heat shields. The tubes growth with a wall thickness of less than 2 millimeters occurs without problems.

Figure 1. The heater temperature increases along the length of the grown sapphire tube with a cross section of 9.5 x 5 mm. The overheating areas correspond to the places where the inner meniscus is torn off from the edge of the shaper.
In the present work, the heat transfer problem in refractory semi-transparent crystals grown from a melt is solved by the asymptotic method. It is shown that the effect of separation of the inner meniscus from the edge of the shaper can be explained by the more intensive absorption of external hot radiation in areas near the inner surface of the tube. With an increase in the length of the grown crystal, it is necessary to increase the external radiant intensity from the heater, since the radiation heat flux through the crystal from the crystallization front increases.

The asymptotic expansion algorithm contains not one, but two small parameters: - the conductive-radiation parameter

\[ N = k \lambda / \left( 4 n_e^2 \sigma T_0^3 \right) \ll 1 \] (1)

and the parameter of the characteristic optical thickness of the crystal cross section:

\[ \tau_d = k d << 1. \]

Here \( k \) is the absorption coefficient of thermal radiation, \( d \) is the characteristic transverse dimension, \( \lambda \) is the thermal conductivity, \( n_e \) is the refractive index, \( \sigma \) is the Stefan-Boltzmann constant, and \( T_0 \) is the melting temperature in \( K \).

For an optically thick medium, the ratio of conductive flux to radiation is 0.75N, and for an optically thin medium:

\[ \frac{q_{\text{cond}}}{q_{\text{rad}}} = \frac{2}{\tau^2} N, \quad \tau << 1 \] (2)

where \( \tau \) is the optical thickness. For sapphire at temperatures close to the melting temperature, the parameter \( N = 0.01 \).

The radiative-conductive heat transfer (RCT) problem will be solved by the asymptotic method under the following assumptions:

1) the absorption coefficient is considered constant for most of the thermal radiation spectrum,
2) the conductive heat transfer of the tube walls with gas will be neglected compared to the radiation heat flux in the opaque region of the spectrum,
3) the ratios \( \bar{R}_2/d \) and \( \bar{R}_1/d \) will be considered constant as \( d \to 0 \) (\( \epsilon_0 = k d \to 0 \)), where \( \bar{R}_2, \bar{R}_1 \) is the external and internal radius of the tube, and \( d \) is equal to \( d = \bar{R}_2 - \bar{R}_1 \),
4) the inner and outer lateral cylindrical surfaces of the crystal are assumed to be smooth and the mechanism of reflection of rays will be considered specular (according to Frenel).

As a result, after the transition to dimensionless variables with

\[ r = \bar{r} / \bar{d}, r_1 = \bar{R}_1 / \bar{d} = \text{const}, r_2 = \bar{R}_2 / \bar{d} = \text{const}, (r_2 - r_1 = 1), \quad z = k \bar{z}, \]
\[ t = T / T_0, t_e = T_e / T_0, t_i = T_i / T_0, t_H = T_H / T_0, \]

RCT problem is converted to the form:

\[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial t}{\partial r} \right) + \tau_d^2 \frac{\partial^2 t}{\partial z^2} = \frac{(\tau_d)^2}{N} \left[ t^4 \Delta_p(t) - G \left( t, t_e^{-1} \right) \right] \] (4)
\[ \frac{\partial t}{\partial r} \bigg|_{r=r_i} = Bi \cdot \left( t^4 \delta_p(t) - t^4 \delta_p(t_0) \right) - \frac{\partial t}{\partial r} \bigg|_{r=r_2} = Bi \cdot \left( t^4 \delta_p(t) - t^4 \delta_p(t_0) \right) \]

\[ Bi = \frac{\tau_d}{N} B_0, \quad B_0 = \frac{\beta \delta_{p0} (T_0)}{4n_e^2}, \quad Bi = Bi_m \left( \tau_d \right)^m, \quad m = 0, 1, \ldots \]

\[ t \big|_{k=0} = 1, \quad t \big|_{k=\tau_d} = t_H. \]

Here \( t \) is the dimensionless temperature, equal to 1 at the crystallization front;

\( T_e, T_i \) are the effective temperatures of thermal radiation coming from outside to the boundary \( r = r_2 \) and \( r = r_i \), respectively;

\( T_H \) is the temperature at the upper end of the tube;

\( \delta_{p0} (T), \Delta_p (t) \) is the fraction of the energy of Planck thermal radiation at a temperature \( T = tT_0 \) for the opaque and semitransparent absorption region of the crystal, respectively (\( \Delta_p (t) = 1 - \delta_{p0} (tT_0) \));

\( G \) is the dimensionless spatial density of the radiation;

\( Bi \) is the Bio number, which may be of the order of smallness \( Bi = O(\left( \tau_d \right)^m) \), where \( m \) is an integer greater than zero;

\[ \delta_p (t) = \delta_{p0} (tT_0) / \delta_{p0} (T_0); \]

\( \beta \) is the coefficient of blackness of the side surface of the crystal.

In equalities (3), where \( r_i \) and \( r_2 \) are assumed to be constant when tending \( \tau_d \) to zero (this actually happens, when \( d \to 0 \)), the condition for constructing the asymptotics is actually contained. If we assume \( R_2 = const \) and tend \( d \) to zero, then we will have a different picture corresponding to the case of an infinitely wide plate. The asymptotic solution for \( \tau_d = kd << N \) and \( \tau_d \) of order \( N \) (similar to the case of opaque media [2]) can be shown that the temperature field in these cases is almost one-dimensional and depends only on the distance from the crystallization front. This approach was used earlier and was called the light-guide model approximation [3-4], which made it possible to reduce the solution of three-dimensional problems to a one-dimensional formulation with a specular (according to Frenel) reflecting side surface of the crystal.

But when \( \tau_d = O(\sqrt{N}) \) the contribution of radiative heat transfer becomes comparable with the conductive component (in the transverse direction with respect to the direction of crystal growth), this approach is not applicable. In this case, for the zero term of the asymptotic expansion of the solution of the problem \( u_0 \) in the external approximation (for \( z >> \tau_d \) and \( (\tau_H - \tau) / \tau_d >> 1 \) ) we obtain the problem:
where it is impossible in the general case to find a one-dimensional solution depending only on the variable \( z \), even for \( u_0 \). Here \( G_0^{\text{ext}} \) is the expansion term of the order \( O(1) \) of the spatial radiation density, which consists of the sum of the light-guide component \( G_{\text{LG}}^{0,\text{ext}} \) and the non-light-guide component \( G_{\text{NLG}}^{0,\text{ext}} \). Light guide rays are called rays that are completely reflected from the side surface of the crystal. In [5], an analytical expression is given in the case of a continuous circular cylinder for the solid angle of the region of non-light guide rays in the form:

\[
\Omega_{\text{NLG}}(r/R, \Theta_B) = 8 \int_0^\Phi(r/R, \Theta_B) \frac{d\varphi}{1 - (r/R)^2 \sin^2 \varphi}, \tag{8}
\]

\[
\Phi(r/R, \Theta_B) = \arcsin \left[ \min \left( 1, \frac{\sin \Theta_B}{r/R} \right) \right].
\]

Here \( r, R \) is the current radius value for the considered section point and the outer radius of the cylinder, respectively; \( \Theta_B \) - angle of total internal reflection; \( \varphi \) - the angle in the spherical coordinate system associated with the point in question and with the \( Z' \) axis parallel to the axis of symmetry.

Figure 2 shows a graph of function (8), which shows its largest values near the central part of the crystal cross section. Heat losses due to its outflow from the central part by radiation to the outside are 3 times higher for sapphire compared with the areas adjacent to the outer side surface.

In the case of a tubular crystal, the same picture will remain. First, the region of non-light guide rays emerging from the volume of the crystal through the inner side surface will be smaller. Secondly, the rays passing through the boundary \( r = r_1 \) will again fall back into the tube volume, without losing their energy, if there is nothing but gas inside. In this case, it can be written in the same form as for a solid cylinder:

**Figure 2.** Distribution of the total solid angle fraction per region of non-light guide rays over the cross section of a straight circular cylinder
The expression for the spatial density of the light guide region of the rays has the form

$$G_{NLG}^{0,\text{ext}}(r, z, t) = \frac{1}{4\pi} \int d\Omega I(r, z, \theta, u_0)$$

where \( I(r, z, \theta, u_0) \) is the dimensionless intensity of radiation coming from the tube volume \( d \) to the point under consideration, in the temperature field \( u_0 \); \( \Omega_{NLG}(r, r_1, r_2) \) - region of light guide rays. For a one-dimensional temperature field, which depends only on the \( z \) coordinate, \( G_{NLG}^{0,\text{ext}} \) has a simple form, which can be found in [3] for the case of the light-guide approximation.

Thus, the spatial density of radiation coming from outside is inhomogeneous along the tube’s cross section and is maximum near its inner walls. This leads to their overheating and the separation of the meniscus from the inner edges of the former. Estimates for sapphire give rise to such an inhomogeneity, starting from the tube wall thickness \( d \) of the order of 5 mm (\( k = 20 \text{m}^{-1} \) is the absorption coefficient of sapphire thermal radiation at a temperature close to the melting temperature), which is consistent with experimental results.

References
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