ISSN 1062-8738, Bulletin of the Russian Academy of Sciences: Physics, 2009, Vol. 73, No. 10, pp. 1320–1323. © Allerton Press, Inc., 2009.
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Analysis of the Profile Curves of the Menisci for the Crystal Growth by the Edge-Defined Film-Fed Growth (Stepanov) Technique

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Abstract—The behavior of the profile curves of the melt menisci for the growth of sapphire tubes of different diameters by the edge-defined film-fed growth (Stepanov) technique has been investigated. The external and internal circular menisci of the crystal tube are considered. The case of positive contact angle contact between the profile curve and working shaper surface is analyzed.

DOI: 10.3103/S1062873809100037

INTRODUCTION

Currently, urgent problems in improving the growth technique of profiled sapphire crystals are the expansion of the range of crystal transverse sizes (both to larger and smaller values) and growth of higher quality single crystals (for example, for optical applications).

In this study we numerically investigated the behavior of the profile curves of melt menisci during growth of sapphire tubes of different diameter. One has to know the characteristics of the profile curves of menisci to apply them in automatic systems with weight sensor control.

The effect of the meniscus shape on crystallization has been analyzed in many studies [1–7]. Here, we investigate the menisci with a positive inclination angle of the tangent to the profile curve, θ_d. The right part of the vertical cross section of the crystal rod and melt meniscus is considered. In this case, θ_d is defined as the angle of between the tangent to the profile curve meniscus and the horizontal at the point of contact between the meniscus and shaper edge. Such angles are possible for shaper skew working surfaces, which are used to control the crystallization front shape in order to optimize the defect distribution in the growing crystal and its mosaicity [8]. The minimum skew angle of the shaper working surface at which meniscus with a positive angle θ_d can be formed is determined. The analysis of the profile curves of menisci with a positive angle θ_d is new in comparison with the known studies.

The profile curves of the melt menisci were investigated based on the numerical solution of the Laplace capillary equation [9] with different parameters and boundary conditions, corresponding to the meniscus catching at the shaper edge:

$$z''r + z'(1 + z'^2) \pm 2(H_d - z)(1 + z'^2)^{3/2} r = 0, \qquad (1)$$

where z is the meniscus current height, r is the meniscus current radius, and H_d is the external pressure caused by the difference in the melt levels in the crucible and at the meniscus base. For the sapphire growth by the edge-defined film-fed growth (EFG) technique, the melt surface in the crucible is generally below the meniscus base; i.e., $H_d < 0$. Therefore, we consider below negative or close-to-zero ($H_d \leq 0$) external pressures. Equation (1) is written in the dimensionless form and normalized to the capillary constant. Hereinafter, all the values obtained by solving this equation are also dimensionless and normalized to this constant.

The boundary conditions for Eq. (1) can be set differently. We mainly used the following boundary conditions:

$$z(R_d) = 0, \quad -\arctan z'(r_c) = \pi/2 - \varepsilon, \qquad (2)$$

where r_c is the crystal radius; R_d is the radius of the shaper working surface; and ε is the growth angle, which is approximately 13° for sapphire [5, 10].

Thus, zero meniscus height is set on the shaper working surface, and the condition related to the growth angle constancy is set at the meniscus end contacting with the crystal edge.

When solving the Laplace equation, the numerical values for the boundary conditions (crystal and shaper radii, external pressure, growth angle) were taken close to real, based on the experimental data for profiled crystal growth.

We analyzed the dependences of the meniscus profile curves on the following factors: crystal radius, external pressure, contact angle θ_d between the meniscus and shaper edge, and the radii of crystal and shaper separated by a constant gap.

The dependences of the meniscus profile curves on the angle θ_d and the radii of crystal and shaper separated by a constant gap has been found for the first time.

EXTERNAL CIRCULAR MENISCI

Dependence of the Meniscus Profile Curves on Crystal Radius

Figure 1a shows typical profile curves of external circular menisci, calculated for different values of crystal tube external radius and positive angle θ_d.

The calculation results indicate that, for the angle θ_d varied approximately from $-\pi/2$ to $+\pi/2$, two solutions to the Laplace equation (1) satisfy boundary conditions (2): one for negative angles θ_d and the other for positive angles.

Realization of a particular version depends on the shaper type: the angle θ_d will be negative for horizontal working edges, whereas for sharply tilted edges the formation of menisci with positive angles θ_d is more likely.

Dependence of the Meniscus Profile Curves on the External Pressure

It can be seen in Fig. 1b that an increase in the external pressure from $H_d = -8$ to $H_d = -2$ increases in the height of the meniscus profile curve and reduces the curvature. The reason is that a reduction of the external pressure in magnitude diminishes the pressure of the curved melt surface, which balances the external one. In this context, the force pressing the meniscus against the shaper working surface decreases.

The closer the external pressure magnitude to zero, the more rapidly the height of the meniscus profile curve increases.

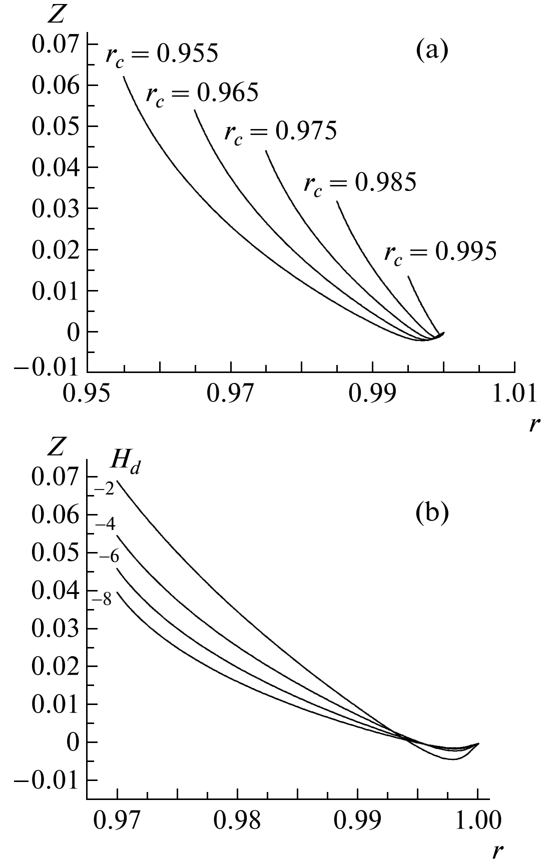

Fig. 1. Profile curves of external circular menisci obtained with a change in the (a) crystal radius and (b) external pressure.

Dependence of the Meniscus Profile Curve on the Contact Angle θ_d between the Meniscus with Shaper Edge

The range of variation in the angle θ_d must be estimated to calculate the program value of the growing crystal mass; this calculation is performed during automatic weight sensor control.

For fixed radii of crystal and working shaper edges, we calculated the profile curves at different angles θ_d. It was not necessary to match the meniscus profile curve with the condition of growth angle constancy near the crystal edge. The calculation results are shown in Figs. 2a and 2b.

The data in Fig. 2b make it possible to determine the range of real angles θ_d. Indeed, taking into account that the growth angles for different materials are in the range of 0°–20°, which corresponds to the change in θ_c from –90° to –70°, one can see in Fig. 2b that this range of variation in θ_c (for specified simulation parameters) corresponds to the following approximate ranges of variation in θ_d: from –50° to –45° and from 45° to 50° for negative and positive angles θ_d, respectively.

Knowing the range of variation in θ_d and using the plot in Fig. 2a, one can find the range of meniscus heights h_m. The values of θ_d and meniscus height h_m are used in the program observation equation for automatic growth control.

In addition, to calculate the program mass, one has to know the sign of θ_d. As was mentioned above, this angle is negative for conventional (non-skew) shapers, whereas menisci with positive angle are more likely to be formed on skew shapers, provided that the shaper working surface is skewed at an angle exceeding θ_d in magnitude.

Dependence of the Meniscus Profile Curve on the Radii of Crystal and Shaper Separated by a Constant Gap

We performed calculations changing simultaneously the crystal radius r_c and the radius R_d of shaper working edges.

As can be seen in Fig. 2c, the meniscus height changes mainly with a change in the crystal radius (and, correspondingly, the shaper radius) within one capillary constant. With a further increase in the crys-

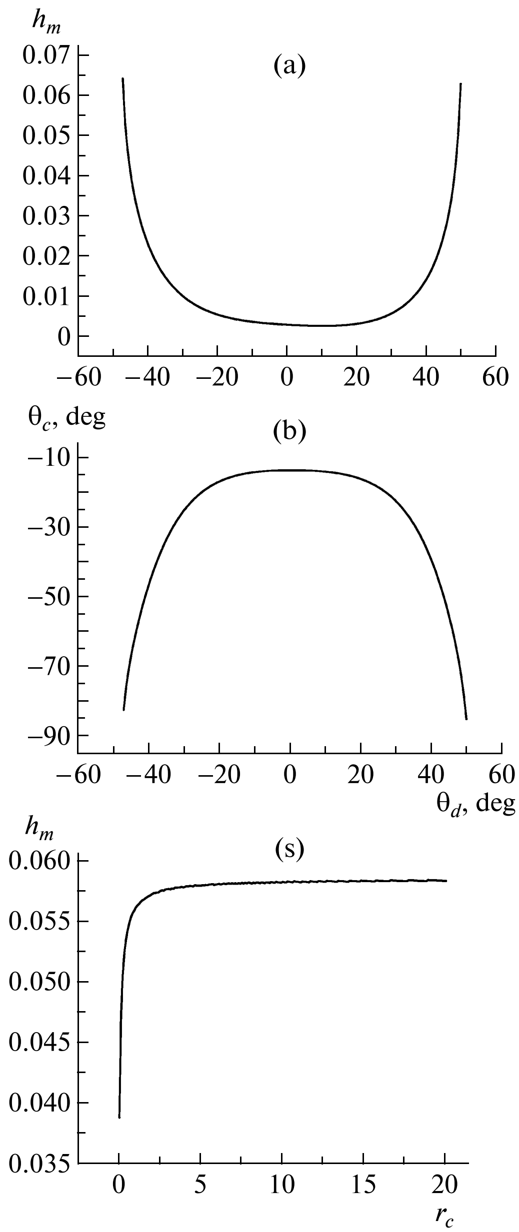

Fig. 2. (a, b) Dependences of the meniscus (a) height h_m and (b) angle θ_c at the triple point on the contact angle θ_d between the meniscus and shaper edge and (c) the dependence of the meniscus height on the crystal radius at an invariable difference between the shaper and crystal radii (for external circular menisci).

tal and shaper radii, the meniscus height barely changes.

The angle θ_d, as well as the meniscus height, changes significantly with a change in the crystal radius (and, correspondingly, the shaper radius) within one capillary constant. The angle θ_d increases in magnitude with an increase in the crystal and shaper radii. This is related to the rise in meniscus height, which requires a sharper rise of the profile curve near the shaper working edges and, accordingly, a larger magnitude of this angle.

The curve in Fig. 2c shows clearly the difference in the meniscus behavior in the cases of "large" and "small" crystal radii. The radius value that is approximately equal to the capillary constant separates the regions characterized by radically different behavior of the dependences of the meniscus height and shaper angle.

INTERNAL CIRCULAR MENISCI

The behavior of internal circular menisci was analyzed for the same cases as for external circular menisci.

The profile curves calculated for internal menisci are generally similar to those for external circular menisci (Fig. 1). The reason for the small differences is that the azimuthal curvature (the second term in Eq. (1)), entering the Laplace equation analyzed, is positive for the internal meniscus, whereas for the external meniscus this curvature is negative. Due to this, he profile curves of internal menisci are located slightly above those of external menisci for the same radii (provided that the other simulation parameters are the same).

Calculations were also performed for fixed crystal and shaper radii and external pressure at different angles θ_d(without matching with the condition of growth angle constancy near the crystal edge). The dependences of the main meniscius characteristics are also similar to the corresponding dependences in Figs. 2a and 2b for external circular menisci.

For internal circular menisci we also performed calculations with a simultaneous change in the crystal radius r_c and the radius R_d of working shaper edges (with a constant difference between these radii).

It was found that the height of internal menisci weakly decreases, whereas that of external menisci increases.

The reason is that the azimuthal curvature has different signs in the cases under consideration. For small internal menisci this curvature is fairly high and its positive additive to the meniscus height is significant. Then, with an increase in the crystal and shaper radii, the effect of azimuthal curvature decreases, due to which the height of internal menisci decreases.

The main change in the angle θ_d and the internal meniscus height corresponds basically to the change in the crystal and shaper radii within one capillary constant. The change in the angle θ_d for internal menisci is insignificant in comparison with that for external

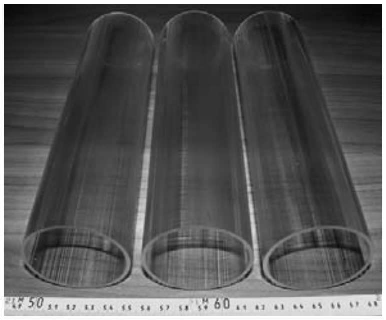

Fig. 3. Sapphire tubes with an external diameter of 55 mm, grown by the EFG automatic technique.

menisci. In this case, the angle magnitude slightly decreases, whereas for external menisci it increases.

CONCLUSIONS

Based on the numerical solution of the Laplace capillary equation, we analyzed the shape of the profile curves and their characteristics at a positive angle θ_d, which are characteristic of skew shaper working surfaces.

It is found that there are two solutions to the Laplace equation for stationary growth: with positive and negative contact angles θ_d between the meniscus and shaper.

Differences in the heights of external and internal tube menisci are revealed, all other factors being equal. These differences are due to the fact that the azimuthal curvature for the external and internal menisci has different signs and, correspondingly, differently affects the meniscus height in a particular case.

It is established that the heights of external and internal menisci behave oppositely, depending on the simultaneous change in the radii of crystal and shaper separated by a constant gap. This effect is also explained to a great extent by the different signs of the azimuthal curvature of external and internal menisci.

It is found that the height of menisci with different signs of shaper edge catching angles similarly (nonlinearly) depends on external pressure.

We plotted the dependences of the meniscus height and profile curve angles near the crystal edge on the angle θ_d, which do not require matching menisci near the crystal edge with the condition of growth angle constancy. For the simulation parameters of cylindrical tube growth under consideration, the range of angles θ_d corresponding to real shaping conditions is found, according to which the magnitude of the real angle θ_d can vary from 45° to 50°. This angle must be set in the automatic growth control to determine the program mass determined by the weight sensor.

An example of application of meniscus profile curve calculations in the automatic weight sensor control is the growth of large sapphire tubes with an external diameter of 55 mm (Fig. 3).

REFERENCES

1. Tsivinskii, S.V., *Inzh. Fiz. Zh.,* 1962, vol. 5, no. 9, p. 59.
2. Tatarchenko, V.A., *Ustoichivyi rost kristallov* (Stable Crystal Growth), Moscow: Nauka, 1988.
3. Shchelkin, Yu.F., *Fiz. Khim. Obrab. Mater.,* 1971, vol. 3, p. 29.
4. Surek, T., Chalmers, B., and Mlavsky, A.I., *J. Cryst. Growth*, 1977, vol. 42, p. 453.
5. Kuandykov, L.L. and Antonov, P.I., *J. Cryst. Growth*, 2001, vol. 232, p. 852.
6. Rossolenko, S.N., *J. Cryst. Growth*, 2001, vol. 231, p. 306.
7. Rossolenko, S.N. and Zhdanov, A.V., *J. Cryst. Growth*, 1990, vol. 104, p. 8.
8. Perov, V.F., Papkov, V.S., and Ivanov, I.A., *Izv. Akad. Nauk SSSR, Ser. Fiz.*, 1979, vol. 43, no. 9, p. 1977.
9. Landau, L.D. and Lifshits, E.M., *Mekhanika sploshnykh sred* (Mechanics of Continuous Media), Moscow: Gostekhteorizdat, 1953.
10. Satunkin, G.A., *J. Cryst. Growth*, 2003, vol. 255, p. 170.