Application of the liquid bridges theory to find the melt menisci shape when growing cylindrical crystals

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Abstract. A vertical liquid bridge of small volume between a conical shaper and a convex crystallization front was investigated. Two variants of the front crystallization shape selection are considered: a conical and a spherical fronts. A variational statement of the original problem is given. As the boundary conditions we used the condition of engagement at the edge of the shaper and a given growth angle at the crystallization front. The Bond number was assumed to be small, and to find a solution of the problem the asymptotic approach was applied. The calculations are carried out for small diameter cylindrical sapphire crystals, grown from the melt by the Stepanov method. The results of the menisci shapes calculations are presented. The comparison of the results of calculations for conical and spherical crystallization fronts is carried out.

1. Introduction

In recent decades, the theory of liquid bridges between different surfaces has attracted the attention of researchers. One of the possible practical applications of this theory is the study of the profile curves of the menisci of cylindrical crystals, grown from the melt by the Stepanov method [1]. Asymptotics of the shape of the melt menisci for single crystals grown by this method were investigated in [2] for the case of a flat crystallization front and a flat shaper. However, in practice, the crystallization front is not flat and its shape can be approximated by a spherical surface. Moreover, as shown in [3], the use of not a flat, but a conical shaper is preferable when growing sapphire crystals. In [4], an algorithm is proposed for solving the problem of the shape of a vertical liquid bridge between arbitrary solid convex surfaces under the assumption that the Bond number is small.

In this paper, a particular case of a liquid bridge is considered - a vertical liquid bridge of a small volume between a conical shaper and a convex crystallization front. Two variants of the front crystallization shape selection are considered: a conical and a spherical fronts. An engagement condition at the edge of the shaper and a given growth angle at the crystallization front are used as boundary conditions. It is assumed that the Bond number is a small parameter of the problem, and the use of the asymptotic approach is justified. The results of calculations of the menisci shape of small-diameter sapphire crystals grown from melt by the Stepanov method are presented.
It should be noted that the algorithm proposed in this work is suitable for several more technological processes, namely: zone melting of silicon and growing oxides from a pedestal when heated by laser and light beams [5].

2. Formulation of the problem

Let us consider the case of a vertical liquid bridge located between two solid surfaces: the bottom and the lid. The bottom is the shaper and the lid is the crystallization front (figure 1). In view of the axial symmetry, we will solve the problem in a cylindrical coordinate system \((r, z)\). Let the surface tensions between media are \(\alpha_{13}, \alpha_{14}, \alpha_{34}, \alpha_{23}, \alpha_{24}\), respectively. Contact area of the liquid bridge with the shaper \(z = f_1(r) = \tan(\gamma_1) r\) (bottom) - circle of radius \(r_1 = \pi - 2\gamma_1\) is the cone angle), and with the crystallization front \(z = f_2(r) = \hat{h} + f_2(r) (f_2(0) = 0)\) (lid) - circle of radius \(r_2\), and we will assume that \(r_2 < r_1\). Consider two cases: front - a spherical surface of radius \(R_0 (f_2(r) = R_0 - \sqrt{R_0^2 - r^2})\), or - a conical surface with cone angle \(\pi - 2\gamma_2\) \((f_2(r) = \tan(\gamma_2) r)\). The volume of the liquid bridge is assumed to be fixed: \(I\{u(r)\} = V\). We introduce into consideration the functional \(J\{u(r)\}\), which includes the surface energy and the gravity force energy. Therefore, we obtain an isoperimetric problem: find the minimum of the functional \(J\{u(r)\}\) under the condition that the functional \(I\{u(r)\}\) takes the constant value \(V\). In accordance with Euler’s theorem, we introduce an extended functional \(J\{u(r)\} + \lambda I\{u(r)\}\), where \(\lambda\) is the Lagrange multiplier. Let us turn to the dimensionless form of writing the task using the following scaling:

\[
\begin{align*}
\xi &= r/V^{1/3}, \quad w(\xi) = u(r)/V^{1/3}, \quad i = 1, 2, \\
\varphi_1(\xi) &= f_1(r)/V^{1/3}, \quad \varphi_2(\xi) = \hat{h}/V^{1/3} + f_2(r)/V^{1/3} = h + \varphi_2(\xi), \quad \mu = \nu V^{1/3}/\alpha_{34}.
\end{align*}
\]

Dimensionless constant \(B = g\rho V^{2/3}/\alpha_{34}\) is the Bond number. Here \(g\) is the gravity acceleration, \(\rho\) is the fluid density.
2.1. Equations and boundary conditions

Euler’s equation for the extended functional in new variables will have the form

$$\frac{d}{d\xi} \left( \int \frac{\xi w'(\xi)}{\sqrt{1 + (w'(\xi))^2}} \right) = B\xi (w(\xi) - \phi_1(\xi)) + \mu\xi, \xi_2 < \xi < \xi_1; \quad (1)$$

The boundary conditions of the problem can be written in the following form:

- the engagement condition at the edge of the shaper is
  $$w(\xi_1) = \phi_1(\xi_1), \quad (2)$$
  and we believe that the radius of the shaper $\xi_1$ is a given value;
- the contact condition of the liquid bridge with the crystallization front is
  $$w(\xi_2) = h + \phi_2(\xi_2); \quad (3)$$
  the growth angle condition is
  $$w'(\xi_2) = -\cot(\varepsilon_0), \quad (4)$$
  then $\theta_2 = \pi/2 + \varepsilon_0 - \arctan(\phi_2'(\xi_2))$.
  And finally, the condition of the volume conservation is
  $$\int_{\xi_2}^{\xi_1} (w(\xi) - \phi_1(\xi)) \xi d\xi + \int_{\xi_0}^{\xi_2} (\phi_2(\xi) - \phi_1(\xi)) \xi d\xi = \frac{1}{2\pi}. \quad (5)$$

So, we have four unknown quantities, namely: $\mu$, $\xi_2$, two integration constants, and four conditions (2)-(5).

3. Algorithm for solving the problem

To construct an efficient algorithm for solving problem (1)-(5), we renormalize of all quantities by $\xi_2$ as follows: a new independent variable $\eta = \xi/\xi_2$; new sought function $v(\eta) = w(\xi)/\xi_2$; modified given functions $\psi_1(\eta) = \phi_1(\xi)/\xi_2$, $\psi_2(\eta) = h/\xi_2 + \phi_2(\xi)/\xi_2 = H + \psi_2(\eta)$; modified Bond number $b = B(\xi_2)^2$; modified Lagrange multiplier $M = \mu \xi_2$.

As a result, equation (1) takes the form

$$\frac{d}{d\eta} \left( \int \frac{\eta v'(\eta)}{\sqrt{1 + (v'(\eta))^2}} \right) = b(v(\eta) - \psi_1(\eta)) + M\eta, \quad 1 < \eta < \eta_1 = \xi_1/\xi_2. \quad (6)$$

Integrating this equation taking into account condition (4), we obtain

$$\frac{v'(\eta)}{\sqrt{1 + (v'(\eta))^2}} = \frac{1}{\eta} \left( 0.5M\eta^2 - 1 - \cos(\varepsilon_0) + b \int_{\eta_1}^{\eta} [v(s) - \psi_1(s)] ds \right) \equiv \Phi(\eta). \quad (7)$$

The auxiliary function $\Phi(\eta)$ must satisfy the conditions: $-1 < \Phi(\eta) < 0$, $\Phi[1, \eta_1], \Phi(1) = -\cos(\varepsilon_0)$.

Resolving equation (7) with respect to $v'(\eta)$ and integrating the resulting equation taking into account the boundary condition (3), which in new variables has the form $v(1) = H + \psi_2(1)$, we get the following expression for the sought function $v(\eta)$:

$$v(\eta) = \int_{1}^{\eta} \frac{\Phi(s) ds}{\sqrt{1 - (\Phi(s))^2}} + H + \psi_2(1). \quad (8)$$
Boundary condition (2), which in new variables has the form $v(\eta_1) = \psi_1(\eta_1)$, gives

$$H = \psi_1(\eta_1) - \psi_2(\eta_2) - \int_1^{\eta_1} \frac{\Phi(s)}{\sqrt{1 - (\Phi(s))^2}} ds.$$  \hspace{1cm} (9)

Writing down the volume conservation condition (5) in new variables and integrating by parts taking into account the boundary conditions, we obtain the following equation for finding the sought value $\eta_1$:

$$\int_1^{\eta_1} \frac{\Phi(\eta)\eta^2 d\eta}{\sqrt{1 - (\Phi(\eta))^2}} - \int_0^{\eta_1} \psi'_1(\eta)\eta^2 d\eta + \int_0^1 \psi'_2(\eta)\eta^2 d\eta = -\frac{1}{\pi(\xi_1)^3} (\eta_1)^3.$$  \hspace{1cm} (10)

Thus, we have been obtained all the relations necessary for solving the problem by the iteration method. Let’s organize an iterative process by the small parameter $b$.

3.1. First iteration (construction of a zeroth approximation)

At the first iteration, we believe that $B = 0$, therefore, $b = 0$ (we construct an approximate solution in zero gravity conditions). Let us take the value of the parameter $M$ from the range of admissible values (thereby determining the function $\Phi(\eta)$). The admissible values of the parameter $M$ are that its values for which the equation (10) have solution. Using the bisection method, we find an approximate solution of the nonlinear equation (10) - the value $\eta_1$, and since $\eta_1$ is a given value, then and the value $\eta_2$. Knowing $M$ and $\eta_2$, we find $\mu$. Further, from equation (9) we find $H$, and hence $h$. The meniscus profile is given by formula (8). If we want to restrict ourselves to the zero approximation, then we take a different value of the parameter $M$ and perform the same actions. We pass with some selected step the entire admissible values range of $M$ (it is bounded both above and below). As a result, we obtain the dependence of the the liquid bridge height $h$ (distance from the shaper to the crystallization front at $\xi = 0$) on the parameter $M$ and find a solution corresponding to the given value of $h$. At this point, the process of obtaining a zero approximation of the problem solution for a given value of the height of the liquid bridge ends.

3.2. Second and subsequent iterations

If we want to find the next approximation, then we continue the calculations process for the parameter value $M$ selected at the first iteration. Namely, we find the value of the parameter $b$. In the expression for the auxiliary function $\Phi(\eta)$, we take into account the term proportional to $b$, moreover the function $v(\eta)$ take from the previous iteration. Further we carry out the actions described above and we take the next step of the iterative process. And so on, until the difference in the values of $\eta_1$ at two successive iterations becomes less than the value, which ensures the given accuracy of the calculations. And so we do for each selected value of the parameter $M$ from the range of admissible values. We construct the dependence of the liquid bridge height on the parameter $M$ and we find the problem solution for the given height value.

4. Calculation of the shape meniscus of a sapphire single crystal

Let us apply the algorithm suggested above for calculating the meniscus shape of a cylindrical sapphire single crystal grown from the melt by the Stepanov method. The cone angle of the shaper is $180^\circ - 2\gamma_1, \gamma_1 = 30^\circ$. The calculations were performed for two variants of the crystallization front shape: 1) spherical front with the radius is $R_0 = 3 \text{ mm}$ and 2) conical front with the cone angle is $180^\circ - 2\gamma_2, \gamma_2 = 25^\circ$. The shaper radius is $r_1 = 3 \text{ mm}$. The sapphire growth angle $\varepsilon_0 = 13^\circ$ [6]. Calculations were carried out for the Bond number equal
to 0.2 and we were been perform the first and second iterations. We are present the results of calculations after the first iteration, since the differences in the solutions are small and the graphs of the profiles practically merge.

Figure 2 gives the profiles of sapphire melt menisci for two variants of the crystallization front shape. It is shown how the profile shape changes when changing of the liquid bridge height. It is seen that these changes are more pronounced for a spherical front. At a some value of the liquid bridge height, the sign of the meniscus profile curvature changes: the meniscus becomes convex from a concave one. Figure 3 also serves for compare two possible variants of the crystallization front shape. It contains dependencies $r_2(h)$ and $\theta_1(h)$ for the spherical and conical crystallization fronts. In the case of a conical front, the radius of the growing crystal ($r_2$) approaches the radius of the shaper ($r_1 = 3$ mm) at higher the liquid bridge height than in the case of a spherical front, which is confirmed by the growing sapphire single crystals practice. In addition, the angle of contact at the point of engagement of the melt over the edge of the shaper (angle $\theta_1$) in this case changes more and with a decrease in the height of the liquid bridge it becomes greater than 90 degrees, which is also confirmed by the practice of growing.

![Figure 2](image)

**Figure 2.** Meniscus profiles. The case (a) - a spherical crystallization front and three values of the liquid bridge height: 1 - $h = 0.84$, 2 - $h = 0.78$, 3 - $h = 0.76$ mm; (b) - a conical front and 1 - $h = 0.92$, 2 - $h = 0.78$, 3 - $h = 0.65$ mm.

5. Conclusions

A study of a vertical liquid bridge of small volume between a conical shaper and a convex crystallization front has been carried out. Two possible variants of the front crystallization shape are considered: a conical front and a spherical front. The condition of engagement at the edge of the shaper and a given growth angle at the crystallization front were used as the boundary conditions. A variational formulation of the melt meniscus shape problem is given. An algorithm for finding its approximate solution is constructed under the assumption that the Bond number is small.

The calculations have been performed for small-diameter cylindrical single crystals of sapphire grown from the melt by the Stepanov method. The results of the menisci shapes calculations for conical and spherical crystallization fronts are presented. The dependences of the growing crystal radius and the angle of contact at the engagement point of the melt over the edge of the shaper from the height of the liquid bridge are investigated. The comparison of the results of calculations for two forms of fronts is carried out. It are determined that the conical shape of the crystallization front is in better agreement with the growing practice.
Figure 3. The dependences of the radius of the growing crystal $r_2$ (a) and of the contact angle of the melt with the shaper $\theta_1$ (b) from the height of the liquid bridge height for spherical (1) and conical (2) crystallization fronts.

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