Modeling Czochralski growth of oxide crystals for piezoelectric and optical applications

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Abstract. Numerical modeling is applied to investigate the impact of crystal and crucible rotation on the flow pattern and crystal-melt interface shape in Czochralski growth of oxide semi-transparent crystals used for piezoelectric and optical applications. Two cases are simulated in the present work: the growth of piezoelectric langatate (LGT) crystals of 3 cm in diameter in an inductive furnace, and the growth of sapphire crystals of 10 cm in diameter in a resistive configuration. The numerical results indicate that the interface shape depends essentially on the internal radiative heat exchanges in the semi-transparent crystals. Computations performed by applying crystal/crucible rotation show that the interface can be flattened during LGT growth, while flat-interface growth of large diameter sapphire crystals may not be possible.

1. Introduction

Czochralski (Cz) technique is used for the growth of a variety of oxide materials such as Al₂O₃ (sapphire), Y₃Al₅O₁₂ (YAG), LiNbO₃, Bi₄Ge₃O₁₂ and La₃Ga₅.₅Taₐ.₅O₁₄ (LGT). Among these oxides, LGT and his homologous compounds, are of interest as raw materials for piezoelectric applications. Sapphire has also many applications, being used for lasers, optical windows or as substrate for Light Emitting Diodes (LED) devices.

The Czochralski method has been well developed in time for the growth of oxide crystals [1]. However, the quality of crystals is still affected by defects as dislocations, inclusions, cracks and facets. Numerical simulation has become an important tool to investigate Cz growth of semi-transparent oxide crystals [2-8]. In their work [2], Xiao and Derby studied the effect of the internal radiation in Cz growth of yttrium aluminium garnet (YAG) and gadolinium gallium garnet (GGG). They found that the conical shape of the crystal-melt interface depends on the internal radiant transfer through the crystal. Other papers investigated the heat and mass transport phenomena at different stages of the Cz growth process [3, 4]. It is generally recognized that the deeply convex shape of the interface in these oxide systems, generate high thermal stress in the crystal, which may promote cracking, facets and dislocations [5, 6]. Crystal and crucible rotation can be applied to flatten the interface and reduce thermal stresses [2, 7, 8]. Numerical computations carried out in [7, 8] have shown that the interface can be flattened in Cz sapphire growth by applying crystal/crucible counter rotation. However, no experimental validation of these numerical results has been provided. It should be pointed that in practice, only crystal rotation is used in Czochralski growth of oxides.

The objective of the present work is to investigate the impact of crystal and crucible rotation on the flow pattern and interface shape in Cz growth of LGT and sapphire crystals.
2. Model description
Numerical modeling is performed by using the finite element code COMSOL Multiphysics. Heat transfer and momentum equations are solved for a 2D-axisymmetric furnace configuration, where all the furnace components are included. The internal radiative heat transfer in semi-transparent LGT and sapphire crystals is simulated by using P1-approximation model. The laminar flow computations include buoyancy, Marangoni and forced convection due to crystal and crucible rotation. Simulations are performed at a given stage of the crystallization process. A model which uses the deformable mesh technique to compute the shape of the crystal-melt interface, has been implemented in COMSOL Multiphysics. Transient computations with deformed mesh are conducted by using initial conditions for temperatures and flow velocities carried out from steady-state simulations.

3. Numerical results
The modeling is performed for two different Czochralski configurations. LGT crystals of 3 cm in diameter are grown in an inductive heated furnace (see Fig. 1). The electromagnetic field produced by a RF coil induces eddy currents in an iridium crucible (6 cm in diameter) which is heated by Joule’s effect. Figure 1 shows the temperature field (at the left side), streamlines and flow velocity (at the right side) computed at the middle stage of the growth process. The solidification isotherm is shown (in blue colour) on the figure. High temperatures, above the melting point are obtained only in the melt and a small region around the crucible. The radial temperature gradient at the free surface of the melt, which drives the Marangoni convection, is \( G_{Tr} = 200 \text{K/cm} \).

![Figure 1. Temperature field (left side), streamlines and velocity field (right side) computed in an inductive heated Czochralski configuration used to grow LGT crystals of 3 cm in diameter.](image-url)
Figure 2 shows a resistive heated Czochralski configuration used to grow sapphire crystals of 10 cm in diameter. The input power in the heater is much higher as compared to the previous case. Numerical computations show a more extended area of high temperatures in the furnace (see the left side of the Fig. 2). It is found that the resistive heating ensures small temperature gradients in the melt. The radial temperature gradient at the free surface of the melt is \( G_{Tr} = 28 \text{K/cm} \).

![Temperature field and streamlines](image)

**Figure 2.** Temperature field (left side), streamlines and velocity field (right side) computed in a resistive heated Czochralski configuration used to grow sapphire crystals of 10 cm in diameter.

### 3.1. Internal radiative effect

The effect of the radiative heat exchanges in the semi-transparent LGT crystal is analysed in Fig. 3. Figure 3a shows the results carried out from a computation which takes into account the internal radiative heat exchanges in the crystal. The radiation is absorbed and emitted by the semi-transparent LGT ingot, leading to a significant increase of the effective thermal conductivity of the crystal. The huge difference between the thermal conductivities in the solid and liquid phases is responsible for the deeply convex shape of the interface. The computed interface deflection \( f \approx 2.2 \text{cm} \) was validated by comparison to experimental measurements in our previous work [9]. The flow pattern is characterized by one main vortex which flows upwards along the crucible wall and downwards at the sample centreline. The interface becomes almost flat in the computation performed for an opaque crystal (Fig. 3b). The radial temperature difference in the crystal at the proximity of the triple solid-liquid-gas point is much higher in the case which accounts for the internal radiative effect: \( \Delta T = 110 \text{K (semi-transparent crystal)} \) and \( \Delta T = 15 \text{K (opaque crystal)} \).
3.2. Effect of crystal and crucible rotation

The effect of crystal and crucible rotation in Cz growth of piezoelectric LGT crystals of 3 cm in diameter was analysed in our previous work [9]. Numerical computations have shown that the interface can be flattened by applying only crystal rotation at high rates ($\omega_{\text{crystal}} = 45 - 50 \text{ rpm}$), due to reversed flow at the sample centreline underneath the crystal. Applying crystal/crucible counter rotation in this configuration, leads to a distorted shape of the interface.

The effect of the forced convection in the case of sapphire crystals (10 cm in diameter) is analysed in Fig. 4. Computations are performed for a crystal length of $L = 4 \text{ cm}$ (conical part of the ingot). The case computed without rotation shows that the liquid downwards at the sample centre, below the crystal (Fig. 4a). The computed interface deflection ($f \approx 5.3 \text{ cm}$) is in agreement with the experimental result in [10]. Applying only crystal rotation at rates greater than a critical value ($\omega_{\text{crystal}} = 20 \text{ rpm}$), changes the flow direction at the sample centreline. The upward flow located below the crystal has a flattening effect on the growth interface. However, this effect becomes important starting with rotation rates of $\omega_{\text{crystal}} = 30 \text{ rpm}$ (Fig. 4b). In this case, the interface curvature decreases, but the convection becomes unstable, having a deleterious effect on the interface shape, which is slightly distorted. If the rotation rate is increased above 30rpm, the interface is more distorted by the flow. Figure 4c shows the case computed with crystal and crucible counter rotation: $\omega_{\text{crystal}} = 30 \text{ rpm}$, $\omega_{\text{crucible}} = -30 \text{ rpm}$. Applying crucible rotation intensifies the downward flow below the crystal, and opposes the beneficial effect of the crystal rotation, so the interface remains curved.

Figure 3. Temperature field and isotherms (left side), streamlines and velocity field (right side) computed for Cz growth of LGT: (a) semi-transparent crystal; (b) opaque crystal.
Figure 4. Temperature field and isotherms (left side), streamlines and velocity field (right side) computed with crystal/crucible rotation in the case of sapphire growth: (a) $\omega_{\text{crystal}} = 0, \omega_{\text{crucible}} = 0$; (b) $\omega_{\text{crystal}} = 30 \text{ rpm}, \omega_{\text{crucible}} = 0$; (c) $\omega_{\text{crystal}} = 30 \text{ rpm}, \omega_{\text{crucible}} = -30 \text{ rpm}$. 
4. Conclusion
Previous numerical analysis of crystal/crucible rotation effect in Czochralski growth of small diameter (3 cm) piezoelectric LGT crystals have shown that the interface can be flattened by applying only crystal rotation at high rates $\omega_{\text{crystal}} = 45 – 50$ rpm [9]. The present computations show that the growth interface cannot be flattened in the case of large size sapphire crystals (10 cm in diameter). Applying only crystal rotation at high rates results in a distorted shape of the interface, while crystal/crucible counter rotation has no flattening effect, due to the opposite directions of the induced flows. These results are in agreement with experimental Cz growth of sapphire crystals, which generally uses only crystal rotation at low rates [6].

5. References
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