Validation of mathematical model for CZ process using small-scale laboratory crystal growth furnace

Kristaps Bergfelds, Andrejs Sabanskis and Janis Virbulis
Faculty of Physics and Mathematics, University of Latvia, Zellu Street 23, LV-1002 Riga, Latvia
E-mail: kristaps.bergfelds@lu.lv

Abstract. The present material is focused on the modelling of small-scale laboratory NaCl-RbCl crystal growth furnace. First steps towards fully transient simulations are taken in the form of stationary simulations that deal with the optimization of material properties to match the model to experimental conditions. For this purpose, simulation software primarily used for the modelling of industrial-scale silicon crystal growth process was successfully applied. Finally, transient simulations of the crystal growth are presented, giving a sufficient agreement to experimental results.

1. Introduction
Semiconductor microelectronics is a large market with sales reaching 335 billion dollars [1]. This market is based on single-crystal (especially silicon) wafer production, where the first technologically complex process is the single crystal growth. Additionally, 40% of solar panels are manufactured from monocrystalline silicon (Si) [2]. Si single crystals are mostly produced by utilizing the Czochralski (CZ) and floating zone (FZ) methods [3]. Continuous crystal growth process development is necessary mostly because of the need to decrease the size of microelectronic circuitry elements [4] and increase the wafer diameter [5]. This leads to demand for larger crystals with higher quality. Larger amounts of polycrystalline Si and larger crucibles used for each crystal growth process causes large financial losses due to any faults during this process and makes large scale industrial experiments extremely expensive. Therefore, mathematical modelling is a key instrument used for the development of the crystal growth processes.

Mathematical modelling makes it possible to obtain information about important physical parameters relevant to the growth process without performing costly experiments. The authors’ research group at the University of Latvia (UL) Faculty of Physics and Mathematics has developed such mathematical modelling tools for the simulation of the CZ crystal growth process [6]. However, it is necessary to perform validation of the mathematical model to ensure its relation to an actual crystal growth process by using practical experiments. As mentioned before, industrial scale experiments are too expensive to be considered as routine tool for mathematical model development. In addition to large-scale experiments, small laboratory scale research techniques must be established to perform cost-effective validation of the developed mathematical methods.
2. Problem definition
Authors of this article have access to small-scale laboratory CZ crystal growth furnace (figures 1 and 2) which can be used for the mathematical model verification described previously. This furnace consists of an electrically heated crucible unit (see schematics in figure 3) and a crystal puller. Crucible contains about 150 ml of NaCl-RbCl salt mixture with molar ratio of 40:60. This composition has eutectic properties which allow for relatively low melting point around 815 K [7, 8]. Optimal growth conditions are achieved at pulling speeds of 0.6 mm/min (see illustrative photography in figure 1). Crystals can be grown with diameter around 20 mm (see example in figure 2). After performing experimental crystal growth shown in figure 2, it is possible to study the application of the available mathematical modelling techniques for the simulation of this experiment — which is the main topic of this article.

![Figure 1. Photography of the NaCl-RbCl salt crystal growth process.](image1)

![Figure 2. Example of a grown crystal.](image2)

Although the crystal growth performed with this material first seems as considerably different from the industrial CZ-Si growth processes (size, temperature range, presence of atmosphere, translucency), system still exhibits all the physical phenomena that can be related to silicon crystal growth and does not have the drawbacks associated with some liquid metals e.g. gallium and its eutectics. Namely, the crystal growth can be performed in room conditions and does not require a shielding liquid or a pressure vessel to protect it from air. Additionally, the temperature field structure in our furnace is similar to industrial CZ process, contrary to gallium which melts at 30°C and might require cooling — instead of heating — to be used in crystal growth experiments.

3. Calibration of model parameters
3.1. Overview of the mathematical model and creation of geometry
For the studies performed in this article, previously developed simulation software “CZ-Trans” was used [6]. It can perform axisymmetric non-stationary simulations of CZ crystal growth process. The following physical phenomena are taken into account: (1) heat transfer by conduction, (2) heat transfer by thermal radiation (view factor approach as in [9]), (3) shape of the melt free surface, (4) shape of crystallization interface and (5) shape of crystal side surface.

To perform simulations with the software, axisymmetric geometry was created by simplifying various system parts to speed up simulations and conform to the limitations of the model (figure 4). As it can be seen, ceramic parts are unified into a single domain and separate
Figure 3. Precise geometry of the crucible-heater unit used in the laboratory crystal growth furnace (dimensions in mm).

Figure 4. Simplified geometry for the creation of FEM domains used in numerical simulations.

windings of the heater wire are modelled by a single FEM block. These simplifications are required to correctly model the radiation dominating heat transfer between these components.

3.2. Data from experimental heating of crucible as means for model calibration
During the experiment, before the crystal growth shown in figure 2 could be performed, crucible was heated up using its full electrical power of 475 W until all its contents were melted and stationary temperature field was achieved. This situation was characterized by the following temperature measurements using a pyrometer: 450 ± 10 K on the outer steel casing and 850 ± 20 K on the crucible edge, just besides melt free surface. This information can be used to calibrate the parameters used in the software to achieve reasonably realistic temperature distribution for numerical simulations.

Physical parameters for the melt [7] and corundum ceramic parts [10] are well known (see figure 4 for summary). However, the thermal conductivity of the silica insulation wool used in the construction of the crucible is not known. This parameter ($\lambda_{\text{ins}}$) can vary from 0.05 to 0.6 W/(m·K) depending on temperature and how tightly it is packed [11]. Because of the highly empirical nature of this parameter, it was chosen as means of calibration of our model — by varying this parameter we would ensure that the calculated temperature field corresponds to the measurements done with pyrometer.

3.3. Approach to convective heat transfer modelling
“CZ-Trans” software currently does not support 3rd type boundary condition and, therefore, convective heat transfer from steel casing to ambient air require a specialized modelling approach. To achieve realistic temperature field within the system, emissivity of the steel case was increased as means to introduce additional heat flux through the insulation layer.
The approach to introduce effective emissivity $\varepsilon_{\text{eff}}$ instead of using the actual material property $\varepsilon_{\text{surf}}$ can be related to the use of effective heat transfer coefficient $h_{\text{surf}}$, described in [9]. First, we assume that the heat flux from the steel casing of the crucible block consists of two parts: radiative and convective heat losses (equation 1).

$$q_{\text{surf}} = q_{\text{rad}} + q_{\text{conv}}.$$ (1)

Then we can write these heat fluxes explicitly by using the effective emissivity $\varepsilon_{\text{eff}}$ and heat transfer coefficient $h_{\text{surf}}$ (equation 2).

$$\varepsilon_{\text{eff}} \sigma_{\text{SB}} (T_{\text{surf}}^4 - T_{\text{amb}}^4) = \varepsilon_{\text{surf}} \sigma_{\text{SB}} (T_{\text{surf}}^4 - T_{\text{amb}}^4) + h_{\text{surf}} (T_{\text{surf}} - T_{\text{amb}}).$$ (2)

We then solve the equation 2 for $h_{\text{surf}}$, to evaluate how large is the heat transfer coefficient effectively applied in our simulation, when we find the appropriate $\varepsilon_{\text{eff}}$ for the particular set of parameters and temperatures (equation 3).

$$h_{\text{surf}} = \frac{\sigma_{\text{SB}} (T_{\text{surf}}^4 - T_{\text{amb}}^4)(\varepsilon_{\text{eff}} - \varepsilon_{\text{surf}})}{T_{\text{surf}} - T_{\text{amb}}}.$$ (3)

It must be noted that using this approach (applying effective emissivity instead of 3rd type boundary condition) is entirely suitable with $\varepsilon_{\text{eff}} > 1$ as it is only means to achieve the required heat loss from a surface that radiates only to ambient environment.

3.4. Simulation results of a stationary temperature field matched to the experiment

At this point during the modelling process, there are two unknown parameters that need to be adjusted to achieve realistic temperature field for the FEM simulations — insulation thermal conductivity $\lambda_{\text{ins}}$ and effective emissivity of the steel casing $\varepsilon_{\text{eff}}$ — which is the necessary amount of free parameters to match the two experimentally measured temperatures: of the crucible edge and outer steel casing (see table 1). A series of calculations were performed to achieve this goal.

The final (fitted) calculated temperature field as well as simulation FEM mesh is shown in figures 5 and 6. We can see that the temperatures are indeed matched to the measured values mentioned earlier — calculated values range from 450 to 550 K on the outer steel casing and from 850 to 900 K on the crucible edge.

The summary of the optimized parameters and their relationship to values found in literature are shown in table 1. The optimal emissivity $\varepsilon_{\text{eff}}$ that ensures this temperature field is 1.0. The heat exchange coefficient $h_{\text{surf}}$ that corresponds to such a choice is 7.5 W/(m$^2$·K) which is entirely realistic given the data in [9]. It can be concluded that the main thermal parameters of our model are successfully fitted to perform transient simulations of the crystal growth experiment.

4. Transient simulations

4.1. Simulation of the laboratory crystal growth experiment

After performing the model calibration, it was possible to use our transient simulation capabilities to replicate the experimental growth conditions as close as possible. PID control algorithms were used to optimize the growth parameters (pull rate and heater power) to match the experimental crystal shape. As seen in figures 7 and 8, calculated crystal shape is matched considerably well to the experimental shape seen in figure 2. It must be noted that temperature field and heater power values are also consistent with experimental conditions.

Despite the reasonable precision of the crystal shape modelling, figure 7 shows that the desired pull rate of 0.6 mm/min during the simulation is not maintained. This fact is due to the way PID controller cascade is set up: 1) pull rate is adjusted to maintain the required shape and 2) the heater power is adjusted to minimize the pull rate adjustment.
Figure 5. Mesh used for FEM simulations of temperature field within the crucible-heater unit.

Figure 6. Result of the stationary temperature field calculation.

Figure 7. Crystal radius, pull rate and heater power during the laboratory crystal growth experiment (dashed line) and transient simulation (solid line).

Figure 8. Temperature field and phase boundaries at the end of transient simulation.
Table 1. Summary of known and calibrated parameters that are featured in the initial stationary temperature field simulations.

| Property                           | Denotation | Value       | Notes                                                                 |
|------------------------------------|------------|-------------|----------------------------------------------------------------------|
| Ambient temperature                | $T_{amb.}$ | 300 K       | Experiment was performed in room temperature.                        |
| Steel casing covered with aluminium paint | $\varepsilon_{surf.}$ | 0.4          | Value found in [9]. Value increased in simulation to account for convective cooling (see $\varepsilon_{eff.}$ below). |
| Average crucible edge temperature  | $T_{edge}$ | 850 K       | Measured experimentally, **must be matched by simulation.**          |
| Average steel casing temperature   | $T_{surf.}$ | 450 K       | Measured experimentally, **must be matched by simulation.**          |
| Effective emissivity of steel casing | $\varepsilon_{eff.}$ | 1.0          | Free **parameter that was optimized** during numerical studies to match temperature field to measured values. |
| Heat exchange coefficient for steel casing | $h_{surf.}$ | 7.5 W/(m²·K) | Value that corresponds to $\varepsilon_{eff.}$ listed above. This corresponds to typical values for free convection found in [9]. |
| Insulation thermal conductivity    | $\lambda_{ins.}$ | 0.5 W/(mK)  | Free **parameter that was optimized** during numerical studies to match temperature field to measured values. |

Evidently, the experimental heater power curve is not suitable for the modelling case and, therefore, large deviation of the pull rate is necessary to maintain the crystal shape. This problem can be solved by iteratively applying the PID-produced power curve for consecutive simulation cases and shifting it towards earlier times. In this way, the temperature near the crystal will be exactly right, despite thermal inertia of the whole furnace.

Furthermore, figure 8 shows a concave crystallization interface. This is not consistent with experimental conditions where it has been observed (by abruptly pulling out the crystal from the melt) that the interface is convex — central part is located lower in the melt.

4.2. **Simplified model for heat losses through the transparent crystal**

The difference of crystallization interface shape between the model and experimental conditions is related to the transparency of the crystal. Presented results consider mathematical model where the radiative heat transfer within the melt and crystal is not included. However, we propose a simplified model that can ensures sufficient precision of simulated phase boundary shape, but do not require substantial model development effort. By knowing the temperature (melting point) and emissivity of the crystallization interface [12] the corresponding radiative heat flux can be calculated as shown in equation 4.
\[ q_{\text{rad.}} = \varepsilon_{SB}T^4 = 0.962 \cdot 6.67 \cdot 10^{-8} \cdot 815^4 = 24.1 \text{ kW/m}^2 \] (4)

The calculated heat flux density is assumed to be lost (radiated) from the crystallization interface and transferred away without absorption and reflection. Therefore, it was applied as a constant negative heat source on the crystallization boundary in our model. By using this approach, results seen in figure 10 are achieved — the crystallization interface is indeed concave, giving better match to experimental conditions.

By introducing the negative heat sources on the interface, the total conductive heat flux from the interface into the crystal is reduced significantly (figure 9) and the crystallization occurs more rapidly. As a result, the presented cases (figures 8 and 10) with roughly the same crystal length of 15 mm have considerably different duration — 28 minutes without the heat flux correction and only 12 minutes for the corrected case. Additionally, crystal has larger diameter as the predefined (experimental) crystal shape is increasingly harder to maintain by using the PID algorithms with the same parameters.

5. Conclusions
Calculation software used primarily for the modelling of industrial-scale silicon crystal growth process has been successfully used for the optimization of unknown material properties of a small-scale laboratory NaCl-RbCl crystal growth furnace. Stationary temperature field that corresponds to experimentally measured temperatures have been calculated. This result was obtained by choosing thermal conductivity of 0.5 W/(m·K) for the thermal insulation layer and choosing effective emissivity coefficient of 1.0 for the outer steel case. Such approach is equivalent to using convective heat transfer coefficient of 7.5 W/(m²·K).

The obtained stationary calculation results and model parameters were used as basis for the transient simulation of the laboratory crystal growth experiment. Sufficient match between the modelled and experimental crystal shape was achieved, although additional studies are necessary to acquire optimal heater power distribution that ensures the required pull rate of 0.6 mm/min.

Simplified model of heat losses from crystallization interface due to radiative heat transfer through the transparent crystal was proposed. By introducing additional, constant heat flux density on the crystallization interface, a convex crystallization interface shape was achieved — a result that corresponds to experimental conditions.
Acknowledgments
The research was carried out with the financial support from SIA “Mikrotikls”. University of Latvia Foundation project “Implementation of Experimental Laboratory Crystal Growth Furnace” (project number: 2183).

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