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Implementation of Small-scale Laboratory Crystal Growth Furnace for the Development of Mathematical Model for Large-Scale Industrial CZ Process

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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 used.

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 author's research group in the UL Faculty of Physics and Mathematics has developed such mathematical modelling tools for the simulation of the CZ crystal growth process [7]. 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.

1. Problem definition

Author of this article has access to small-scale laboratory CZ crystal growth furnace which can be used for the mathematical model verification described previously. This furnace consists of an electrically heated crucible unit (see schematics in Fig. 2, left) 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 [8]. 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, translucent melt and crystal), system still exhibits all the physical phenomena that can be related to silicon crystal growth.

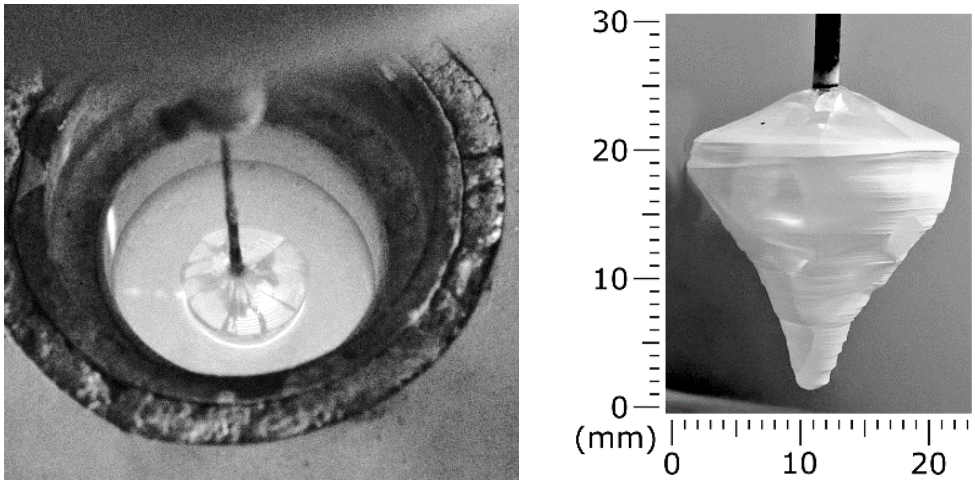

Fig. 1. Photography of the NaCl-RbCl salt crystal growth process (left) and example of a grown crystal (right)

The general construction of the furnace as well as material property combination (latent heat and thermal conductivity ratio) ensures that overall structure of the temperature field, its gradients as well as phase boundary dynamics are very similar for both industrial-scale silicon furnace and small-scale salt crystal growth. This can be verified when comparing results in Section 3 to results in [7]. Translucency of salt crystal and melt is ignored during the initial studies performed in this article – future developments and experimental results will dictate the necessity to include this phenomenon in the mathematical model.

The crystal growth with NaCl-RbCl can be performed under atmospheric pressure and do not require any type of enclosure or pressure vessel. Optimal growth conditions are achieved at pulling speeds of 0.6 mm/min (see illustrative photography in Fig. 1, left). Crystals can be grown with diameter around 20 mm (see example in Fig. 1, right). After performing experimental crystal growth shown in Fig. 1, 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.

2. Calibration of model parameters

2.1. Overview of the mathematical model and creation of geometry

As described in [8], applied simulation software can perform axisymmetric non-stationary simulations of: (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 (Fig. 2). As it can be seen, ceramic parts are unified into a single domain and separate windings of the heater wire are modelled by a single FEM block. These simplifications are required to correctly model the radiation heat transfer between these components.

2.2. Data from experimental heating of crucible as means for model calibration

During the experiment, before the crystal growth shown in Fig. 1 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 [8] and corundum ceramic parts [11] are well known (see Fig. 2 for summary). However, the thermal conductivity of the silica insulation wool used in the construction of the crucible is not known. This parameter ($\lambda_{ins.}$) can vary from 0.05 to 0.6 W/(m×K) depending on temperature and how tightly it is packed [12]. 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.

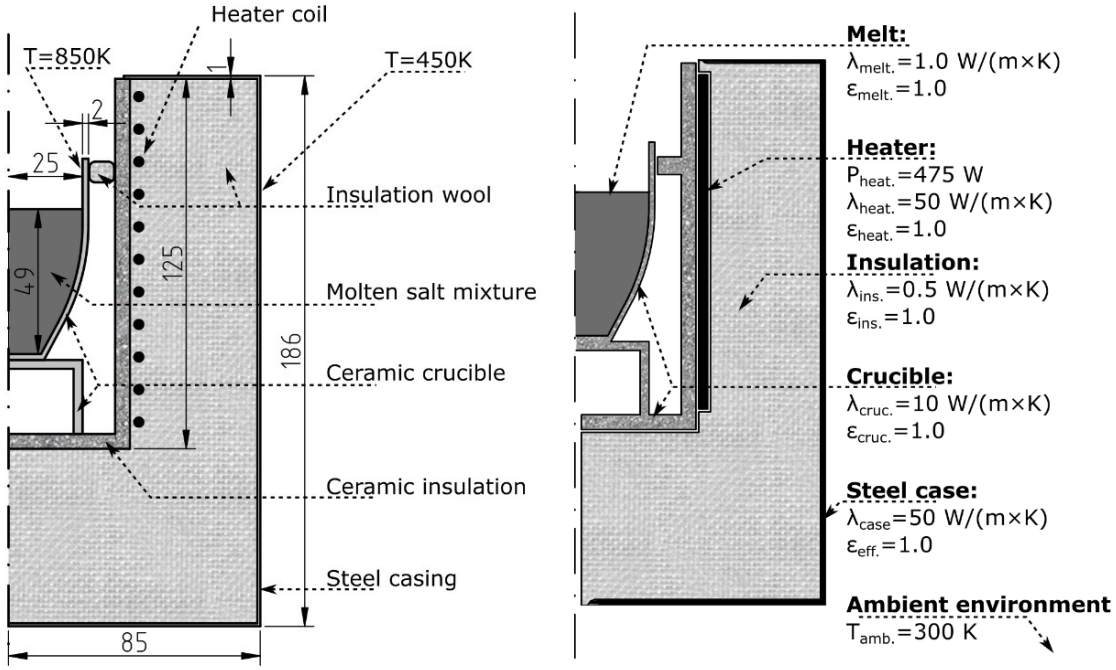

Fig. 2. Schematics of the precise geometry of the crucible-heater unit used in the laboratory crystal growth furnace (left, dimensions in mm) and simplified geometry for the creation of FEM domains used in numerical simulations (right)

2.3. Approach to convective heat transfer modelling

Our mathematical model does not include convective heat transfer from steel casing to ambient air. To mitigate this problem and 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 $\varepsilon_{eff.}$ instead of using the actual material property $\varepsilon_{surf.}$ can be related to the use of effective heat transfer coefficient $h_{surf.}$ described in [10]. First, we assume that the heat flux from the steel casing of the crucible block consists of two parts: radiative and convective heat losses (Eq. 1).

$$q_{surf.} = q_{rad.} + q_{conv.} \tag{1}$$

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

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

We then solve the Eq. 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 (Eq. 3).

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

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

In our case, all the temperatures in Eq. 3 are known experimentally as well as the emissivity of the painted steel surface. By using this information and various effective emissivity $\varepsilon_{\text{eff.}}$ and thermal conductivity $\lambda_{\text{ins.}}$ values, a series of calculations was performed to achieve realistic temperature field that corresponds to the temperature measurements.

The final (fitted) calculated temperature field as well as simulation FEM mesh is shown in Fig. 3. We can see that the temperatures are indeed matched to the measured values. The summary of the optimized parameters and their relationship to values found in literature are shown in Tab. 1. The 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 [10]. It can be concluded that the main thermal parameters of our model are successfully fitted to perform transient simulations of the crystal growth experiment.

Tab. 1. Summary of known and calibrated parameters that are featured in the initial stationary temperature field simulations.

Property	Value	Notes
Ambient temperature, $T_{\text{amb.}}$	300 K	Experiment was performed in room temperature.
Steel casing covered with aluminium paint, $\varepsilon_{\text{surf.}}$	0.4	Value found in [10]. Value increased in simulation to account for convective cooling (see $\varepsilon_{\text{eff.}}$. below).
Average crucible edge temperature, T_{edge}	850 K	Measured experimentally, must be matched by simulation.
Average steel casing temperature, $T_{\text{surf.}}$	450 K	Measured experimentally, must be matched by simulation.
Effective emissivity of steel casing, $\varepsilon_{\text{eff.}}$	1.0	Free parameter that was optimized during numerical studies to match temperature field to measured values.
Heat exchange coefficient for steel casing, $h_{\text{surf.}}$	7.5 W/(m^2×K)	Value that corresponds to $\varepsilon_{\text{eff.}}$ listed above. This corresponds to typical values for free convection found in [10].
Insulation thermal conductivity, $\lambda_{\text{ins.}}$	0.5 W/(m×K)	Free parameter that was optimized during numerical studies to match temperature field to measured values.

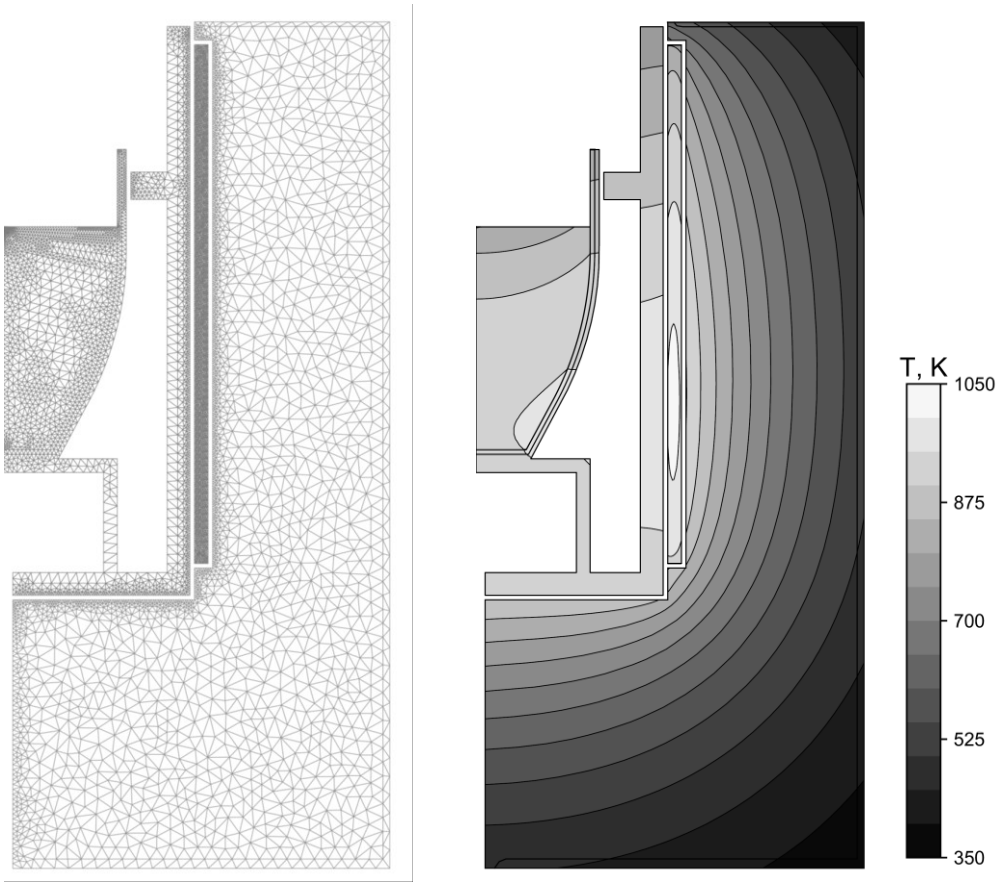

Fig. 3. Mesh used for FEM simulations of temperature field within the crucible-heater unit (left) and results for temperature field calculation (right)

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/(m^2×K).

The obtained result is a critical first step for the continuation of the numerical studies with small-scale laboratory furnace, i.e. stationary calculations will be used as initial conditions for the fully transient simulations. After the development of full, non-stationary model for the laboratory crystal growth experiments, various modelling approaches can be tested to validate the performance and precision of the developed calculation software.

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