



Virtual reactor as a new tool for modeling and optimization of SiC bulk crystal growth

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Abstract

We propose a new approach to optimization of SiC bulk crystal growth based on modeling. The idea is to employ a special software tool “virtual reactor” (VR) operated by the user of the code as an actual crystal growth system. The software tool includes the models necessary to simulate global heat transfer in the whole growth system and inside the crucible including radiative transport through the semi-transparent SiC crystal. It is known that accurate material properties are crucial for thermal modeling of SiC growth. A database with material properties of SiC crystal and powder, graphites and insulation is included into the VR-software. An advanced model of species transport during sublimation growth of SiC crystals is developed. The model includes convective and diffusive species transport, surface kinetics based on the Hertz–Knudsen equations and chemical models for all solid surfaces (SiC crystal, SiC source, graphite wall). A model to predict type of parasitic deposit and the corresponding deposition rate is combined with the mass transport model available in the VR-software. In this paper, we show the results of simulation of a large-size SiC bulk crystal growth using the VR-software tool with the focus on poly-SiC deposit formation on the graphite crucible lid around the crystal. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Sublimation growth of SiC bulk crystals is the basic method to obtain high-quality large-size SiC

single crystals. Experimental optimization of the growth process normally takes much effort and is time-consuming. Thus, modeling of SiC sublimation growth can be quite beneficial if the simulation tool can predict important features of the process.

Many studies in the recent years have aimed at the development of a comprehensive model for SiC

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sublimation growth. Heat transfer in the systems used for growth of bulk SiC crystals was studied in Refs. [1–3]. Species transport inside the crucible coupled with the heat transfer modeling was simulated in Refs. [4,5].

Most modeling approaches consider SiC sublimation growth to be a steady-state process. However, non-stationarity is an inherent feature of the growth technique, since several factors result in a gradual change of the SiC growth rate and of the powder properties. An important unsteady aspect of the sublimation growth ignored by most theoretical studies is the formation of polycrystalline SiC deposit on the graphite walls of the crucible [6].

Temporal evolution in the sublimation growth has been recently studied in Refs. [7–10]. An extensive experimental analysis made in [7,8] was focused on the powder source degradation. Theoretical and experimental study of the crystal shape development was carried out in [9]. Evolution of shape of the growing SiC bulk crystal was first simulated in [10].

In the present paper, we report on modeling of a long-term silicon carbide bulk crystal growth. The simulations have been carried out using a specialized software tool with automatic geometry modification (the so-called “virtual reactor”).

2. Concept of the “virtual reactor” software for modeling sublimation growth of SiC bulk crystals

We propose a quasi-steady-state approach to simulate a long-term sublimation growth of bulk SiC crystals. The overall modeling is carried out by solving a series of steady-state problems at various discrete moments of growth. The quasi-steady-state concept implies that the transient times of all the processes occurring in the system are much less than the growth duration. This allows one to replace an actual growth process with the slow variation of parameters by a number of coupled steady-state steps with the constant parameters.

A special software called “virtual reactor” has been developed to implement the approach. It is important to note that the growth system geometry can be introduced using CAD files followed by

automatic generation of the computational grid. Material properties are also automatically transferred into the code from the database. A key feature of the VR-software tool is that it is operated by the user of the code like an actual growth system. The user has to program the growth process by prescribing heating of the system, variation of inert gas pressure and of the coil movement during the growth process. All other temporal changes in the system are made by the VR-software automatically. At each stage, heat transfer coupled with the reactive species transport is computed. Then the SiC growth rate on the seed and the rates of formation of various deposits at the crucible walls are calculated, and the system geometry including the moving inductor coil is automatically modified with corresponding grid regeneration. The VR-software allows also continuous “virtual characterization” of the growing crystal by calculating thermoelastic stress, dislocation distribution, by predicting graphite inclusion formation in the crystal during growth, etc. The software tool is found to be a useful approach for prior examination of the effects of growth system design and process parameters modification on the properties of the grown SiC bulk crystal. The software is PC-compatible with computing time in the range of a few hours.

3. Prediction of deposit formation on the crucible walls and of Si- or C-inclusion formation on the growth surface

A model has been developed for prediction of parasitic deposition on the crucible walls and of the Si- or C-inclusion formation on the growth surface to automatically adjust the type of heterogeneous equilibria to be used in the boundary conditions for the mass transport equations [11]. The model considers all types of the condensed phases possible in the Si–C system to be in equilibrium with the initial SiC or graphite surface. Spontaneous generation of the microscopic islands of the condensed phases on the surface is assumed. Since liquid silicon cannot be in equilibrium with graphite at the temperatures of practical importance, the following cases are considered in the

model: graphite islands on SiC, liquid silicon droplets on SiC, and islands of polycrystalline SiC on graphite. A detailed mathematical description of the deposition criteria can be found in [12].

If the initial surface is graphite and the islands of poly-SiC are generated on the surface, SiC–C three-phase equilibrium is used in the boundary conditions [11]. From the partial pressures of the gaseous reactive species (Si, Si₂C and SiC₂) we calculate the growth rates for both condensed phases — SiC and graphite [9]. If both growth rates are positive, SiC and graphite co-exist on the wall surface. The values of the rates provide the fractions of the surface covered by SiC and graphite, respectively. If SiC growth rate is positive, whereas that of graphite is negative, this means that the SiC islands spread over the surface, i.e. SiC deposition dominates over that of graphite. If the SiC growth rate is negative, the spontaneously generated SiC islands disappear and the surface remains to be graphite. It should be noted that etching of graphite walls of the crucible plays an important role in the supply of carbon atoms into the growing SiC crystal [5] and

this mechanism is allowed for in the model of species transport during sublimation growth.

4. Modeling approach

To study crystal shape evolution and dynamics of deposit formation, we simulated SiC bulk crystal growth in a system shown schematically on the left side of Fig. 1. Heat transfer was modeled accounting for both conductive and radiative mechanisms. The species transport inside the crucible was analyzed by using a set of Navier–Stokes equations. Modeling of the coupled species transport and heterogeneous processes was carried out in a self-consistent manner. First, the mass-transport equations were solved with the boundary conditions obtained from the initial phase state of all the surfaces. Then, formation of deposits was analyzed and, if the phase state of a surface area was predicted to be changed, the species transport simulations were repeated again with new boundary conditions. The iterations were made until convergence of the calculations was reached. As a

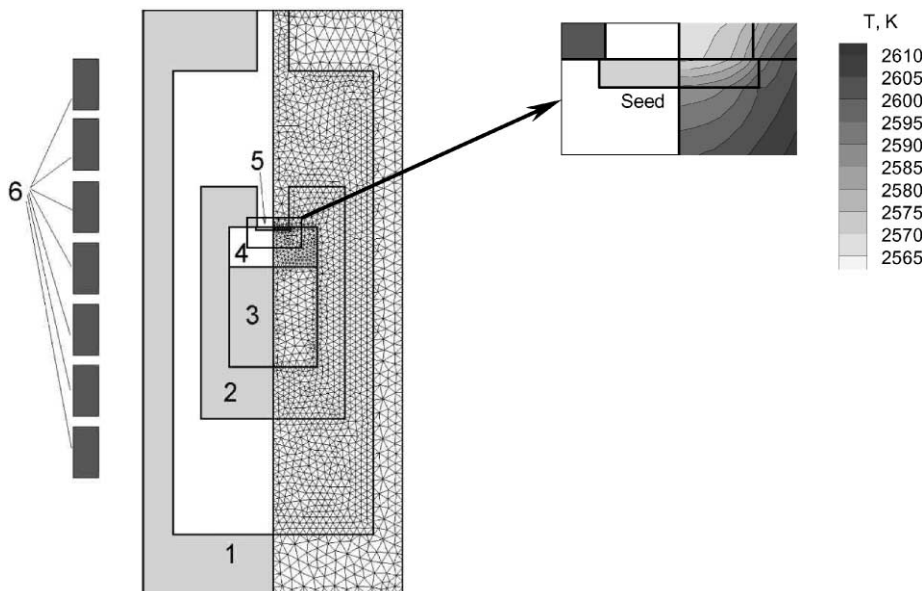


Fig. 1. Schematic view of a growth system: (1) thermal insulation (2) crucible (3) powder charge (4) growth cell (5) seed (6) inductor coil. Computational grid is shown on the right side. The inset shows a fragment of the crucible near the seed combined with the initial temperature distribution.

result, a steady-state distribution of species concentrations inside the crucible is computed along with various deposits on the crucible wall, sublimation of SiC source, etching of graphite, and growth of SiC crystal on the seed. The equations were solved using a finite-volume technique on unstructured computational grids [13]. A typical grid used in the calculations is displayed on the right side of Fig. 1.

5. Results and discussion

Temperature distribution in the crucible near the seed is shown in Fig. 1. Growth was carried out with the temperature at the upper central point of the seed being 2470 K, thus the heat power generated due to the inductive heating was varied with time to maintain this temperature. The current frequency in the coil was 8 kHz, the total pressure inside the crucible was kept at 30 mbar. Fig. 2 demonstrates the initial and the current phase states of the graphite crucible surfaces at the onset of growth combined with the temperature distribution in the growth cell. One can see that poly-SiC deposit is predicted to appear at the crucible lid around the seed under the conditions considered. No SiC deposition occurs at a hotter side wall of the crucible. Besides, graphitization of SiC powder is predicted to occur near the side wall of the crucible. These results agree well with the experimental observations reported in [6,8].

Simulation of the initial growth, i.e. during the first hour, was carried out with time steps of

15 min. The further growth was modeled with time steps of 1 h. Meanwhile, the inductor coil was moved downward at a speed of 1 mm/h. The important role of the poly-SiC deposition in species transport and crystal shape control can be seen from Fig. 3. Calculations without taking into account poly-SiC deposition give non-realistic shape of the growing crystal.

Next, we examined possible ways for suppression of poly-SiC deposition around the seed in the growth system considered. First, effect of process parameters like the seed temperature or the inert gas (Ar) pressure was studied. It was found that the deposit formation could not be reduced significantly by only changing the process parameters. Another possibility is to optimize the inductor coil position that controls the temperature gradients in the growth cell. The coil position was varied in the range of 180 mm (~30% its length), whereas the temperature at the control point was maintained at 2470 K. It was found that the coil position has a significant effect on the deposit formation in this growth system. In the case of lowered coil the poly-SiC deposition is significant and results in reduced lateral growth of SiC crystal. On the contrary, in the case of lifted coil the deposition is significantly suppressed and allows considerable enlargement of SiC single crystal. Fig. 4 compares the temperature distribution and the crystal shape after 10 h of growth for the two coil positions discussed above.

These results qualitatively agree with the experimental observations during SiC bulk crystal growth by the group of Vodakov [14]. The VR-software is tested and extensively used in this

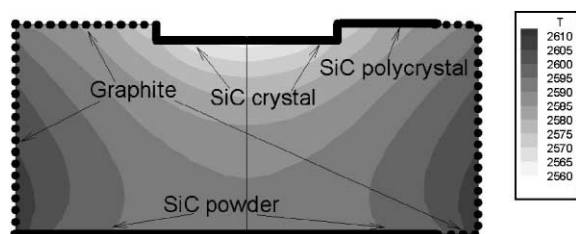


Fig. 2. The initial (left) and predicted (right) phase states of the reactive crucible walls for the onset of growth. Solid and dotted lines denote SiC and graphite surfaces, respectively. The initial temperature distribution in the growth cell is also shown.

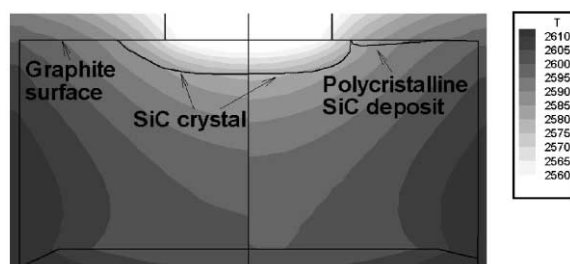


Fig. 3. The crystal shape after 4 h of growth accounting (right) and ignoring (left) the deposit formation and the corresponding temperature distribution.

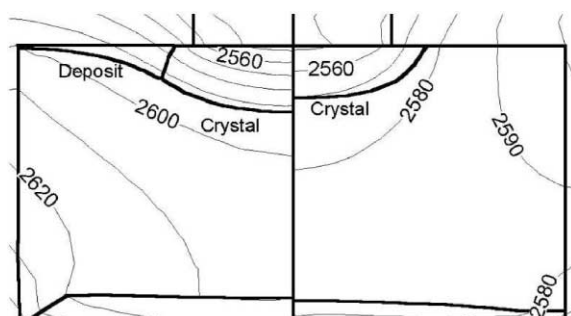


Fig. 4. The crystal shape obtained after 10 h of growth accounting for deposit formation for two coil positions. Inductor coil was shifted downwards at 60 mm (left) and upward at 110 mm (right) from a medium position. The temperature distribution for both cases is also shown.

group for optimization of the growth system design and the process conditions to provide SiC bulk crystals with reduced defect concentrations. Results of the crystal growth will be reported elsewhere.

6. Conclusion

Simulation of a long-term sublimation growth of SiC bulk crystals has been carried out using a special software tool named “virtual reactor”. It was shown that formation of the polycrystalline SiC deposit at the crucible lid around the seed strongly affects evolution of the main SiC crystal shape. It was found that the poly-SiC deposition can be effectively suppressed by optimizing the position of the inductor coil during the growth process.

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