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Numerical modeling and experimental verification of modified-PVT crystal growth of SiC

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Abstract

This work focuses on numerical modeling of temperature field and mass transport in a so-called modified physical vapor transport (M-PVT) growth configuration, which is a further development of the state-of-the-art SiC bulk crystal growth technique. Modeling has been carried out by adapting the commercial computer code CFD-ACE+ (CFD Research Corporation) to the special application. In particular, linked to experimental results in the own lab, we will first show that modeling may be applied to numerically reproduce the impact of an additional gas inlet on the resulting effective mass transport inside the growth setup. In a second step, we will use modeling to understand the physical and chemical processes during crystal growth. We will show how additional gases may be used to tailor the gas phase composition, i.e. C/Si ratio, in front of the SiC growth interface.

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1. Introduction

The physical vapor transport (PVT) technique has become the state-of-the-art process for commercial SiC single crystal growth [1]. Using a modified physical vapor transport (M-PVT) setup (Fig. 1) with an additional gas pipe for fine tuning of the gas phase composition, improved doping of SiC has been reported [2]; today, for example, the highest aluminum p-type doping of SiC has been achieved ($\rho = 0.1$–$0.2\,\Omega\,\mathrm{cm}$), which for the first time meets device fabrication needs. In addition, the M-PVT growth configuration opens the possibility to directly adjust the C/Si ratio in the gas phase in front of the SiC growth interface. For the latter numerical modeling of temperature field and mass transport inside, the growth setup is an essential tool to plan SiC growth experiments and also to evaluate the results. This paper addresses numerical modeling of the M-PVT process and its interplay with experimental verification.

2. Growth process and numerical modeling

2.1. Modified physical vapor transport growth technique

SiC single crystals are fabricated at elevated temperatures above 2000 °C by the so-called PVT method in a closed graphite crucible. Typically, SiC powder is placed in the hot zone at the bottom of the growth cell (e.g. 2200 °C). After sublimation it re-crystallizes in the colder zone in the top area at the SiC seed (e.g. 2150 °C) (Fig. 1, left). The growth process may be controlled by setting of the inductive heating power (→ global growth temperature, typically $T > 2000$ °C), proper crucible design and position of inductive heating coil (→ temperature gradients; axial temperature gradient, typically 10–30 °C/cm; radial temperature gradient as low as possible), and inert gas pressure (→ adjustment of diffusive species transport from the hot SiC source to the colder SiC seed; typical inert gas pressures and growth rates are 30–50 mbar and

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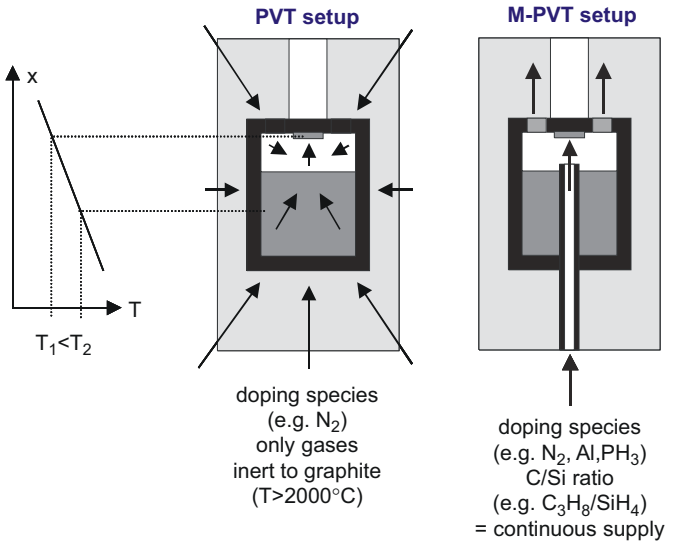

Fig. 1. Sketch of the PVT and M-PVT growth configuration. In the case of M-PVT an additional gas pipe is used for species feeding for fine tuning of the gas phase composition (doping or C/Si ratio) in front of the SiC crystal growth interface.

300–500 μm/h, respectively). All these process parameters influence the C/Si ratio in front of the SiC growth interface; a direct control, however, is not possible. The M- PVT technique (Fig. 1, right) [3] accounts for the drawbacks of the conventional growth configuration by offering an additional degree of freedom through feeding of gases like silane (SiH_4), propane (C_3H_8) or doping species (N_2, PH_3, Al vapor) through the pipe and, hence, the possibility of tuning of the gas phase composition (adjustment of C/Si ratio and controlled doping). Compared to the so-called high-temperature chemical vapor deposition (HTCVD) [4] growth setup, which uses silane and ethylene (C_2H_4) as source material in the M-PVT process the additional gases are solely used to fine tune the well-established PVT growth process where SiC powder is used as the main source material.

2.2. Numerical modeling

While numerical modeling of the conventional SiC PVT process may be called "state-of-the-art", the M-PVT configuration faces some new challenges because it combines PVT and CVD. In this work, we have used the CFD-ACE+ software package (CFD Research Corporation, Huntsville, AL, USA), which offers a wide spectrum of built-in modules for heat and mass transport calculations, including chemistry [5]. In addition, user subroutines can be implemented. We have carried out numerical modeling of the M-PVT growth considering heat transfer, SiC mass transfer from source to seed and impact of inner gas flux on heat and mass transfer inside the growth cell. In the case of propane as additional gas we performed calculations with and without consideration of thermal propane decomposition during feeding. In the case of heat transfer, we took into account heat conduction, radiation, convection as well as Stefan flow. In the CFD-ACE+ software package, heat transfer by radiation is calculated by the so-called discrete ordinates model (DOM); the surface of the various materials, i.e. graphite, SiC powder, and SiC crystal, were treated as gray radiators with an emissivity smaller than 1 (e.g. 0.9 for graphite). Materials properties at the elevated growth temperatures above 2000 °C are based on theoretical extrapolation of experimental data acquired up to 1700 °C [6]. SiC mass transport, i.e. sublimation and crystallization, was calculated using a user subroutine. A main assumption was stoichiometry of SiC sublimation and crystallization. In an accurate treatment one would consider decomposition of SiC into the major gas species Si, Si_2C, and SiC_2. In this particular case (graphitization of source material is less than 3%–5% [7], no silicon droplet formation is observed), the assumption of stoichiometric SiC mass transfer is justified. We treated the SiC source material as a porous media with a porosity of 70% (derived from initial SiC source material density). The parameters for pore size, permeability, and effective surface were chosen for a granular powder morphology as in the initial growth stage; sintered SiC blocks with hollow cores as they typically occur during later stage of growth were not taken into account [8]. When feeding propane (C_3H_8) into the growth cell, we considered gas decomposition and carbon deposition on the inlet pipe wall but did not take into account hydrogen etching of the graphite pipe and crucible walls. All calculations were performed in 2D with axial symmetry.

3. Results and discussion

3.1. Modeling of principle M-PVT process

With the above-mentioned modeling approach, we recently reported on numerical calculations of the principle M-PVT process operation considering the feeding of the inert gas helium into the growth cell (for details see Ref. [2]). We could verify in modeling the impact of the additional gas flow on the SiC mass transport from source to seed and semi-quantitatively reproduce growth rates and evolution of crystal growth interface, indicating that all crucial physical and chemical effects have been considered in the computer code. In short: we found that an intermediate gas flux comparable to the SiC PVT species flux is ideal; in this case, a convection roll is stabilized, which supports mixture of SiC PVT flux and additional gas flux and leads to homogeneous SiC deposition on the seed. A minimum gas flux is necessary to establish long-term stable inlet conditions (no closure of the pipe); a too high gas flux, however, will cause negative SiC growth rates on the seed (etching of the seed). Based on these results, in this work we have refined our modeling, including (i) chemical reactions between the inlet gas and the main SiC powder sublimation species Si, Si_2C, and SiC_2 as well as growth cell material and (ii) variation of isolation property changes in