

Free growth of 4H-SiC by sublimation method

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Abstract. 4H-SiC crystals of cylindrical shape 25 mm and 45 mm in diameter have been grown by the Modified Lely method in a graphite crucible with a guide. The crystals had not mechanical contacts with the walls of the crucible and no formation of polycrystalline SiC during the growth to decrease the stresses. This was confirmed by the facets observed at the edges of the crystals. Numerical simulations of the growth process have been performed.

Introduction

Silicon carbide is a wide band gap semiconductor with unique properties that make it suitable for application in high frequency, high temperature and high power devices. These applications prompted increased interest in growth of high quality, large diameter single crystals with defect reduction. The objective of this paper is the study of 4H-SiC growth by the Modified Lely method without polycrystalline rim and mechanical contact with graphite walls of the crucible in order to decrease the stresses. Besides, the aim was to obtain crystals with a cylindrical shape and flat top surface to minimize the number of low grain boundaries. Numerical modeling has been used to optimize the design of the crucible.

Experimental details

4H-SiC crystals, 25 and 45 mm in diameter, have been grown by the Modified Lely method over the 2000-2050°C temperature range (measured by the top pyrometer : 1 in Fig. 2) in argon [1] (Fig.1). An experimental set-up with RF heating and graphite crucible was used. The crucible (Fig.2) was wrapped in a graphite felt for thermal insulation, and the whole assembly was placed inside a water cooled quartz reactor. Under these experimental conditions we obtained 4H single crystals with a thickness of 5-10 mm. The growth rate was 200-300 μm/h. An increase of the growth rate resulted in 6H-SiC growth. The SiC source powder was loaded both between a dense graphite crucible and a thin walled inner graphite cylinder and inside of the inner cylinder. With this configuration it is possible to reduce the influence of the leakage of Si and the reaction of Si from the main central source with the walls in order to maintain an excess of Si over the seed. To eliminate the influence of the periphery we cut out the seed after gluing to the graphite holder and avoided 6H polycrystalline SiC growth at the periphery. 4H-SiC wafers of 25 and 45 mm in diameter were used as seeds. The ingots were grown on the C-face of seeds which are 8° off-axis. The distance between the powder source and the seed was 15-20 mm.

The growth process consisted of :

- (a) annealing in vacuum at a temperature lower than 1000°C,
- (b) heating of the crucible up to growth temperature at a high argon pressure,
- (c) decrease of the pressure (3-5 torr) and growth at low argon pressure.

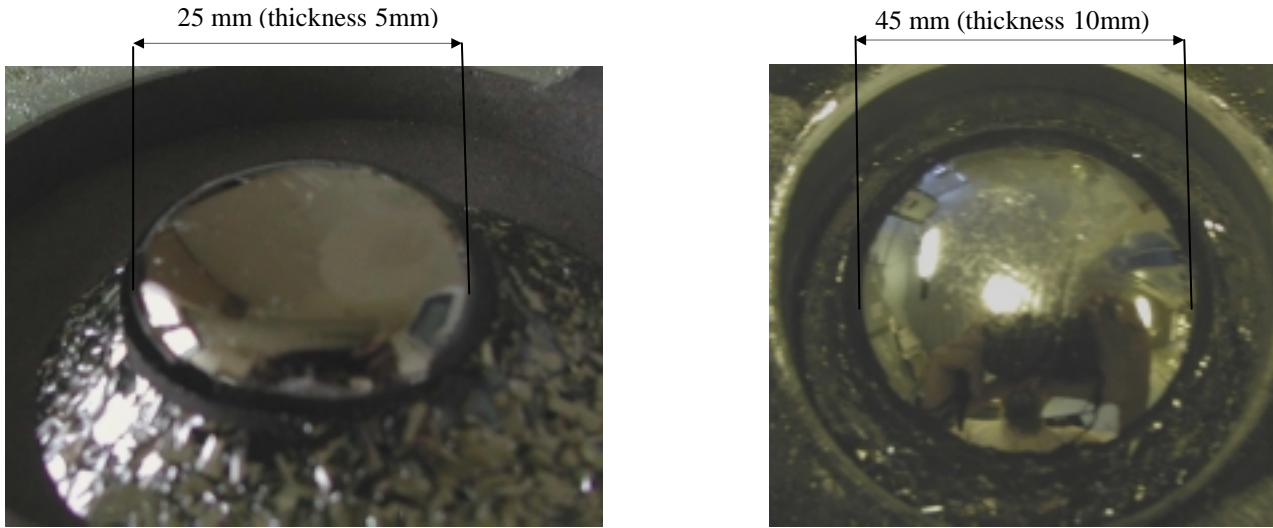


Fig.1. Photographs of “as-grown” 4H-SiC crystals

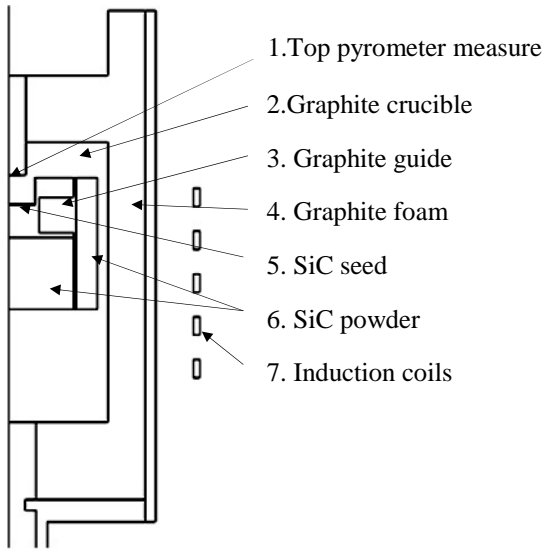


Fig. 2 : Schematic representation of the right-half part of the reactor with guide

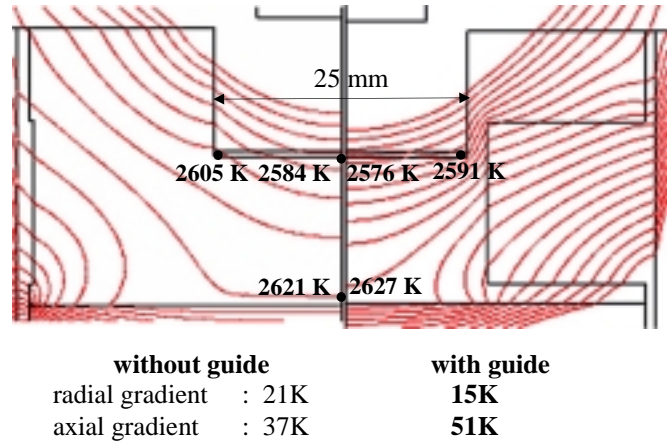


Fig. 3 : Comparison of temperature distribution in the growth cavity in different configurations

Numerical details

Intense effort has been focused on the crucible design to optimize thermal and concentration profiles in the growth area [1-7]. Electromagnetodynamics must be coupled with heat transfer, especially radiative heat transfer within the growth cavity [1]. The design of the crucible with and without a graphite guide (3 on Fig. 2) was optimized by heat transfer simulation in order to minimize the radial temperature gradient (Fig. 3). In order to consider chemical reactions that occur in growth cavity, we have used a model based on the heterogeneous chemistry [2]. We use Hertz-Knudsen equations (Eq. 1) to relate the partial pressures of the species P_i ($i=SiC_2, Si_2C, Si$) to their total mass flux R_i^S :

$$R_i^S = \gamma_i (P_i - \alpha_i P_i^{eq}) \sqrt{\frac{M_i}{2\pi RT}} \quad (i=Si, SiC_2, Si_2C) \quad (1)$$

Here γ_i is the sticking coefficient and α_i is the evaporation coefficient of the i -species. The equilibrium pressures P_i^{eq} obey to mass action law.

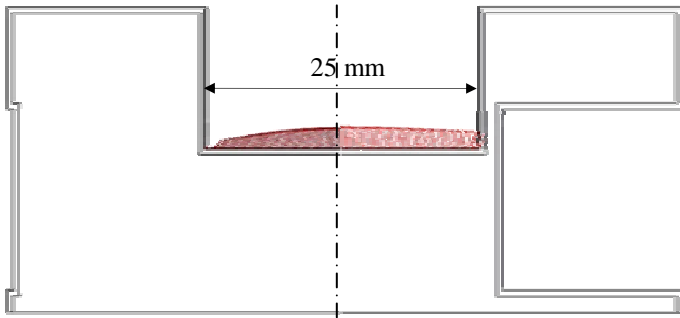


Fig. 4 : Comparison of deposited carbon flux in different configurations

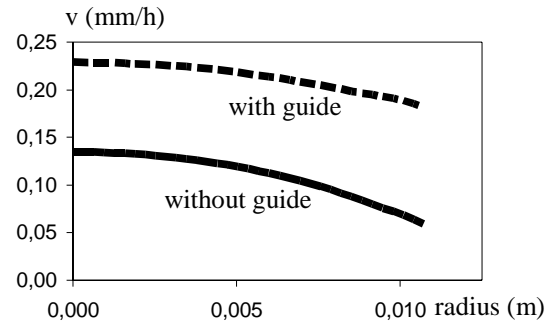


Fig. 5 : Growth rate vs. radius in different configurations

At the beginning of the growth, the carbon flux (Fig. 4) and growth rate (Fig. 5) have been computed on the seed surface. It was clear by comparison of the two different configurations (with vs. without guide) that the growth rate depends ($230\mu\text{m/h}$ vs. $140\mu\text{m/h}$) on the axial temperature (51K vs. 37K) and the crystal shape (flat vs. convex) depends on the radial temperature gradient (15K vs. 21K). Temperature gradient control is indeed an efficient way for lowering the defect density in SiC bulk crystals [8].

The coupled heat and mass transfer is simulated on triangular mesh of second order at every temporal step using a quasi-steady-state approach. The data obtained at the previous time step provide the necessary information (normal carbon flux, growth rate) for the next time step, and the computational mesh is regenerated between the steps in order to take into account the geometry modifications.

It is found that crystal with a flat growth surface can be obtained in the configuration with a guide (Fig. 5 and 6) compared to the configuration without a guide for a 25mm crystal diameter. In optimised geometry the growing crystal does not touch the crucible due to a high axial temperature gradient (Fig. 3, right side).

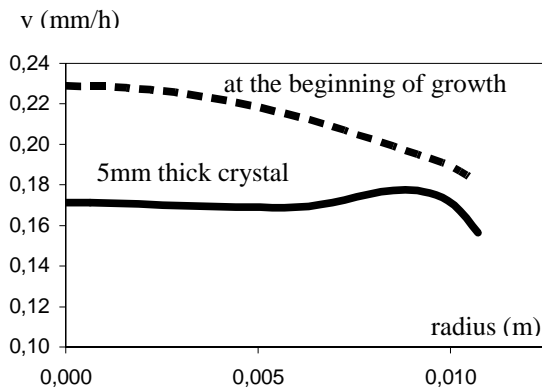


Fig. 6 : Growth rate vs. radius for the configuration with a guide

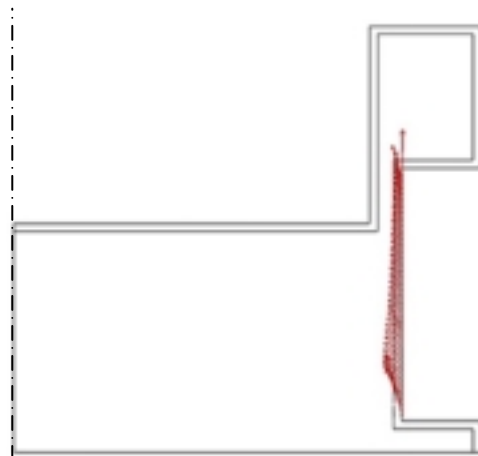


Fig. 7 : No deposited carbon flux on the guide

Negative carbon flux (no deposition) on the guide has been calculated by simulation (Fig. 7) and experimentally observed for crystal diameters of 25 and 45 mm. The facets were found at the edge of the grown crystals confirming the absence of contact between the crucible and the growing crystal. In the case of the crystal of 45mm in diameter, a higher radial temperature gradient was simulated (Fig. 8) and the crystal shape at different time step with the quasi-steady-state approach has been computed (Fig. 9). Due to a higher radial temperature gradient than for the case of the

crystal of 25mm in diameter (38K vs. 15K), we obtain numerically a slightly convex shape with a growth rate at center around 250 $\mu\text{m}/\text{h}$.

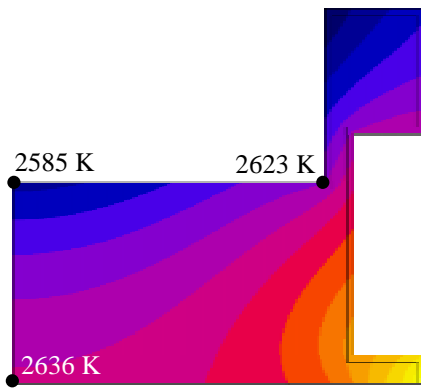


Fig. 8 : Temperature distribution

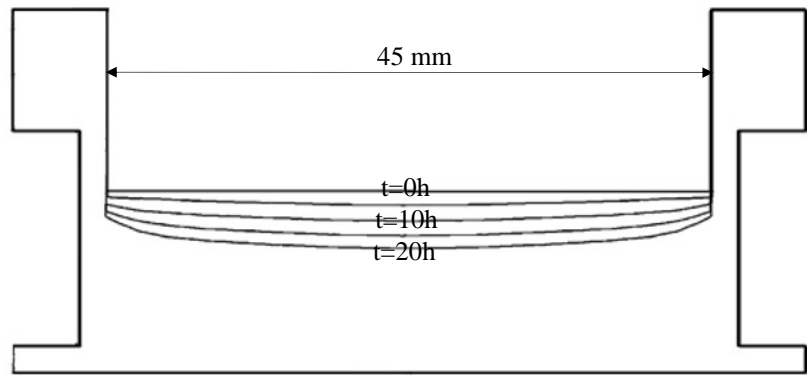


Fig. 9 : Temporal evolution of the crystal shape

Results and discussion

Polytype transformation

The 4H-6H SiC transformation was usually observed at the growth rates above 300 $\mu\text{m}/\text{h}$. This transformation usually started near the interface seed-ingot close to the edge of the seed. The beginning of the growth process has been investigated. It was shown that inhomogeneous contact seed-graphite holder could deteriorate the quality of the grown ingot. This was especially important at the periphery of the seed where even polycrystalline 6H-SiC growth was observed [1].

Shape of the crystals

Numerical modeling has been used to optimize the design of the crucible for a 25mm crystal diameter. It was found that the crystal shape largely depends on the radial and axial temperature. The crystals of 25 mm in diameter had flat top surface due to a low gradient temperature. On the contrary, the crystals of 45 mm in diameter had convex top surface. No deposition on the guide has been observed. This result has been confirmed by simulation.

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