Optimization of the SiC powder source size distribution for the sublimation growth of long crystal boules

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Abstract

In this work we studied the influence of three different SiC powder size distributions and the sublimation behavior during physical vapor transport growth of SiC in a 3 inch crystal processing configuration. The evolution of the source material as well as of the crystal growth interface was carried out using in-site X-ray computed tomography. Two SiC powders exhibited a single modal D90 size distribution of ca. 50 µm and ca. 200 µm, respectively. In both cases the average SiC powder density was 1.2 g/cm^3. The third powder was a mixture of the above mentioned source materials and exhibited a bimodal particle size distribution. The corresponding average SiC powder density was 1.7 g/cm^3. In this latter case the in-itu X-ray computed tomography study revealed an improved growth interface stability that enabled a much longer crystal growth process. During process time, the sublimation-recrystallization behavior of the mixed SiC powder showed a much smoother morphology change and slower materials consumption as well as much more stable shape of the growth interface than in the case of the less dense SiC source. By adapting the size distribution of the SiC source material we achieved to significantly enhance stable growth conditions. Copyright © 2018 VBRI Press.

Keywords: Silicon carbide, physical vapor transport, source material, in-situ visualization, computed tomography.

Introduction

In recent years silicon carbide (SiC), and in particular the 4H-SiC polytype, has become a standard semiconductor material for power electronic applications. The most common growth technique for the preparation of single crystalline wafer material is the so called physical vapor transport (PVT) method [**1-6**]. Growth is carried out in a closed graphite container at elevated temperatures above 2000°C (up to 2400°C). As source material, SiC powder is placed at the crucible bottom. At the top crucible interior a single crystalline seed is mounted at a slightly lower temperature. The typical temperature between source material surface and SiC seed lies in the range of 50 °C to 100°C. The type of SiC source material, i.e. its powder size distribution and effective packaging density has a significant impact on the temperature field and the growth process itself[**7**]. In particular morphological changes of the SiC powder source material during sublimation growth influence the stability of the growth process in terms of constant growth conditions as well on the ability to grow long boules.

In this work we investigate the optimization of the SiC source material packaging density in order to perform long crystal growth runs that result in long SiC boules. The behavior of the sublimation and recrystallization process is monitored using advanced 3D in-situ computed tomography X-ray visualization [**6**].

Experimental

SiC powder source material

Three different SiC powder size distributions provided by the Industrial Technology Research Institute of Taiwan (samples ITRI-1,-2,-3) were investigated to optimize the source material packaging density. In addition, a technologically well-established SiC reference powder from the University of Erlangen (sample FAU reference) was included into the scientific discussion. Two SiC powders exhibited a single modal D90 size distribution of ca. 50 µm (sample ITRI-1, **Fig. 1a**) and ca. 200 µm (Sample ITRI-2, **Fig. 1b**), respectively. In both cases the average SiC powder density was 1.2 g/cm^3. The third powder (sample ITRI-3, **Fig. 1c**) was a mixture of the above mentioned source materials and exhibited a bimodal particle size distribution. The corresponding average SiC powder density was 1.7 g/cm^3. **Table 1** summarized the properties of the applied SiC powder charges.

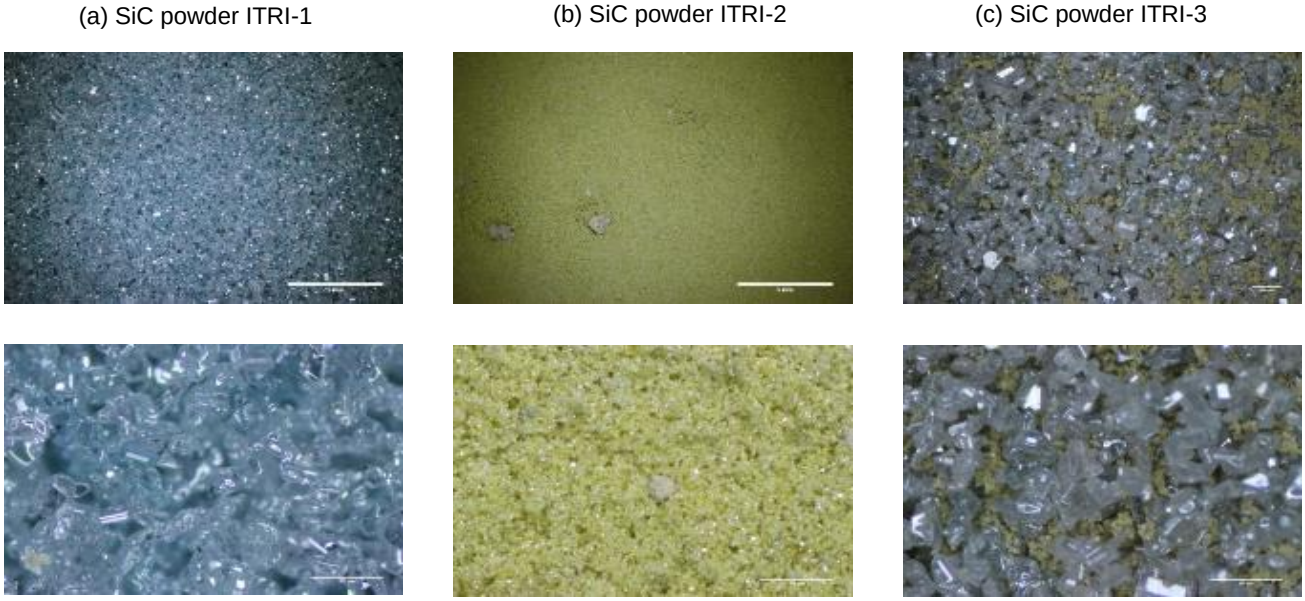

Fig. 1. Optical microscopy images of the three SiC powder sources ITRI-1, ITRI-2 and ITRI-3.

Table 1. Properties of the SiC powder materials during charging of the growth cell.

	SiC powder charge in the 3 inch crucible [g]	SiC powder density [g/cm^3]	Degree of filling
ITRI-1	493	1,18	36,9 %
ITRI-2	485	1,16	36,3 %
ITRI-3	694	1,66	51,9 %
FAU reference	490	1,17	36,6 %

PVT growth of SiC boules

Crystal growth was carried out in an inductively heated PVT setup using a crucible configuration to grow 3 inch SiC boules of up to thickness of ca. 25 mm. The temperature on the top of the growth cell varied between 2030 °C and 2170 °C for the four experiments of this study, respectively. The corresponding crystal growth interface temperatures are ca. 2150°C to 2250°C. Although the growth experiments were carried out with the goal to establish equal process conditions, differences in the thermal SiC source material properties and in addition the degradation of graphite isolation parts from growth run to growth run, caused a variations of the average growth temperature of up to 50°C. The inert gas pressure to control growth rate in diffusion limited growth mode was 20 mbar.

In-situ 3D process visualization

In-situ 3D process visualization of the morphological changes of the SiC source material as well as of the progression of the SiC crystal growth interface were performed using computed tomography [CRAT15]. 2D projections of the crucible interior were extracted from the 3D computed tomography data **(Fig. 2).**

Results and discussion

Fig. 2 shows four series of 2D-projections of the evolution of the SiC powder sources during the growth processes using in-situ 3D computed tomography X-ray visualization. The reference growth run in **Fig. 2a** uses SiC powder which has been developed at FAU and which has been used for SiC PVT growth in recent years as standard material in the own lab. The D90 size distribution lies between 50 µm and 90 µm. The SiC powder consumption as depicted in **Fig. 2a** exhibits a smooth sublimation and re-crystallization behavior in the upper area throughout the process. The condensed top area of the SiC source is formed due to a strong drop of the axial temperature gradient at the interface from the SiC source to the gas room. It remains stable with a large diameter facing towards the growing SiC boule, as it is a prerequisite for the processing of high quality SiC crystal. Below the condensed top area a second SiC powder block of a lower density is evolved which is separated by bigger SiC particles from the top.

To investigate the improvement potential of the SiC source material performance we have investigated two SiC powders with strongly deviating single modal D90 size distributions of 200 µm and 50 µm (SiC powder ITRI-1 and ITRI-2 in **Table 2**) as well as its mixture with a bimodal size distribution and a significantly enhanced packaging density (SiC powder ITRI-3 in **Table 2**).

The SiC powder ITRI-1 (large grains) and SiC powder ITRI-2 (small grains) exhibit sublimation behavior to perform a basic bulk growth process of SiC by the PVT method. Densification in the central powder area ensures stable growth conditions.

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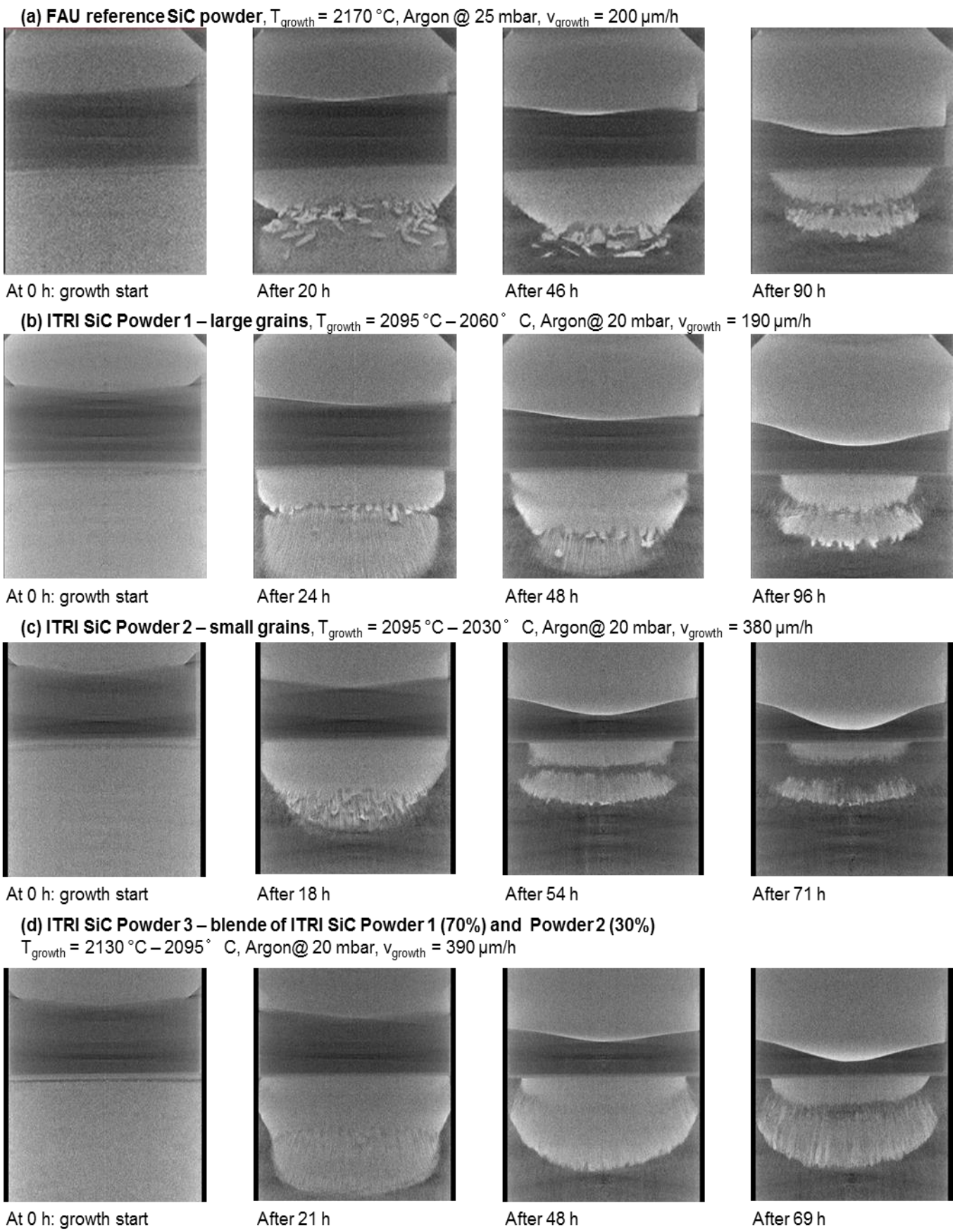

Figure 2. Evolution of the SiC source material and SiC powder source using in-situ computed tomography X-ray visualization of the growth process. The 2D projection images are extracted from the 3D data sets. (a) Reference growth run using the FAU reference SiC powder. (b, c, d) SiC powder optimization study using the SiC powders ITRI-1, ITRI-2 and ITRI-3.

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Immediate consumption of the SiC powder in the periphery of the source material top and side, however, causes a slight temperature field disturbance. I.e. the radial temperature gradient is altered and, hence, the growth interface exhibits an increasing convex shape with increasing growth time. The latter is a source for stress within the grown crystal. As main reason for this immediate SiC powder consumption in the side area of the crucible the low packing density of the SiC powder may be identified. This leads to an enhancement of the sublimation-recrystallization paths at the powder rim and to an immediate SiC powder consumption.

The SiC powders ITRI-1 and ITRI-2 exhibit a slightly more pronounced shrinkage of the condensed SiC source top compared to the FAU reference powder. The nature of the irregular powder grain shape could be considered as origin for the early SiC powder consumption in the crucible rim.

The SiC powder ITRI-2 with the smaller D90 grain size exhibits a much faster overall sintering. The double growth rate using the SiC powder ITRI-2 (small SiC grains) compared to the SiC powder ITRI-1 (large SiC grains) during nominally identical growth process parameters could be partially related significant different values of the SiC source heat conductivity which influences the axial temperature gradient in the growth cell. In the diffusion limited growth regime (inert gas pressure of 20 mbar), however, kinetical sublimation differences can be excluded to a large extend as explanation for the growth rate differences.

SiC powder ITRI-3 which was established as a mixture/blend of the large grain powder ITRI-1 (70 wt%) and the small grain powder ITRI-2 (30 wt%) overcomes the drawbacks of the single powder ITRI-1 and ITRI-2. Densification in the central powder area as well as SiC powder consumption in the periphery ensured stable growth conditions (i.e. homogeneous T-distribution inside the growth cell). In addition, the high packing density enabled the growth of long SiC crystal of ca. 25 mm. The SiC powder ITRI-3 exhibits reasonable sublimation behavior to carry out high quality PVT bulk growth
of SiC!

Conclusion

The SiC powder ITRI-3 with its bimodal SiC powder size distribution exhibits the most favorable growth conditions: In-situ X-ray computed tomography revealed an improved growth interface stability that enabled a much longer crystal growth process. During process time, the sublimation-recrystallization behavior of the mixed SiC powder showed a much smoother morphology change and slower materials consumption as well as much more stable shape of the growth interface than in the case of the less dense SiC source. By adaption of the particle size distribution of the SiC source material the growth process stability was significantly enhanced. The latter is believed to be an important step towards a better polytype stability as well as towards a more constant dopant incorporation during bulk growth of SiC.

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Author's contributions

Conceived the plan: MA, TH, PW; Performed the expeirments: MA; Data analysis: MA, TH, PW; Wrote the paper: PW. Authors have no competing financial interests.

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