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Filling-Design Effect of Powder Source in the Crucible on SiC Single-Crystal Growth

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Abstract. An effective powder consumption is indispensable for enlarging the diameter and thickness of SiC crystals. We employed three types of filling designs for SiC source powder with different distances between the surface of the seed and the source powder. To maintain the shape of the designs, the SiC source powder was heat-treated in an Ar atmosphere at 680 torr within a temperature range of 1500 to 1600°C. The SiC source powder consumption and contribution to growth in well-structured layouts increased due to the increase in the surface area of SiC source powder, despite its lower initial powder filling. The numerical simulation showed that the well-structured layouts with a higher surface area of SiC source powder have a higher partial pressure of Si and SiC_2 gases (supersaturation of these gas phases) near the seed region compared to the without well-structured layouts. The computed tomography (CT) analysis of the cross-section of SiC source powder after the growth run clearly showed that the source powder was previously sublimated at the region of the crucible wall, and recrystallization at the surface region of the source powder physically retarded the pathway of SiC source gases to the region of the SiC seed crystal. The newly designed well-structured layouts of the source powder have an economical advantage in achieving effective powder consumption during crystal growth.

Introduction

SiC single crystal has nearly 3 to 4 times higher thermal conductivity, breakdown voltage, and band gap energy compared to Si. Due to their superior physical properties, SiC has gathered the most attention as a power semiconductor material [1]. Physical vapor transport (PVT) method has more advantage for enlarging the diameter and thickness of SiC crystals compared with solution-base one. However, during the crystal growth, the recrystallization at the center region of SiC source powder occurs due to relatively lower temperature along the radial direction within crucible wall. The region of recrystallization of source powder interrupts sublimation from the SiC sources, such as Si, Si_2C and SiC_2 phases, toward the SiC seed crystal region [2]. This is the reason why the PVT method has the disadvantage of a low contribution to growth and consumption of the SiC source powder compared with other growth methods [3]. Despite these disadvantages of the PVT method, the current SiC wafer market is rapidly growing. Recently, the fabrication of SiC single crystals with 8 and 12-inch diameters using by the PVT method is being actively introduced. To enlarge the diameter and thickness of the SiC crystal, it requires additional time and cost for growth. It is necessary to improve the efficiency of SiC source powder consumption in PVT growth. In this study, we proposed the fabrication process and design of SiC source powder in the graphite crucible to improve the efficiency of SiC powder consumption. 4-inch SiC single crystals were prepared by the PVT method adopted with several designs of the SiC source powder. The surface area of the powder, powder consumption, and contribution to growth, depending on the design of the SiC powder, were investigated before and after the SiC growth.

Experiments

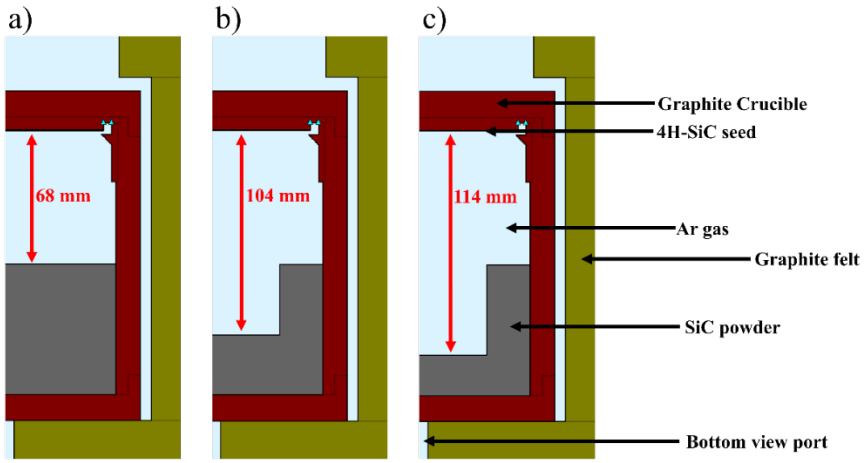

Fig.1. A Schematic diagram of three types of powder filling-design based on VR (Virtual Reactor, *STR*) 8.4: a) Design A with no well-structured layout, b) design B with a 104 mm depth of well-structured layout, and c) design C with a 114 mm depth of well-structured layout.

Figure 1 shows a schematic diagram of three types of powder filling-design in graphite crucible designed by VR (Virtual Reactor, *STR) 8.4* software. The powder filling-design was classified as the distance from surface of the SiC seed crystal to the center of SiC source powder. Designs A, B, and C have distances of 68 mm, 104 mm, and 114 mm, respectively. In design A, physical interference due to powder recrystallization occurs from the formation of an interparticle cavity within the source powder caused by the radial temperature difference between the center (relatively low) and the nearby crucible wall (relatively high). On the contrary, designs B and C with well-structured layouts have more efficient powder consumption due to the increased surface area of the source powder placed near the crucible wall. Figure. 2 exhibits a fabrication process of well-structured layouts of SiC source powder. The fabrication process is as follows: 1) A base powder was filled into the graphite crucible. 2) A small inner graphite crucible was placed on the base powder to form the well-structured layouts. 3) The additional source powder was filled into the empty space of in 2) (between the inner and the outer crucible).

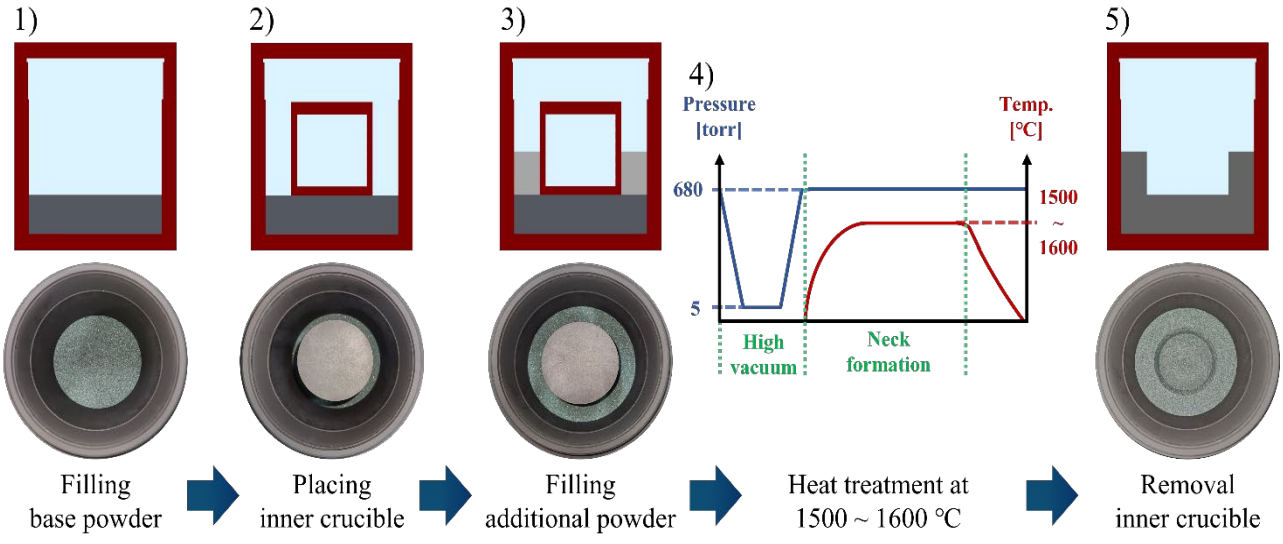

Fig.2. The fabrication process of well-structured layouts of SiC source powder (Design B and C).

4) To sustain the structure, the heat treatment was conducted in an Ar atmosphere at 680 torr in the temperature range of 1500 ~ 1600℃ for 10 hours. Design A did not carry out the heat treatment. In general, heat treatment of a typical SiC source powder is performed at 2000 ~ 2200°C, this temperature range is well known as the sintering temperature for SiC source powder. The sintering process leads to neck formation and the removal of internal pores, resulting in grain growth of the source powder [4]. The pores in the source powder act as paths for SiC source gases such as Si and SiC_2. The grain growth could be directly linked to the improvement of SiC powder sublimation [5], however, the aspect of the SiC crystal could change with the use of different grain sizes of source powder, even under identical growth conditions. Since the neck formation was conducted at relatively lower temperatures between 1500 and 1600°C, the grain growth was suppressed, and the minimal inter-grain bonding maintains the shape of the powder. 5) The inner crucible was removed after the heat treatment. Crystal growth using designs A, B, and C was carried out under identical growth conditions. SiC crystals were prepared in an Ar atmosphere of 5 torr at a temperature of approximately 2300℃ for 50 hours by the physical vapor transport (PVT) method. An n-type 4H-SiC crystal was used as a seed crystal.

Results and Discussions

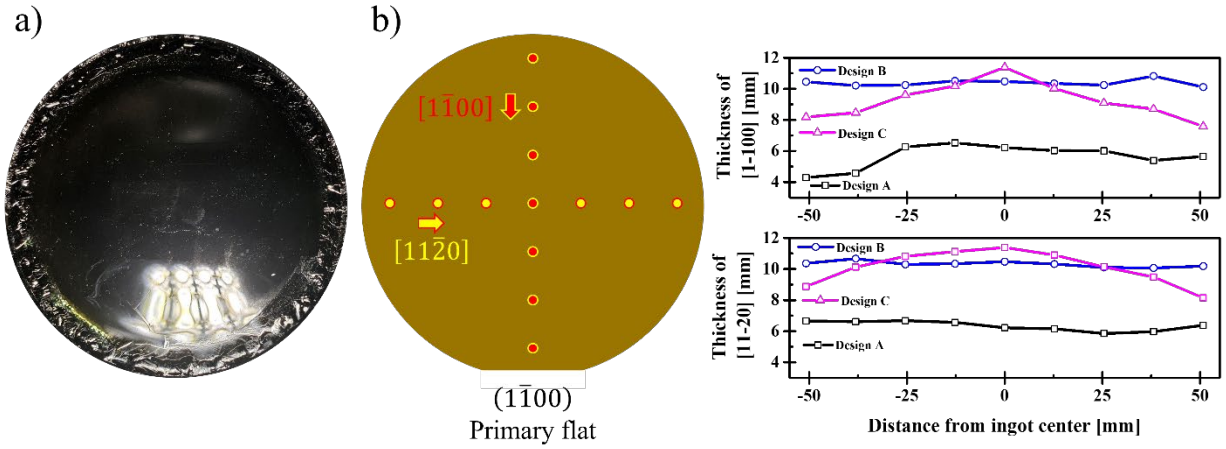

Fig.3. The representative image of the SiC crystal grown using design C a), the measured points of thickness of the SiC crystal b), and the plot of thickness variations along the vertical and horizontal directions on the SiC crystal grown by each design.

4H-SiC crystals with a diameter of about 100 mm were grown. The growth rates for SiC crystals (maximum values) with design A, B, and C were 126 μm/h, 208 μm/h, and 214 μm/h, respectively. Figure 3a) shows the representative image of the SiC crystal grown by design C, and Fig. 3 b) shows the thickness plot along the directions [1-100] and [11-20] based on the primary flat of the SiC crystal grown using each design. The seven points of thickness were obtained along each direction. The thickness of SiC crystals grown using designs B and C is definitely higher than that of design A. The thickness range (mean thickness) was 4.3 ~ 6.7 mm (5.5 mm) for design A, 10.1 ~ 10.7 mm (10.4 mm) for design B, and 7.6 ~ 11.4 mm (9.5 mm) for design C, respectively. The SiC crystal adoption with designs A and B shows almost a flat surface; however, that of design C shows a convex shape. Design C, with a deeper well depth (114 mm) shows a higher contribution to growth compared with design B (104 mm). This is directly related to an increase in the surface area of the powder. The factor for crystal growth of 4H-SiC crystals using designs A, B, and C is as shown in Table 1. Here, the powder consumption [%] and contribution of growth [%] were calculated by the weight usage of powder/total weight of powder and the weight of crystal/weight usage of powder, respectively. As shown in Fig. 4 a), the powder consumption increases with an increase in their surface area, even decreasing in the filling powder. Design A has a higher filling powder of 1,202.6 g in the crucible and a higher contribution to growth of 98% after crystal growth. This means the SiC source powder was almost consumed and contributed to the growth. However, interestingly, the powder surface area

increases in designs B and C which have well-structured layouts compared to design A which does not have well-structured layout as shown in Fig. 4 b). Design A has a lower growth rate compared with that of the other designs. In contrast, designs B and C have a higher growth rate than design A, even though they have lower filling powder and contribution to growth as shown in Fig. 4 c).

Table 1. The factors for crystal growth depending on each design.

Factors for crystal growth	Design A	Design B	Design C
Filling powder [g]	1,202.6	919.9	845.1
Surface area of powder [mm^2]	2,704 π	4,624 π	5,264 π
Powder consumption [%]	20	29	39
Growth rate [μm/h]	126	208	214
Contribution to growth [%]	98	88	90

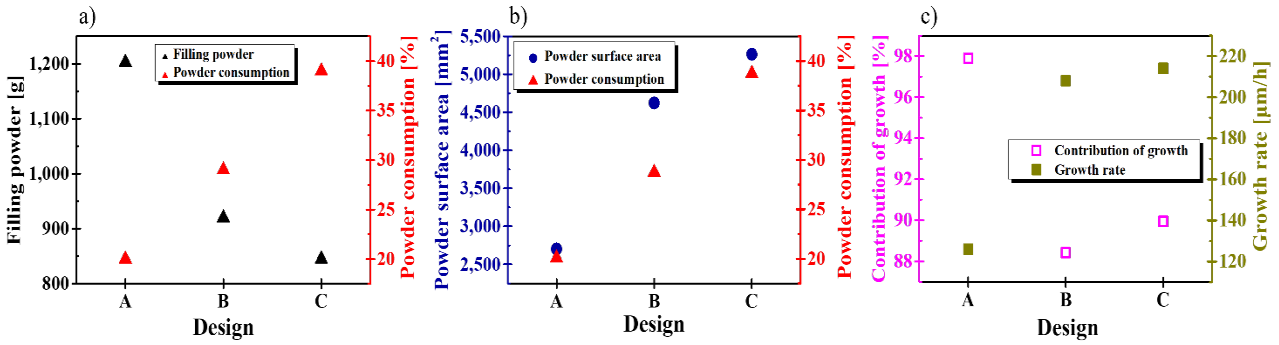

Fig. 4. The plot of filling powder a) and surface area b) as functions of powder consumption depending on design. c) The plot of contribution to growth as a function of growth rate for each design.

In the crystal growth of SiC, one of the most important parameters is the supersaturation of sources near the seed region. Supersaturation with an appropriate level of SiC source allows for the stable growth of SiC single crystals. Chen et al. [6] studied the relationship between supersaturation and growth rate depending on growth pressure. Lower growth pressure leads to an increase in supersaturation; as a result, the growth rate increases. In general, the degree of supersaturation is about 0.0075 torr or less when the growth pressure is in the range of 60 ~ 105 torr. Since crystal growth was performed at a growth pressure of 5 torr in this study, the degree of supersaturation increases as more energy is needed for the phase transition from solid to gas. This is why a sufficient amount of SiC source powder should be consumed to create supersaturation in the seed region. As a result, designs B and C have higher powder consumption of 29% and 39%, respectively; however, design A has lower powder consumption (20%) due to lower supersaturation, despite its higher contribution to growth. Figure 5 shows the partial pressure distribution of P_{Si} and P_{SiC_2} gas phases and SiC source gas flow based on the crucible calculated by VR 8.4 software. In the crucible, the partial pressure of designs B and C was higher than that of design A, as shown in Fig. 5 (a) ~ (c). Figures 5 d) and e) show the partial pressure of P_{Si} and P_{SiC_2} in the nearby seed region for each design. Since designs B and C have higher partial pressure than design A, they show the same trend of

increased powder consumption depending on their powder surface area, as mentioned above at Fig. 4 b). Wang et al. [7] explained this behavior toward different partial pressures in the source powder by using Darcy's law and Van Goff's equation, which are related to the momentum equation and the driving force of gas phase shifting from a given point within the powder to the powder surface.

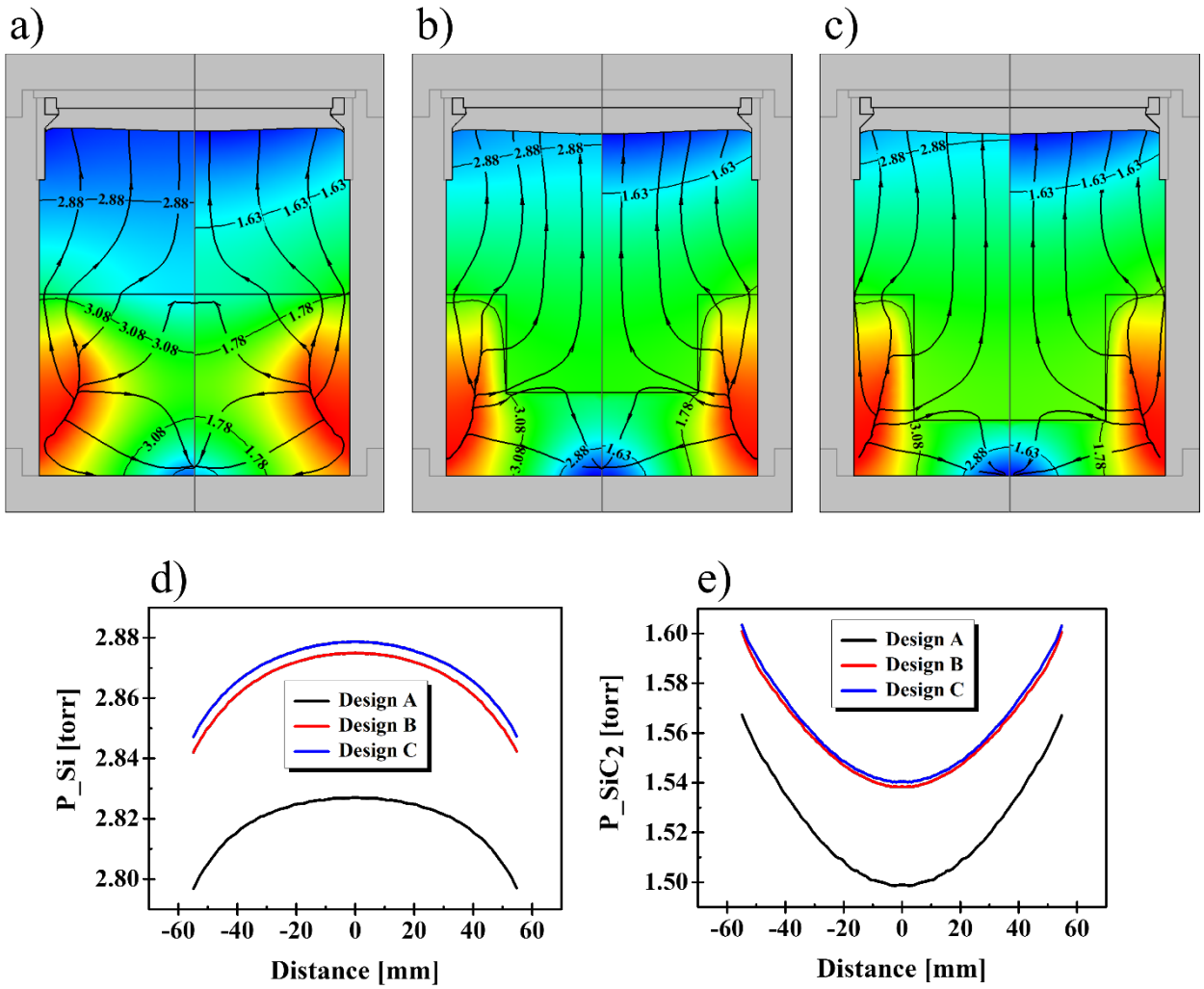

Fig. 5. The numerical simulation of partial pressure and SiC source gas flow for Si and SiC_2 gas phases. The half of left and right of crucible for each design A, B, and C (a, b, and c) show the partial pressure of P_{Si} and P_{SiC_2}, respectively. The partial pressures of Si d) P_{Si} and SiC_2 e) P_{SiC_2} are plotted in the nearby seed region.

According to their research, since the temperature of the powder at the region of the crucible wall is consistently higher compared to that of the central region, the partial pressure is also high. However, the temperature of the powder at the surface region is relatively lower and the partial pressure is lower as well. This means that sublimation of the SiC source can theoretically occur through the surface of the given powder. The flow of SiC source gases (flux) is represented by the black arrows in Fig. 5 (a) ~ (c). As a result, both the powder consumption and the partial pressure of the SiC source around the seed region increase with the surface area of the powder. Figure 6 shows the cross-sectional photographic images and CT (Computed Tomography) images of the remaining source powder after growth run. We found that the height of the source powder for all designs increases after growth run due to surface recrystallization. The final increase in height of the center region was 2 ~ 5 mm for design A, 2 ~ 12 mm for design B, and 15 mm for design C, respectively. In design A, the sublimation of the source powder occurred at the crucible wall, which had a relatively higher temperature, resulting in only porous carbon powder remaining (as shown in Fig. 6 b). This means source gases

can be previously sublimated at the crucible wall region. At the same time, source gases at the center of the powder were recrystallized on powder surface. The center region of the source powder is likely to recrystallize because this region is relatively lower than the other regions in the crucible. The concept of large sublimation of the source powder near the crucible wall and surface recrystallization of the center region of the source powder could be applied in designs B and C.

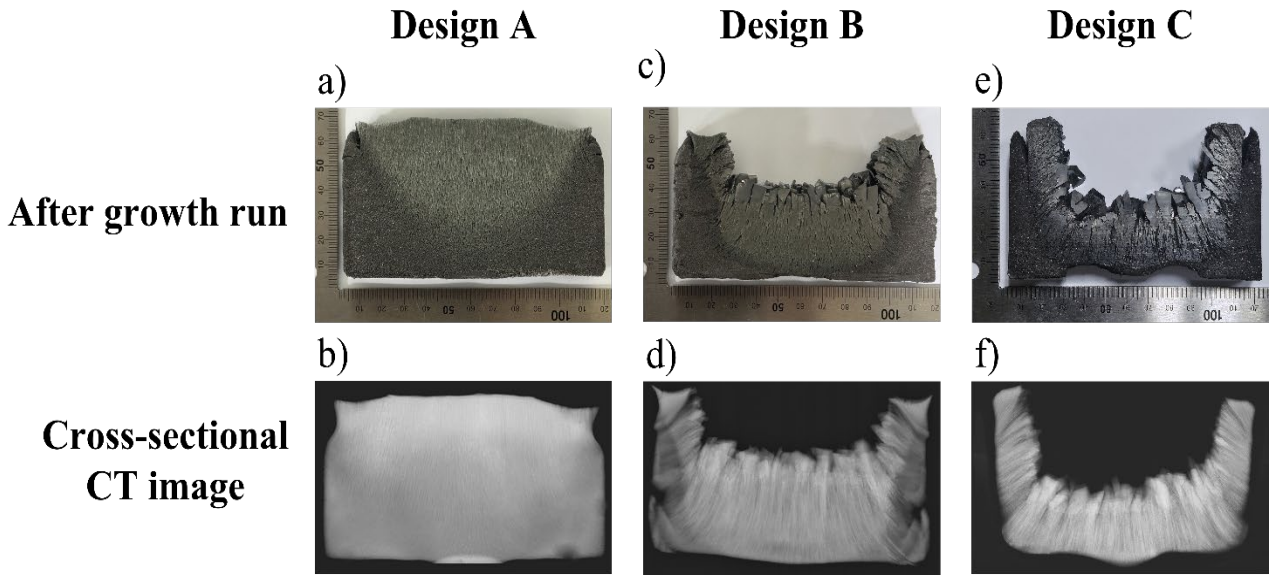

Fig. 6. The photographic images a), c), and e) and CT (Computed Tomography) images b), d), and f) of the remaining source powder after the growth run adopting with designs A, B, and C.

We can easily observe the path of source gases in the CT images for designs B and C despite the presence of a large recrystallization region within the source powder after growth, as shown in Fig. 6 d) and f). This leads to effective powder consumption both at the center of the powder and near the crucible. As a result, design C has an economic advantage in achieving higher powder consumption and a higher growth rate of crystals, even with its lower filling of powder, because the powder surface area is the highest. This increases the volume of source gases from the SiC source powder to the seed region. Sufficient supersaturation can form near the seed region, resulting in thicker SiC crystals.

Summary

We prepared the 4-inch 4H-SiC crystals using three types of SiC source powder filling-design with well-structured layouts. The consumption of SiC source powder and the contribution to growth at the well designs increased due to the increase in the surface area of the SiC source powder, despite its lower initial powder filling. The numerical simulation revealed that designs B and C, which have well-structured layouts, exhibit a higher surface area of SiC source powder and a higher partial pressure (higher supersaturation at the seed region) of Si and SiC_2 gases near the seed region. The CT images for design A, which lacks a well-structured layout, showed that the source powder was previously sublimated at the region of the crucible wall, leading to recrystallization at the surface of the source powder due to variation in radial temperature. However, the path for source gases during grain growth can be clearly observed in the CT images for designs B and C, despite the presence of a large recrystallization region within the source powder. This leads to effective powder consumption both at the center of the powder and near the crucible. The new design with a well-structured layout for the source powder paves the way to achieving effective powder consumption in SiC crystal growth.

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References

[1] P. J. Wellmann, Review of SiC crystal growth technology, *Semicond. Sci. Technol.* 33 (2018) 103001

[2] X. Liu, B. Y. Chen, L. X. Song, E. W. Shi, Z. Z. Chen, The behavior of powder sublimation in the long-term PVT growth of SiC crystals, *J. Crys. Growth.* 312, 9 (2010) 1486-1490

[3] H. Norihiro, et al., High-speed, high-quality crystal growth of 4H-SiC by high-temperature gas source method, *Appl. Phys. Express.* 7 (2014) 065502

[4] T. Hidehiko, Silicon carbide powder and sintered materials, *J. Ceram. Soc. Japan*. 119, 3 (2011) 218-233

[5] I.G. Yeo, et al., The quality investigation of 6H-SiC crystals grown by a conventional PVT method with various SiC powders, *TEEM.* 11, 2 (2010) 61-64

[6] Q. Chen, J. Yan, V. Prasad, Application of flow-kinetics model to the PVT growth of SiC crystals, *J. Crys. Growth*. 303 (2007) 357-361

[7] X. Wang, D. Cai, H. Zhang, Increase of SiC sublimation growth contribution by optimizing of powder packaging, *J. Crys. Growth.* 305 (2007) 122-132