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Improvement of the thermal design in the SiC PVT growth process

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ABSTRACT

The physical vapor transport (PVT) method is used to grow silicon carbide (SiC) crystals, which are difficult to be grown by other methods. In this paper, a field-coordination theory is involved to optimize the SiC PVT growth process. By using a finite volume-based computational method, we calculate the flow field as well as species concentration field before and after improvement of the thermal design, respectively. The shape of the SiC crystal grown using the improved thermal design is also shown.

1. Introduction

The main method of growing SiC bulk is the PVT method at temperatures above 2000 °C, which involves many important physical phenomena, such as electromagnetic induction, mass and heat transfer, chemical reactions and so on. Simulation of PVT growth process is a useful tool for optimizing the thermal design and improving the crystal growth procedure. Researchers have done significant works on modeling and simulation of SiC crystal PVT growth. Hofmann et al. [1,2] calculated the temperature distribution and used the simulation technique to investigate the heat and mass transfer in the SiC single crystal growth process for the first time. Lilov [3] indicated the main components of the evaporation of SiC are Si, SiC_2 and Si_2C. The effect of the other components of evaporation in the vapor is insignificant. Muller et al. [4,5] simulated the temperature distribution in the growth system, it is shown that the temperature in the powder is highly non-uniform and the radial variations of 30–50 K were observed along the powder surface. Chen et al. [6–10] developed a 2-D coupled flow-kinetics model for the PVT growth of SiC crystal. The model couples calculations of the 2-D gas flow and the growth kinetics at the crystal interface. The species equation is used to simulate species transfer taking into account convection and diffusion and the growth rate is supposed to be determined by system temperature, temperature gradient and inert gas pressure.

Although a lot of work has been done on modeling of growth processes, there is still no theories on how to optimize the growth processes. In this paper, a field-coordination theory is involved to optimize the SiC PVT growth system. The flow field and species concentration field as well as growth rate profile are calculated after the thermal design is improved.

2. PVT growth method of SiC

The schematic of a typical PVT SiC growth system is shown in Fig. 1. The graphite crucible is filled with SiC powder charge at the bottom, and a SiC seed is placed at the top of the crucible. The SiC powder charge is heated by the RF induction heating, then the powder sublimates and transport to the SiC seed and crystallizes at the seed surface. At the seed surface, it can be divided into two zone, the crystal zone and non-crystal zone. At the non-crystal zone, the mass transfer of SiC species should be restrained in order to limit the growth of polycrystalline SiC.

The coupled flow-kinetic model composed of calculations of the multi-phase flow, mass transfer and the growth kinetics. We assume that the fluid in the crucible is Newtonian and incompressible, and the buoyancy effect is negligible. The Navier–Stokes equations are used to describe the flow in the graphite crucible

$$\nabla\cdot(\overline{\rho}\mathbf{v})=0, \quad (1)$$

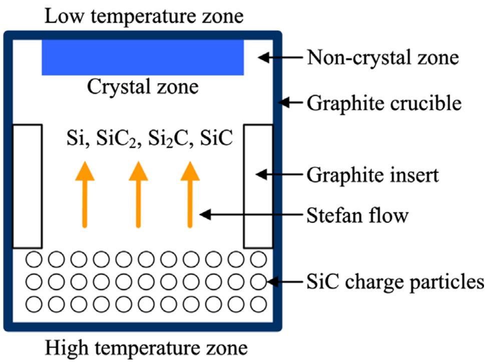

Fig. 1. Schematic of the SiC growth chamber.

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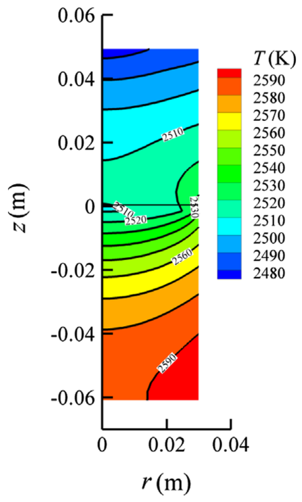

Fig. 2. Temperature field in the SiC growth chamber.

$$\frac{\partial \bar{\rho} u}{\partial t} + \nabla\cdot(\bar{\rho}\mathbf{v}u) = -\frac{\partial p}{\partial r} + \mu\nabla^2 u - \mu\frac{u}{r^2}, \tag{2}$$

$$\frac{\partial \bar{\rho} v}{\partial t} + \nabla\cdot(\bar{\rho}\mathbf{v}v) = -\frac{\partial p}{\partial z} + \mu\nabla^2 v, \tag{3}$$

$$\frac{\partial c_A}{\partial t} + \nabla\cdot(\mathbf{v}c_A) = D_{AB}\nabla^2 c_A, \tag{4}$$

where A, B denote the reaction gas and inert gas (Ar), respectively. D_{AB} is a binary diffusion coefficient, it is approximately given by $D_{AB} = D_0(T/T_0)^n(p_0/p)$.

The boundary conditions are described as the Hertz–Knudsen equation at the seed

$$\left(uc_A - D_{AB}\frac{\partial c_A}{\partial z}\right)\Big|_{z=L} = \chi_A(p_A(L) - p_A^*(L)), \quad \left(uc_B - D_{BA}\frac{\partial c_B}{\partial z}\right)\Big|_{z=L} = 0.$$

At the SiC charge

$$p_A|_{z=0} = p_A^*(T_{max}), \quad \left(uc_B - D_{BA}\frac{\partial c_B}{\partial z}\right)\Big|_{z=0} = 0.$$

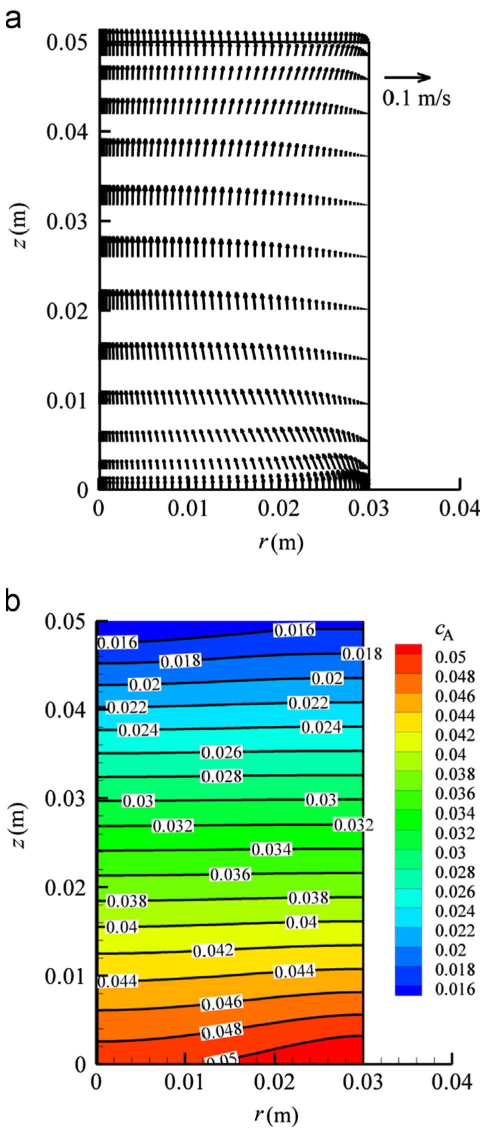

Fig. 3. (a) Flow and (b) concentration fields before improvement of the design at a pressure of 3 kPa.

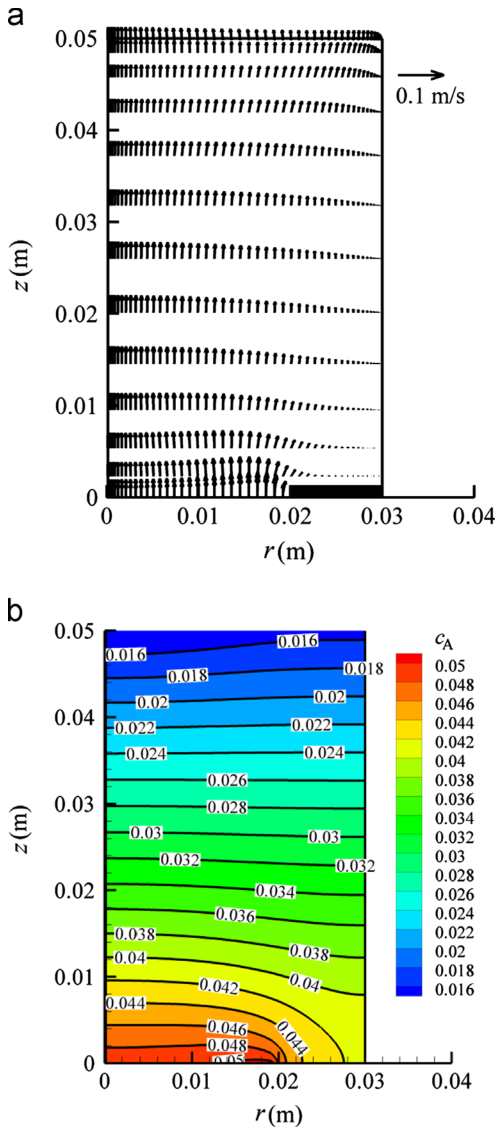

Fig. 4. (a) Flow and (b) concentration fields after improvement of the design at a pressure of 3 kPa.

At the graphite insert above the powder charge

$$u = v = 0, \quad \frac{\partial c_A}{\partial z} = 0.$$

At the axis and the wall of the crucible, the boundary conditions are:

$$\frac{\partial u}{\partial r}\Big|_{r=0} = 0, \quad \frac{\partial c_A}{\partial r}\Big|_{r=0} = 0, \quad \frac{\partial p}{\partial r}\Big|_{r=0} = 0,$$

$$u|_{r=R} = v|_{r=R} = 0, \quad \frac{\partial c_A}{\partial r}\Big|_{r=R} = 0, \quad \frac{\partial^2 p}{\partial r^2}\Big|_{r=R} = 0.$$

3. Numerical simulation and optimization

The field-coordination theory proposed by Guo et al. [11,12] indicates that decreasing the angle between the velocity and temperature gradient vectors is a significant way to enhance the heat transfer. On the contrary, increasing the angle between the velocity and temperature gradient vectors could weaken the heat transfer. This theory could also be used for the mass transfer. We could decrease the angle between the velocity and concentration gradient vectors to enhance the mass transfer and increase the angle between the velocity and concentration gradient vectors to weaken the mass transfer. In this paper, the improvement is achieved by inserting a graphite insulation above the power source. The effect of the graphite insert on the shape of the SiC crystal growth face will be discussed.

We set up an axi-symmetric model for the PVT growth system, and calculate the temperature field in the crucible. The current in the coil is set as 1100 A and the induction frequency is set as 10 kHz. The temperature difference between the edge region and central axis as well as the temperature difference between SiC powder surface and seed surface can be observed in Fig. 2. We assume that the thermal field does not change after inserting the graphite insulation above the powder charge.

We calculate the flow field and species concentration field when the system pressure is set as 3 kPa. The flow and concentration field before improvement of the thermal design are presented in Fig. 3a and b. There is a boundary layer of velocity along the inner surface of the crucible. The directions of the velocity vector and concentration gradient vector are almost the same, the SiC crystal growth would be enhanced at the non-crystal zone. The flow and concentration fields after improvement

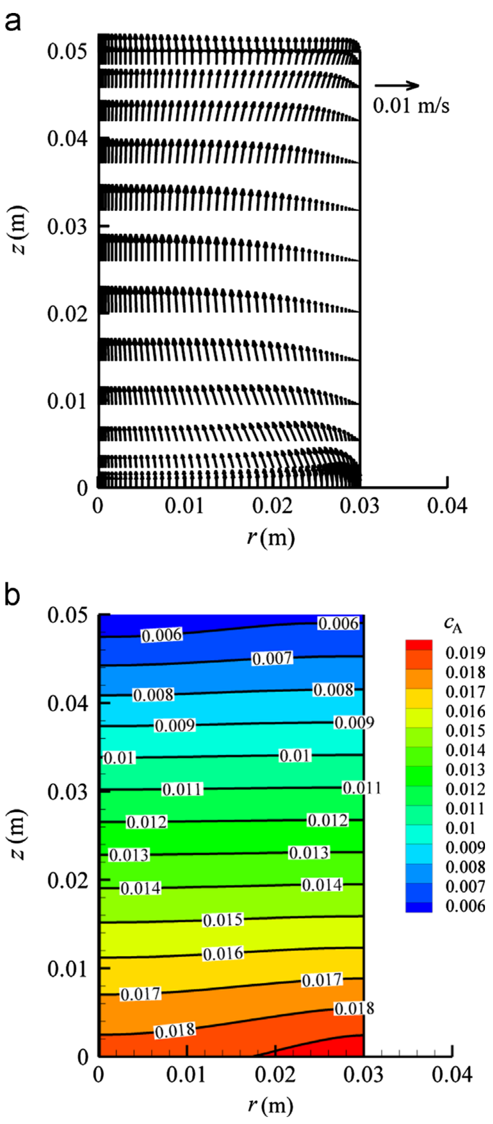

Fig. 5. (a) Flow and (b) concentration fields before improvement of the design at a pressure of 8 kPa.

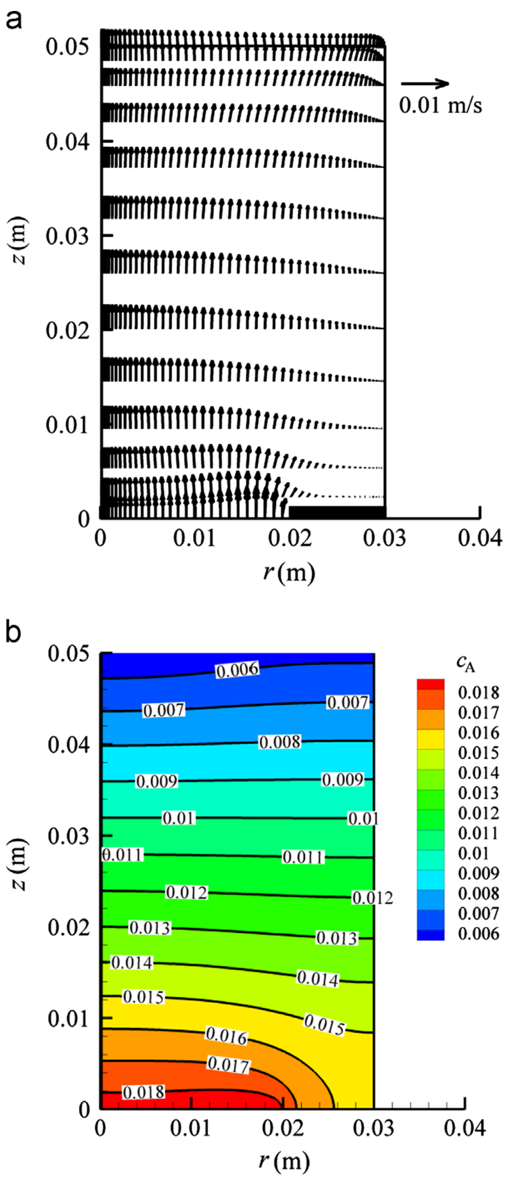

Fig. 6. (a) Flow and (b) concentration fields after improvement of the design at a pressure of 8 kPa.

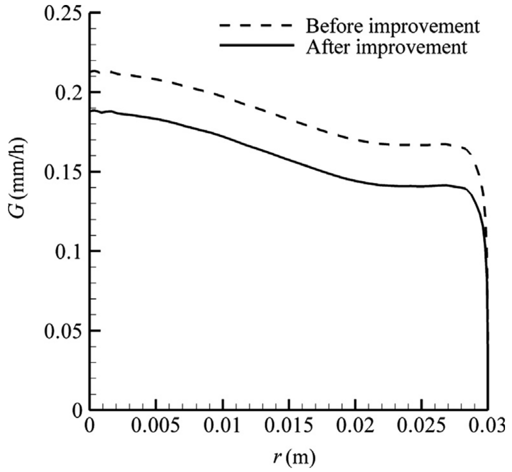

Fig. 7. Profiles of the SiC growth rate before and after improvement of the design at a total pressure of 3 kPa, dash line—before improvement, solid line—after improvement.

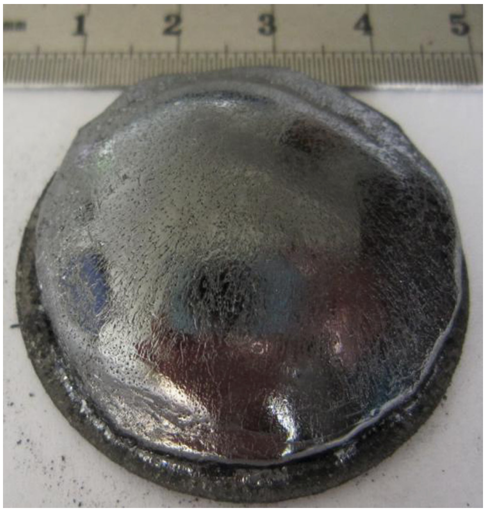

Fig. 8. The shape of the grown crystal interface after improvement of the thermal design.

are presented in Fig. 4a and b. At the non-crystal zone, the angle between the velocity vector and concentration gradient vector increases slightly, so the growth of polycrystalline would be restrained.

Furthermore, we calculate the flow and species concentration fields (Fig. 5a and b) at induction frequency of 10 kHz, current of 1100 A and pressure of 8 kPa. The species concentration and flow field are significantly influenced by the total system pressure. The flow and concentration fields after improvement of the thermal design are presented in Fig. 6a and b.

Fig. 7 shows profiles of the growth rate on the SiC crystal surface before and after improvement of the thermal design. By inserting a graphite insulation above the powder charge, the flow velocity is enhanced at the axis while restrained at the edge of the crystal. The growth rate decreases slightly due to blocking of the vapor by the graphite insulation above the powder source. However, the shape of the growth rate profile improves after inserting the graphite insulation. An appropriate temperature difference between the edge and the center of the crystal causes a convex crystal surface. The crystal surface changes to a concave shape when the temperature difference is negative. An appropriate growth rate of SiC crystal could be obtained by controlling the total system pressure.

Fig. 8 shows the shape of the grown crystal interface after improvement of the thermal design. The shape of the crystal interface is improved, thus the yield of the crystal growth can be improved and the cost of growing high-quality SiC crystals can be reduced.

4. Conclusion

A field-coordination theory is involved to optimize the SiC PVT growth system. We calculate the flow field as well as species concentration field before and after improvement of the thermal design. By inserting the graphite insulation above the powder source, the mass transfer can be improved in the crystal zone. We also performed growth experiments to verify the field-coordination theory. The yield of crystal can be increased by improving the shape of the crystal growth interface. The effect of the graphite insert inside the crucible on the heat transfer will be studied in the future.

Acknowledgments

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References

[1] D. Hofmann, M. Heinze, A. Winnacker, F. Durst, L. Kadinski, P. Kaufmann, Y. Makarov, M. Schafer, On the sublimation growth of SiC bulk crystals: development of a numerical process model, Journal of Crystal Growth 146 (1995) 214–219.
[2] D. Hofmann, R. Eckstein, M. Kolbl, Y. Makarov, G. Muller, E. Schmitt, A. Winnacker, R. Rupp, R. Stein, J. Volkl, SiC-bulk growth by physical-vapor transport and its global modelling, Journal of Crystal Growth 174 (1997) 669–674.
[3] S.K. Lilov, Study of the equilibrium processes in the gas phase during silicon carbide sublimation, Materials Science and Engineering 21 (1993) 65–69.
[4] G. Muller, R. Eckstein, D. Hofmann, L. Kadinski, P. Kaufmann, M. Kolbl, E. Schmitt, Modelling of the PVT-SiC bulk growth process taking into account global heat transfer, mass transport and heat of crystallization and results on its experimental verification, Materials Science Forum 264–268 (1998) 57–60.
[5] G. Muller, R.C. Glass, H.M. Hobgood, V.F. Tsvetkov, M. Brady, D. Henshall, J.R. Jenny, D. Malta, C.H. Carter Jr., The status of SiC bulk growth from an industrial point of view, Journal of Crystal Growth 211 (2000) 325–332.
[6] Q.S. Chen, H. Zhang, V. Prasad, C.M. Balkas, N.K. Yushin, S. Wang, Kinetics and modeling of sublimation growth of silicon carbide bulk crystals, Journal of Crystal Growth 224 (2001) 101–110.
[7] Z.B. Zhang, J. Lu, Q.S. Chen, V. Prasad, Thermoelastic stresses in SiC single crystals grown by the physical vapor transport method, Acta Mechanica Sinica 22 (2006) 40–45.
[8] J. Lu, Z.B. Zhang, Q.S. Chen, Numerical simulation of the flow field and concentration distribution in the bulk growth of silicon carbide crystals, Journal of Crystal Growth 292 (2006) 519–522.
[9] Q.S. Chen, J.Y. Yan, V. Prasad, Application of flow-kinetics model to the PVT growth of SiC crystals, Journal of Crystal Growth 303 (2007) 357–361.
[10] Qisheng Chen, Yanni Jiang, Junyi Yan, Ming Qin, Progress in modeling of fluid flows in crystal growth processes, Progress in Natural Science 18 (2008) 1465–1473.
[11] Z.Y. Guo, D.Y. Li, B.X. Wang, A novel concept for convective heat transfer enhancement, International Journal of Heat and Mass Transfer 41 (1998) 2221–2225.
[12] Z.Y. Guo, Mechanism and control of convective heat transfer-coordination of velocity and heat flow fields, Chinese Science Bulletin 46 (2001) 596–599.