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Design of a high temperature chemical vapor deposition reactor in which the effect of the condensation of exhaust gas in the outlet is minimized using computational modeling

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ABSTRACT

Tetramethylsilane (TMS) was recently proposed as a safe precursor for SiC single crystal growth through high temperature chemical vapor deposition (HTCVD). Because the C content of TMS is much higher than Si, the exhaust gas from the TMS-based HTCVD contains large amounts of C which is condensed in the outlet. Because the condensed C close to the crystal growth front will influence on the thermodynamic equilibrium in the crystal growth, an optimal reactor design was highly required to exclude the effect of the condensed carbon. In this study, we report on a mass/heat transfer analysis using the finite element method (FEM) in an attempt to design an effective reactor that will minimize the effect of carbon condensation in the outlet. By applying the proposed reactor design to actual growth experiments, single 6H–SiC crystals with diameters of 50 mm were successfully grown from a 6H–SiC seed. This result confirms that the proposed reactor design can be used to effectively grow 6H–SiC crystals using TMS-based HTCVD.

1. Introduction

Silicon carbide (SiC) is a promising wide band gap material that can be used as the single crystal substrates for high performance power devices, since it has a high-heat resistance, high-thermal conductivity and can withstand high voltages [1,2]. To fabricate SiC single crystal substrates, physical vapor transport (PVT) is currently widely used as a commercialized technique for growing bulk crystals of SiC [3,4]. PVT has contributed significantly to the recent progress of the scale-up in the SiC crystal wafer.

However, the PVT method is basically based on a closed system, which means that the process conditions continuously change during crystal growth. As an alternative a growth technique in which SiC is used in conjunction with high temperature chemical vapor deposition (HTCVD) was proposed, because, using HTCVD, it is theoretically possible to obtain constant crystal quality along the growth direction [5–8].

Conventionally, HTCVD involves the use of SiH_4, which is explosive, and a hydrocarbon as the Si and C sources, respectively. However, in a previous study, we proposed a novel HTCVD process in which a safer precursor, namely tetramethylsilane (TMS, $Si(CH_3)_4$) was used as a source of both Si and C, rather than SiH_4 and hydrocarbons [9–11]. The outlets of the HTCVD reactor could become closed by the condensation of unreacted vapor phases in the exhaust gas, which constitutes a significant issue, since long-term crystal growth can be interrupted when SiH_4 and a hydrocarbon are used [3,4]. Since TMS contains much more carbon than silicon in its molecule, the outlet closing issue could be even more severe in the case of TMS-based HTCVD [12].

The goal of this study, therefore, was to design an effective reactor in which the effects of carbon condensation are minimized. To accomplish this, carbon condensation in a reactor was thermodynamically investigated based on the geometric locations and temperature distribution. Multiphysics simulations were then carried out, based on computational fluid dynamics with the intent of proposing a design for the efficient, long-term crystal growth of SiC. Finally, the proposed reactor design was applied to actual experimental crystal growth to produce high quality SiC bulk crystals.

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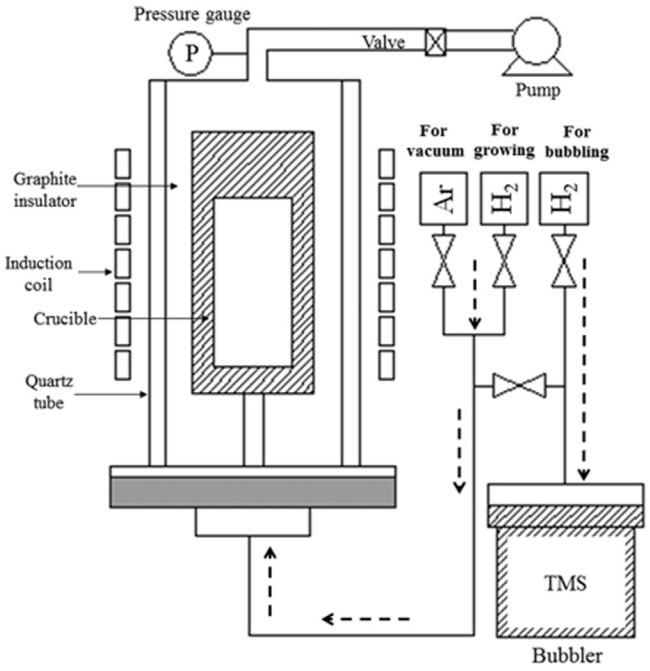

Fig. 1. Schematic of a high temperature chemical vapor deposition system employing tetramethylsilane as the precursor.

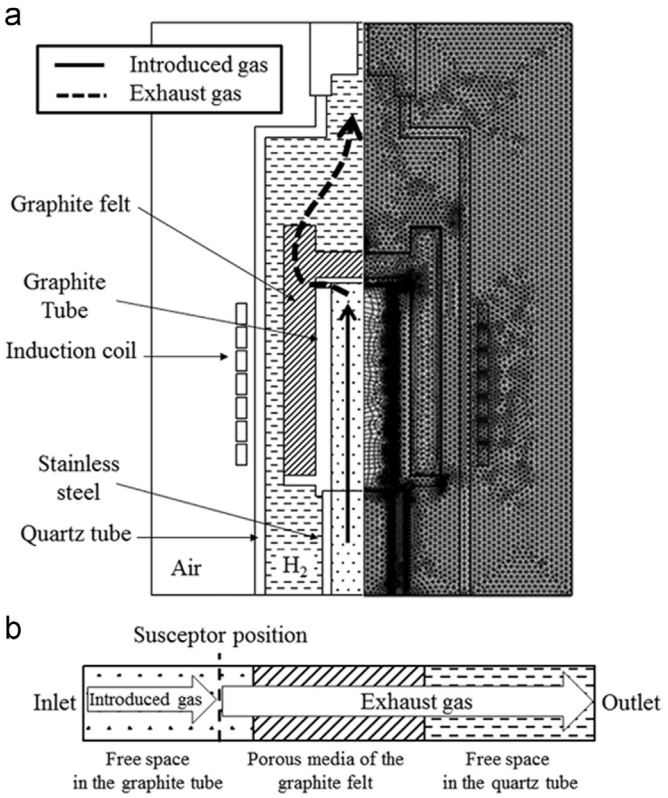

Fig. 2. (a) Modeling of HTCVD system with a meshed model for finite element analysis. (b) Distinguished gas flow in the HTCVD system depending on the media.

2. Process and model description

Fig. 1 shows a schematic of HTCVD reactor used in this study. A conventional vertical hot-wall reactor was used to grow bulk crystals on 6H–SiC seeds. The quartz tube and the RF coils were water-cooled. The crucible inside the quartz tube was thermally insulated with graphite felt. The base and working pressure were set at 76 mTorr and 550 Torr, respectively. The precursor TMS was introduced to the reactor by the carrier gas H_2. The Si/H ratio was fixed at 6.2×10^{-4} as determined by thermodynamic modeling as described in the literature [9]. In a conventional vertical reactor, the growth experiments were conducted at 2000 °C for 2 h. 6H–SiC single crystalline samples were used as seed crystals for each test.

Several simulation techniques were employed in the computational analysis of the TMS-based HTCVD, which included a computational thermodynamic analysis using the Factsage™ 6.4 software with the FactPS database [13] and mass/heat transfer was calculated by means of a finite element analysis (FEA) using the COMSOL Multiphysics 4.3 a software [15]. Details of the calculations for thermodynamic analysis of HTCVD can be found in the literature [9]. The FEA model for HTCVD was designed as axisymmetric 2 dimensional, as shown in Fig. 2a. All the materials properties used in the computation are shown in Table 1. According to Lenz's law, a time-dependent source current along RF coils induces eddy currents in conductive materials which are heated by a Joule effect. The electromagnetic field distribution was computed using the Maxwell equation. The RF frequency and heating time were fixed at 12.5 kHz and 1 h, respectively.

Heat transfer in solids involves conductive heat transfer in all solid components and radiation heat transfer on the all heated surfaces in the quartz tube. The radiative heat exchange is modeled on the basis of the assumption of surface to surface radiation [14]. The temperature of the outer wall of the quartz tube was set to room temperature considering the cooling conditions. The introduced gas passes through the graphite tube and is then condensed on the seed crystal. The exhaust gas from the graphite tube passes through the graphite felt and the quartz tube, as illustrated in Fig. 2b. Because the gas flow in porous media like graphite felt is quite different with that in the free space, we divided the gas channel depending on the flowing media which was classified as shown in Fig. 2b. Hence, the fluid velocity and pressure fields of the TMS/H_2 gas feeding chamber were evaluated using free and porous media interface which were provided by the COMSOL Multiphysics software packages [15]. The free and porous media interface is applicable to a flow passing through free channels and porous media such as graphite felt. The flow in the free channel was described by the Navier–Stokes equations taking non-compressible laminar flow into account, while the flow in the porous media was evaluated by the Brinkman equation, as described in the literature [16]. The heat transfer in fluid was evaluated on the assumption of steady state problem. The simulations were conducted using fully-coupled solver.

3. Results and discussion

3.1. Carbon condensation in the TMS-based HTCVD

As reported in the literature, the process conditions for HTCVD using TMS as the precursor were successfully achieved in the growth experiments for 1 h [10,11]. However, the process conditions for the crystal growth of SiC did not persistent for growth times longer than 2 h because the carbon concentration on the growth front steadily increased in the HTCVD, when TMS was used [12]. The increase of the carbon concentration brings about a change in the thermodynamic conditions. For the thermodynamic transition mechanism in TMS-based HTCVD process, see Ref. [12] in details [12]. Fig. 3a schematically presents the change of the thermodynamic conditions as increasing growing time. In the