Computer Simulation of the Preparation of C/SiC Composites in the F-CVI Reactor

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Abstract

Computer simulation for the preparation of C/SiC composites from methyl-trichlorosilane and hydrogen by F-CVI (Forced-flow Chemical Vapor Infiltration) reactor was studied. The mathematical modeling for the actual processes was carried out.

With one overturning, the process time could be continued for 50 min longer than that without overturning. Additionally, the total amount of deposition increased 1.4 times with several times of overturning. Additionally, effects of many operation parameters were observed for the process with four overturnings. Effects of the flow rate, the initial porosity, and the inlet concentration were observed.

Key words: CVI, C/SiC composites, F-CVI, Computer simulation

Introduction

Ceramic composites are paid a lot of attention to in the several industrial fields recently. That is because shortcomings of each ceramic component can be compensated by being composite materials. Their shortcomings can be improved by reinforcing with fibers and making composites by Chemical Vapor Infiltration (CVI). Fibers absorb energy when ceramic composites are elongated and broken.

CVI was originated in efforts to densify porous graphite bodies by infiltration of carbon¹. In CVI, the precursor gas diffuses into a porous perform, and reacts at pore walls, and deposits matrix materials². Ideally, the result is a dense composite. It can produce large pieces of composites with complex shape.

Manufacturing and modeling of fiber-reinforced ceramic composites by CVI were studied by many researchers³–⁶. Manufacturing ceramic composites reinforced with multilayer woven fabrics and its mathematical modeling were studied by Chung, et al.⁷–¹⁰. Reactant gases diffuse into the three void regions and react to produce solid deposit. The numerical simulations were used to optimize parameters of the CVI process¹¹,¹².

The objective of this work is modeling the preparation of fiber reinforced SiC/C composites by F-CVI of SiC from methyltrichlorosilane (MTS) and H₂. Effects of several variables of infiltration reaction could be predicted by mathematical modeling. Time changes of pore size, porosity, amount of deposition, and pressure gradient, etc. were estimated.

Model development

The cylindrical preform that is composed of layers of fiber bundles. Fibers are assumed nonporous. It is also assumed that pores distributed evenly in the whole preform as shown in Fig. 1.

Reactant gases, i.e., H₂ and MTS, flow from one side of the preform to another by forced convection in the isothermal reactor. For the gas concentration distribution, a pseudo-steady state was assumed. It is supposed that SiC infiltration reaction is a first order reaction of MTS by which 1 mole MTS creates 1 mole SiC and 3 mole HCl.

\[ \text{CH}_3\text{SiCl}_3 \rightarrow \text{SiC}+3\text{HCl} \]  (1)

There is a z-directional convection in the pores between fibers. Following the reaction equation,
the mole balance of each ingredient was made. Poreosity, mole fractions, fiber radius, and amount of infiltration were calculated. Equations and methods of calculations are explained in the reference 13. Calculations were terminated when pores are plugged or pressure becomes more than a limiting pressure.

Results and discussion

Mathematical modeling was carried out with the parameter values listed in Table 1 and the reaction rate constant of $10 \text{ cm/min}^8$. Dimensions of the preform were taken from a sample used in the experiment done in our laboratory. With the mathematical modeling, changes of concentration, pressure, porosity, and amount of deposition were estimated.

Changes of dimensions during deposition the process

Fig. 2 is the time changes of fiber radius at different positions in the preform. As time goes on, fiber radius increases gradually at almost all parts of the preform. As expected, changes of fiber radius around the gas entrance ($z=0$) are big. The slopes of curves are rather decreasing after 80 min due to plugging of gas entrance.

Changes of the mole fraction of each component with time in the middle of the preform were also obtained. As the infiltration process goes on, the surface area around fibers becomes large. Hence, the deposition rate increases with the increasing surface area. As a result, the mole fraction of MTS decreases with time.

The mole fraction of HCl increases steadily first, but, steeply later. It is due to plugging of pores at the gas entrance. Since the entrance pressure rises, the pressure in the middle of the preform rises and the rate of deposition becomes faster. So the mole fraction of HCl increases rapidly after 110 min of reaction time.

Overturning of the preform

As shown in Fig. 3, the deposition reaction can not proceed after 130 min, because pores in preform are plugged. At this time, removing plugged...
surfaces, grinding surface, and overturning the preform are tried and the deposition process can be started again.

**Changes of pressure during the overturning process**

The changes of pressure at the gas entrance with time were obtained. Due to deposition, fiber radii increase and pore sizes decrease. Pressure increases rapidly at around 100 min, i.e. the overturning time. When the preform was overturned once, the reaction time extended up to 210 min.

**Changes of porosity during the overturning process**

Fig. 3 shows the changes of porosity with time after overturning the preform. Because the preform was overturned at 100 min, the porosity at the gas outlet \((z=H)\) decreased rapidly. The amounts of deposition at the gas entrance and the exit are more than those in the middle of the sample \((z=0.2H\) and \(0.5H)\).

Fig. 4 shows changes of porosity in the process of 5-overturning. Porosities at both sides of the preform, i.e., \(z=0, z=H\), became almost 0.1. With 5-overturning, the infiltration was carried out more effectively. However, the porosity at \(z=0.15H\) is 0.47. The infiltration reaction is still not enough in the middle of the preform. The plugging time increased to 250 min after 5-overturning process.

The total amount of deposition increases steadily with time as shown in Fig. 5.

**Effectives of the volumetric flow rate during the overturning process**

Fig. 6 is the changes of porosity at the initial gas entrance \((z=0)\) with the flow rates of 100, 500, and 1000 cm³/min. The preform was overturned at 60, 100, 140, and 180 min. The faster the flow rate is, the faster the infiltrate reaction ends.

**Conclusions**

Computer simulation of the operation process of the chemical vapor infiltration of SiC in the fabric preform has been studied. When the reaction time is 130 min, pore entrances are plugged and the pressure at the gas entrance increases sharply. The time when the preform should be overturned in the middle of the process is decided by observing the time changes of fiber radius at the gas entrance.

Changes of porosity and amount of deposition in the overturning process were obtained. Plugging time increased by 50 min with 1-overturning processing. Total amount of deposition increased about 1.4 times.

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References