Chemical Vapor Deposition of Carbon and Silicon Carbide in a Static Bed Reactor for the TRISO Coating Technology

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1. Introduction

Chemical vapor deposition (CVD) of carbon and silicon carbide (SiC) has been applied to TRISO-coated fuel particles for high-temperature gas-cooled reactors (HTGR). The TRISO-coated fuel particle consists of a microspherical fuel kernel surrounded by four layers, porous pyrolytic carbon (PyC), dense inner PyC, SiC, dense outer PyC [1]. The porous PyC coating layer, called the buffer layer, attenuates fission recoils and provides void volume for gaseous fission products and carbon monoxide. The inner PyC layer acts as a containment to gaseous products. The SiC layer provides mechanical strength for the particle and acts as a diffusion barrier to metallic fission products, which diffuse easily through the inner PyC layer. The outer PyC layer protects the SiC coating layer by inducing a compressive stress and provides chemical compatibility with a graphite matrix in the fuel compact. In order to insure the integrity of each layer after fabrication and in use the microstructure of the PyC and the SiC layers should be controlled properly [2]. The microstructure of the coating layer depends largely on the CVD conditions in a fluidized-bed reactor.

In this study, we performed some preliminary experiments on the PyC and SiC coating in a static bed CVD reactor to obtain a general implication about the effect of the CVD conditions on the properties of the PyC and the SiC layers.

2. Experimental procedure

Isotropic graphite (IG-11, Toyo Tanso, Japan) was used as a substrate for the deposition of the PyC and the SiC layers. Deposition was performed in a horizontal alumina tube furnace with a constant system pressure of 700 torr. Methyltrichlorosilane (MTS, CH_3SiCl_3, 99 %, Aldrich Chemical Co., Ltd.) and acetylene (C_2H_2) gas were used as sources of SiC and PyC, respectively. In the case of the deposition of PyC, Ar gas was used as a carrier gas with a total flow rate of 7000 sccm. The flow rate of the C_2H_2 gas was varied from 200 to 400 sccm and the deposition temperature was 1250°~1500°C. Hydrogen (H_2) gas was used as a carrier/diluent gas for the deposition of SiC. The flow rate of the MTS was controlled by controlling the flow rate of the carrier gas. The total flow rate was 7000 sccm and the ratio of H_2 to MTS was 44. Microstructures of the coating layers were observed using a scanning electron microscopy (SEM).

3. Results and Discussion

3.1. Deposition of PyC

Fig. 1 shows the deposition rate of the PyC coating layer as a function of the deposition temperature and the flow rate of C_2H_2 gas. When the flow rate of the C_2H_2 gas was 200 sccm, the deposition rate shows little change with a variation of the deposition temperature. However, the deposition rate increases abruptly above
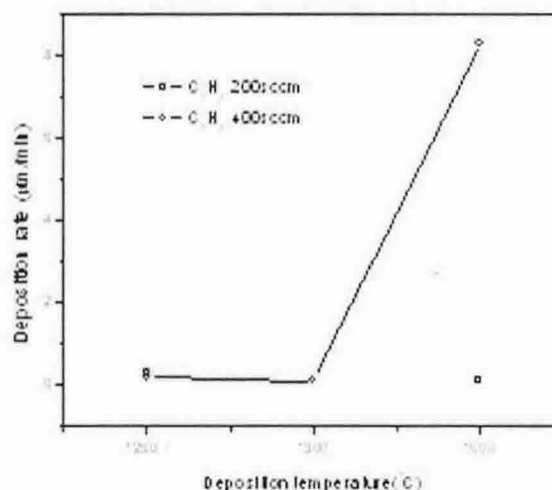

Fig. 1. Variation of the deposition rate of the PyC layer as a function of the deposition temperature and the flow rate of C_2H_2 gas.

1300°C with the C_2H_2 flow rate of 400 sccm. Generally, the porous buffer layer is deposited with a coating rate of 6~10 μm/min while the dense PyC layer is deposited with a lower rate of 4~6 μm/min. Therefore, the deposition at 1500°C with the C_2H_2 flow rate of 400 sccm appears to have an adequate deposition rate for the deposition of the buffer layer. Difference in the deposition rate greatly affects the microstructure of the PyC layer as shown in Fig. 2. As expected in Fig. 1, the sample with the highest deposition rate shows a porous PyC layer. The other samples show dense and laminar microstructures. The laminar structure tends to have a great degree of anisotropy and thus have a inferior performance. To obtain a more isotropic structure, it is required to increase the deposition rate because a low deposition rate leads to a higher degree of texture in the deposit as reported by Kaae et al. [3].
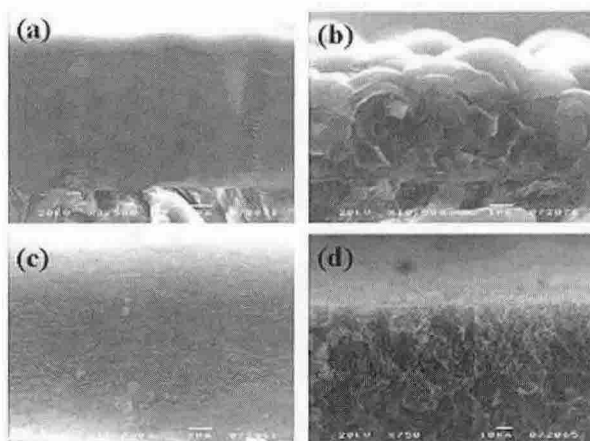
Fig. 2. SEM microstructures for the cross-sections of the samples deposited with the C_2H_2 flow rate of 200 ((a), (b)) and 400 sccm ((c), (d)) at 1250° ((a), (c)) and 1500°C ((b), (d)).

3.2. Deposition of SiC

Equilibrium mole fractions of solid species were calculated using the SOLGASMIX-PV program in order to obtain deposition conditions for stoichiometric SiC phase. Fig. 3 shows the calculation results with a variation of deposition temperature. It can be seen from this figure that a stoichiometric SiC is obtained at deposition temperatures above 1300°C if we control the dilution ratio to be higher than 20. No free Si phase is formed in this condition.
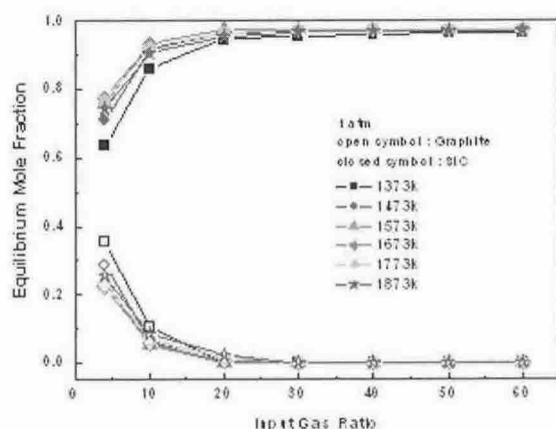

Fig. 3. Calculation results for the equilibrium mole fraction of C and SiC as a function of input gas ratio at various deposition temperatures.

Fig. 4 shows the surface and the cross-sectional microstructures of the SiC coating layer deposited at 1500°C for 2 h. The SiC coating shows a columnar structure consisting of very large grains. Additionally, the deposition condition shows a very high growth rate

around 5 ☐m/min. It has been reported that the smaller grained SiC with the laminar structure shows a better retention of metallic fission products than the larger grained SiC with the columnar structure [2]. The optimum deposition rate for the SiC layer has been reported as 0.2 ☐m/min. Therefore, the deposition rate should be decreased by controlling the CVD condition, e.g., by increasing the dilution ratio, and this will lead to a decrease of the grain size.
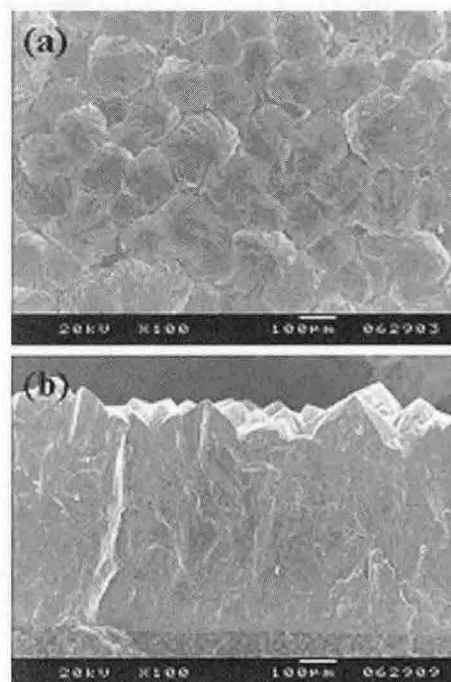

Fig. 4. SEM microstructures for the surface (a) and the cross-section (b) of the SiC layer deposited at 1500°C for 2 h.

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