FABRICATION AND CHARACTERIZATION OF SIC MATRIX COATING LAYER WITH ZRC PARTICLE

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ABSTRACT*: SiC coating on TRistructural-ISOtropic (TRISO) coated fuel particles serves both as a structural integrity and as a protective layer from radioactive fission gas. However SiC has the limitation for high temperature application because of the active oxidation above 1650 ^{o}C. ZrC is being studied as a substitute coating. ZrC is one of the most promising ceramics for ultrahigh temperature application due to the formation of a zirconia layer which is stable at high temperature. In this study, SiC matrix coating layer was deposited by chemical vapor deposition (CVD) method. ZrC particle was fabricated in SiC matrix coating layer by the simultaneous deposition with SiC matrix. Two phases of SiC and ZrC were clearly observed and separated by SEM and TEM analysis. There was no composite phase of Zr and Si in grain boundary. Oxidation properties of SiC matrix coating layer were investigated. Surface oxide layer was analyzed by XRD and SEM.*

KEYWORDS*: SiC coating, ZrC coating, CVD, Oxidation*

I. INTRODUCTION

SiC coating has been suggested as structural materials for nuclear application due to its excellent high irradiation resistance and high temperature mechanical properties[1,2]. TRISO coated fuel particles embedded in a carbonaceous matrix is considered as the fuel of high temperature gas-cooled reactors. TRISO coated particles consist of a kernel, a buffer porous pyrolytic carbon (PyC) layer and an IPyC/SiC/OPyC trilayer. SiC coating on TRistructural-ISOtropic (TRISO) coated fuel particles plays an important role as a protective layer from radioactive fission gas and a mechanical structural layer. SiC has the disadvantage of active oxidation at high temperature above 1650 ^{o}C and air atmosphere[3]. ZrC is one of the most promising ceramics for ultrahigh temperature application due to the formation of a protective zirconia layer at high temperature[4,5]. In this study, SiC and ZrC composite coating and SiC matrix coating with ZrC particle was deposited on the TRISO coated particle via fluidized bed chemical vapor deposition (FBCVD) method from adjusting ratio of Zr and Si source materials. The microstructure and phase of SiC matrix coating with ZrC particles were analyzed by SEM and TEM work. Oxidation properties of SiC and ZrC composite coating layer and SiC matrix coating with ZrC particles were investigated.

II. METHODS AND RESULTS

TRISO coating layer was deposited on a spherical particle by a FBCVD method. The ZrO_2 spherical particles were used as a simulant kernel. SiC coating layer was deposited using the MTS (Methyltrichlorosilane: CH_3SiCl_3), ZrC coating layer was deposited using $ZrCl_4$ and CH_4. Since the $ZrCl_4$ source material is in a solid state at room temperature, a sublimation system was installed to vaporize the source material. The flow rate of the $ZrCl_4$ gas was controlled by feeding amounts of $ZrCl_4$ source powder. The screw feeder system of a source material was introduced to supply the source material uniformly. The detail deposition process of SiC and ZrC coating on TRISO was explained in other paper[6,7]. SiC and ZrC composite coating layer of different microstructure were prepared by adjusting the gas flow rate of MTS, $ZrCl_4$ and CH_4. TEM specimens for microstructural analysis were taken at the center of the coating layer using a focused ion beam (FIB) technique.

Fig.1 shows the SiC/ZrC composite coating and SiC coating with ZrC particles. The deposition condition of each coating layer was shown in Table 1. The deposition temperature of all coating was 1500 ^{o}C and $ZrCl_4$ source gas sublimation temperature was 400 ^oC. Each coating layer has the thickness of 20 ~ 30 μm and well deposited on inner PyC layer. From the XRD results of each coating, it is confirmed that each coating layer consists of SiC and ZrC phase. It is confirmed that white phases are ZrC and gray phases are SiC from the EDS analysis.

Table 1. The deposition condition of each coating layer

Coating	Q	$ZrCl_4$	CH_4	MTS	Ar(D)	H_2(D)	A_r(C)	H_2(C)
SiC and ZrC composite	3925	40	10	30	1581	1543	341	380
SiC matrix coating with ZrC particles	4000	100	25	30	1581	1543	341	380

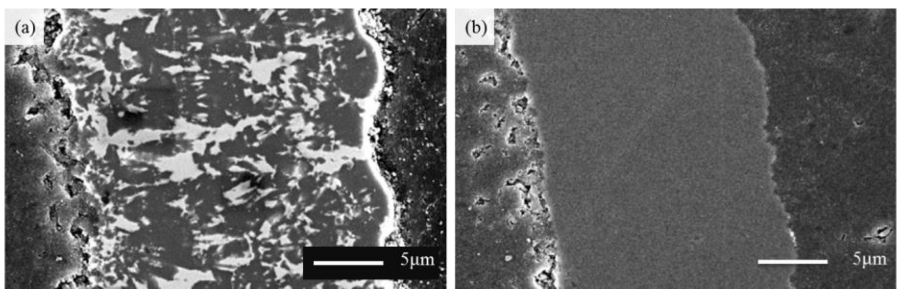

Fig. 1. Polished surface of (a) SiC and ZrC composite coating layer. (b) SiC matrix coating with ZrC particles.

Fig. 2 shows TEM images of SiC and ZrC composite coating layer and SiC matrix coating with ZrC particles. Two phases of SiC and ZrC were clearly observed and separated. Fig. 2 (b) shows high resolution TEM images of boundary between SiC and ZrC phase. Clean grain boundary between SiC and ZrC phase was observed. There was no composite phase of Zr and Si in grain boundary. In Fig. 2 (c), many small particles with size of 10 ~ 30 nm are present in SiC matrix. As a result of EDS analysis of small particle, Zr elements were detected differently from SiC matrix and so small particles were found to be ZrC phase. From XRD and TEM analysis, it is observed that SiC matrix coating layer with ZrC particles were successfully deposited on TRISO particles.

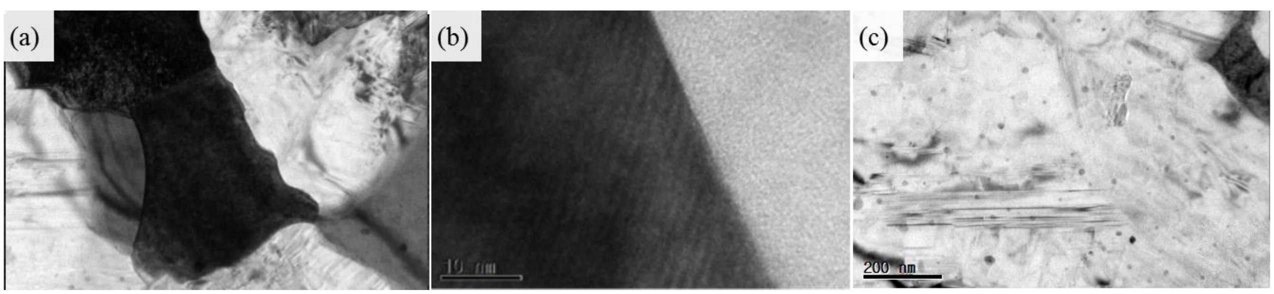

Fig. 2. TEM images of (a) SiC and ZrC composite coating layer, (b) SiC matrix coating with ZrC particles.

Oxidation experiments of each coating layer were conducted at 600 ~ 1400 ^oC. SiC and ZrC composite coating layer couldn't keep its structure after oxidation above 800 ^oC due to the formation of porous ZrO_2. In the specimens oxidized at 600 ^oC, the structure of the coating layer was maintained and oxide film which consists of ZrO_2, SiO_2, and $ZrSiO_4$ was formed. SiC matrix coating with ZrC particle endured after oxidation until 1400 ^oC. In the specimens oxidized at 1400 ^oC, thin oxide film was formed. Most of oxide film was SiO_2, but very small amounts of ZrO_2 and $ZrSiO_4$ were observed.

III. SUMMARY

SiC and ZrC composite coating and SiC matrix coating with ZrC particles on spherical particle were successfully deposited via FBCVD method by adjusting source gas flow rate. In the coating layers, SiC phase and ZrC phase were observed by XRD and SEM analysis. In the condition of 100 sccm of $ZrCl_4$, 25 sccm of CH_4, and 30 sccm of MTS, only two phases of SiC and ZrC were observed and two phases are presented with clean grain boundary. In the condition of 40 sccm of $ZrCl_4$, 10 sccm of CH_4, and 30 sccm of MTS, SiC coating is mainly deposited and many small particles were observed inside the coating. As a results of XRD and TEM analysis, it is observed that small particles with size of 10 ~ 30 nm are ZrC phase. An oxide film composed of ZrO_2, SiO_2, and $ZrSiO_4$ were formed outside the each coating layer after oxidation tests.

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