

Fabrication and Characterization of TRISO Coated Particles with ZrC Barrier Layers

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1. Introduction

Zirconium carbide (ZrC) is a candidate to replace the silicon carbide (SiC) of the TRISO coated particles for a very high temperature gas-cooled reactor (VHTR) as it has good phase stability and resistance to chemical attack by fission products [1]. In this study, ZrC-TRISO coated-particles were fabricated by a fluidized-bed chemical vapor deposition (FBCVD) method with a chloride process. Mechanical and microstructural properties of the ZrC coating layer were estimated.

2. Experimental

ZrC-TRISO coated particles were fabricated by the fluidized-bed chemical vapor deposition (FBCVD) method as shown in Fig. 1. Yttria-stabilized zirconia (ZrO₂) kernels with 534 μm of average diameter are chemically deposited with four coating layers. A ZrC layer was deposited by the chloride process. Reaction gases were supplied with $\alpha = \text{H}_2/(\text{ZrCl}_4 + \text{CH}_4) = 25$. A sublimation temperature of ZrCl₄ powders was 310°C and total pressure in a sublimation chamber was 760 torr. Deposition conditions of coating layers of ZrC-TRISO coated particles are summarized in Table I.

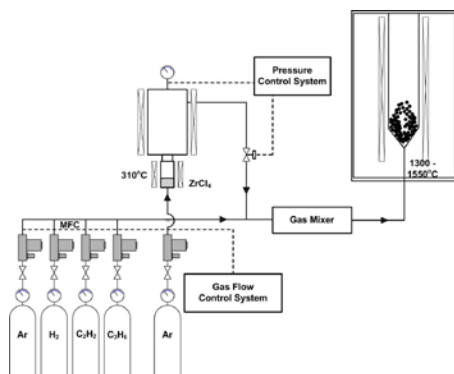


Fig. 1. A schematic of the coating system for ZrC-TRISO coated particles.

Table I: Deposition Conditions of Coating Layers of ZrC-TRISO Coated Particles.

	Deposition Temp. [°C]	Deposition Time	Reaction Gases
Buffer	1350	2m 40s	C ₂ H ₂
Inner PyC	1300	6m 50s	C ₂ H ₂ , C ₃ H ₆
ZrC	1400-1550	240m	ZrCl ₄ , CH ₄ , H ₂
Outer PyC	1350	2m 40s	C ₂ H ₂ , C ₃ H ₆

3. Results and Discussion

3.1 Fabrication of ZrC-TRISO coated particles

Fig. 2 shows typical microstructure of the TRISO coated-particle with the ZrC barrier layer which was deposited at 1500°C. It is composed of four coating layers such as a porous pyrolytic carbon layer (buffer PyC), a dense inner pyrolytic carbon layer (IPyC), a ZrC layer, and a dense outer pyrolytic carbon layer (OPyC).

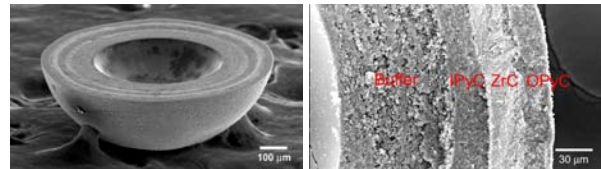


Fig. 2. Typical microstructure of the ZrC-TRISO coated particle fabricated by the FBCVD method.

A deposition rate of the ZrC layer was strongly dependent of a deposition temperature. The rate linearly increased in a temperature range of 1400 – 1550°C, as shown in Fig. 3.

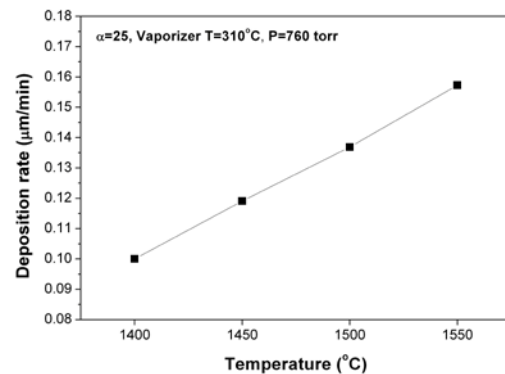


Fig. 3. Deposition rate of the ZrC layer as a function of the deposition temperature.

3.2 Characterization of the ZrC layer

Fig. 4 shows the crystallographic orientations of the ZrC grains of ZrC-TRISO coated particles deposited at 1450°C and 1500°C. ZrC grains have random texture, and average grain size increased generally in proportion to the deposition temperature.

Based on the auger electron spectroscopy (AES) depth profile analysis, a C/Zr ratio of the ZrC layer was approximately 1.0 in a temperature range of 1400 -

1500°C, as listed in Table II. On the other hand, carbon was quite rich in the ZrC layer deposited at 1550°C.

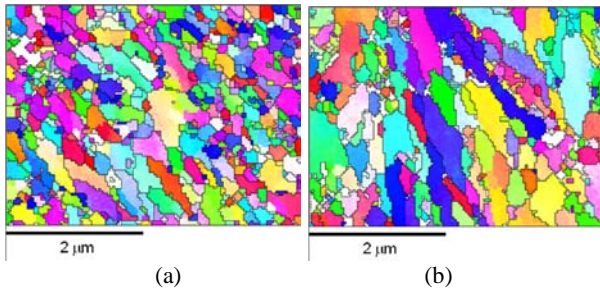


Fig. 4. EBSD orientation maps of ZrC layers deposited at (a) 1450°C and (b) 1500°C.

Table II: The C/Zr Ratio and Grain Sizes of ZrC Deposited at Different Deposition Temperatures.

	Deposition temperature, °C			
	1400	1450	1500	1550
C/Zr ratio	1.04	0.98	1.01	1.24
Grain size, μm	0.15	0.21	0.29	0.28

Fig. 5 shows the nanoindentation hardness and elastic modulus of ZrC layers as a function of the deposition temperature. Hardness and elastic modulus continuously decreased with increasing the deposition temperature.

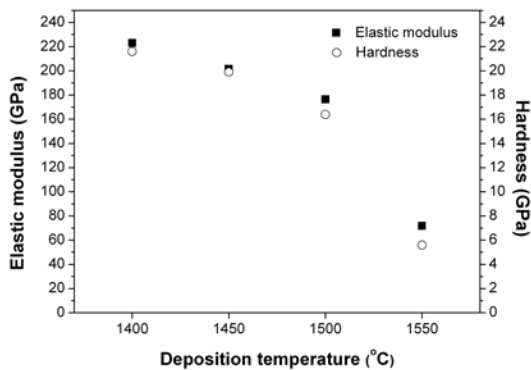


Fig. 5 Variation of nanoindentation hardness and elastic modulus of ZrC with deposition temperatures.

Reduction of hardness of ZrC relies on the difference in grain size in a temperature range of 1400 - 1500°C. The relationship between hardness and grain size can be described by Hall-Petch relation,

$$H = H_0 + K_H d^{-1/2},$$

where H_0 and K_H are constants. H and d are hardness and grain size, respectively.

As shown in Fig. 6, hardness of ZrC was linearly increased according to Hall-Petch relation as a function of $d^{-1/2}$. However, hardness of ZrC deposited at 1550°C was approximately 1/3 of that deposited at 1500°C, though both ZrC had almost same grain size.

AES line profiling results revealed that the carbon-rich phase was formed in the ZrC layer, as shown in Fig.

7, which resulted in the significant reduction of hardness

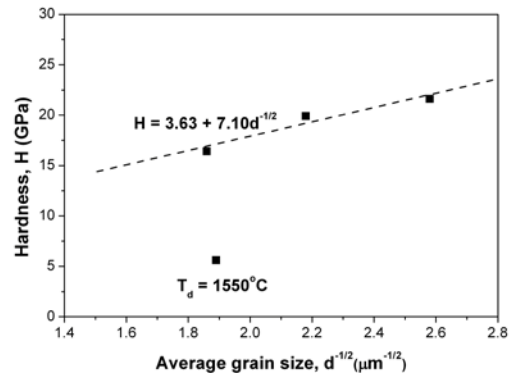


Fig. 6. Relationship between hardness and grain size for ZrC according to Hall-Petch relation, $H = H_0 + K_H d^{-1/2}$.

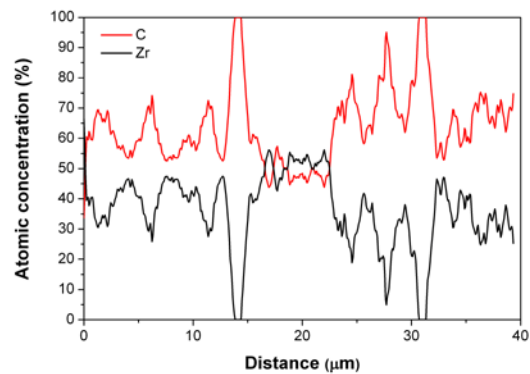
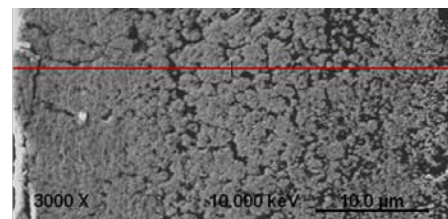


Fig. 7. SEM image and corresponding AES line profiling results of the ZrC layer coated at 1550°C.

In summary, ZrC-TRISO coated particles were fabricated by the FBCVD method with the chloride process. Hardness and elastic modulus of ZrC continuously decreased with increasing the deposition temperature. It was revealed that the reduction was primarily caused by the growth of grain size and formation of the carbon-rich phase.

Acknowledgement

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- [1] S. Ueta, J. Aihara, A. Yasuda, H. Ishibashi, T. Takayama, and K. Sawa, Fabrication of uniform ZrC coating layer for the coated fuel particle of the very high temperature reactor, Journal of Nuclear Materials, Vol. 376, p.146, 2008.