

## Deposition of Stoichiometric ZrC on TRISO Coated Particle by Optimizing Input Gas Ratio and Using Impeller ZrCl<sub>4</sub> Vaporizer

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### 1. Introduction

Advanced TRISO coated particle with ZrC as substitute for SiC has many higher performance capability in terms of corrosion resistance by fission products, fission product retention at higher temperature, and phase stability. However, it is difficult to obtain stoichiometric ZrC by CVD methods [1, 2]. Excess carbon in ZrC deteriorates the performance of the ZrC coating layer. In this study, TRISO coated particles with stoichiometric ZrC were fabricated by optimizing the input gas ratio and ZrCl<sub>4</sub> sublimation method.

### 2. Experimental

ZrC-TRISO coated particles were fabricated by a fluidized-bed chemical vapor deposition (FBCVD) method. A ZrC coating layer was deposited by the chloride process using ZrCl<sub>4</sub> powder at 1500°C. Static and impeller vaporizers were used for sublimation of ZrCl<sub>4</sub> powder. Source gas ratio in input gas ( $\beta = \text{ZrCl}_4 / (\text{ZrCl}_4 + \text{CH}_4)$ ) was controlled from 0.8 to 0.9 and dilution gas ratio ( $r_D = \text{Ar} / (\text{Ar} + \text{H}_2)$ ) was varied from 0.2 to 0.67. Sublimation temperature of ZrCl<sub>4</sub> powder was 310°C and pressure of sublimation chamber was 760 torr.

### 3. Results and Discussion

#### 3.1 Effect of source and dilution gas on ZrC deposition

H<sub>2</sub> as a dilution gas has an effect to reduce the pyrolysis process of CH<sub>4</sub> which is a source gas for the formation of excess carbon in ZrC. As shown in Fig. 1, the amount of excess carbon was gradually reduced as  $r_D$  changed from 0.67 to 0.2. Although a dilution gas was almost H<sub>2</sub> (Ar:H<sub>2</sub>=1:4), excess carbon still existed about 3 % in ZrC.

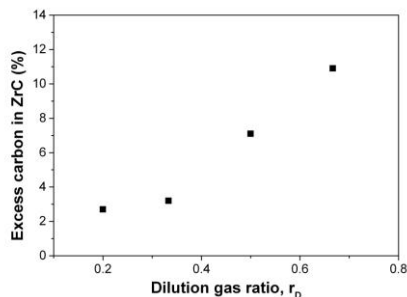


Fig. 1. Changes of excess carbon in ZrC depending on dilution gas ratio.

As CH<sub>4</sub> concentration in input source gases decreased, excess carbon disappeared and ZrC became near-stoichiometric. The amount of excess carbon decreased from 7.1 % for  $\beta=0.8$  to 0.2 % for  $\beta=0.88$ . Excess carbon was more reduced when  $\beta$  was 0.9 as shown in Fig. 2(a). However, an amount of excess carbon was still observed in outer regions of the ZrC layer while little excess carbon existed in inner regions (Fig. 2(b)). It could be due to a decrease in the sublimation rate of the ZrCl<sub>4</sub> as a deposition time. At the initial stage of the ZrC coating process, a sublimation rate of ZrCl<sub>4</sub> was quite steady. However, as deposition progressed, ZrCl<sub>4</sub> powder was observed to be agglomerated at the high temperature and flow channels for a carrier gas was formed at the final stage. Therefore, source gas ratio ( $\beta$ ) significantly decreased.

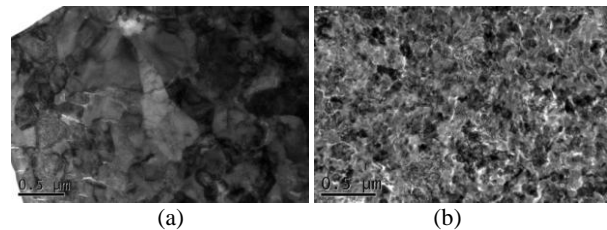


Fig. 2. TEM microstructure of ZrC deposited by a FBCVD method with  $\beta=0.9$ : (a) inner region and (b) outer region.

In the mean time, ZrC grains in the outer region became much smaller because excess carbon along grain boundaries retarded ZrC grain growth.

#### 3.2 Deposition of stoichiometric ZrC by using impeller vaporizer

The impeller type vaporizer was employed to keep up a constant sublimation rate of ZrCl<sub>4</sub> powder. Fig. 3 shows the deposition rates of ZrC coating layers as a

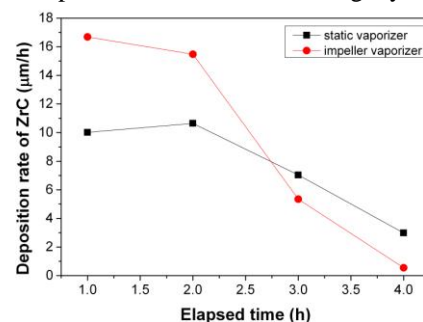


Fig. 3. Deposition rates of ZrC as a function of deposition time.

function of deposition time. Each ZrC layer was deposited for 1 hour at regular intervals of 5 minutes for PyC coating between ZrC depositions. Compared with the static vaporizer, the deposition rate of ZrC increased above 50%. After 2 hours of deposition, deposition rate was sharply decreased.

Fig. 4 shows microstructure of ZrC deposited by a FBCVD using impeller ZrCl<sub>4</sub> vaporizer. Stoichiometric ZrC without excess carbon was formed.

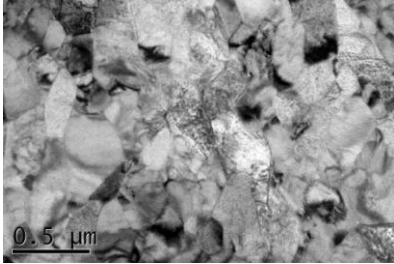


Fig. 4. TEM microstructure of ZrC deposited by a FBCVD using impeller ZrCl<sub>4</sub> vaporizer at 1500°C.

#### **4. Conclusions**

1. The C/Zr ratio decreased by increasing H<sub>2</sub> as a dilution gas. However, hydrogen has little effect on the formation of stoichiometric ZrC compared with other methods.

2. Excess carbon almost disappeared as CH<sub>4</sub> concentration was reduced. However, an amount of excess carbon in the outer region of a ZrC layer was formed owing to a gradual decrease in the sublimation rate of ZrCl<sub>4</sub> powder.

3. Stoichiometric ZrC without excess carbon was deposited by using impeller ZrCl<sub>4</sub> vaporizer.

#### **Acknowledgement**

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