

Strength evaluation method of ZrC coating layer on TRISO particles using a crush test

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1. Introduction

TRISO coated fuel particle used in HTGR(High Temperature Gas-cooled Reactor) is composed of four layers of buffer PyC layer, inner PyC layer, SiC layer, and outer PyC layer coated on UO₂ kernel. SiC layer maintains the structural integrity of TRISO coated fuel particle at high temperature and retains fission products since its excellent high temperature mechanical properties. However, β -SiC deposited by CVD causes a phase change to α -SiC above 1800 °C, which may cause damage to the TRISO coating layer. Since the SiC layer reacts with the fission product Pd to form Pd₂Si, which causes corrosion, resulting in a loss of ¹³⁷Cs trapping ability of TRISO coated fuel particle[1,2].

ZrC, which has more high stability than SiC at high temperature and low reactivity with fission product, has been proposed as an alternative coating layer. ZrC has been reported to have excellent thermal stability that does not cause phase transformation up to 2850 °C and has excellent corrosion resistance due to Pd so that the coated particles are not broken even at 2400 °C[3,4]. ZrC is thermodynamically more likely to form ZrC+C or Zr_{1-x}C than pure ZrC, and the resulting free carbon and carbon vacancy reduces the mechanical and radiation resistance of the ZrC layer[5,6]. Therefore, it is necessary to investigate the change of mechanical properties according to the stoichiometry of the ZrC coating layer. In this study, the strength measurement method of ZrC coating layer on TRISO coated particle were investigated by crush test and the mechanical properties of ZrC coating layer were measured according to the stoichiometry of ZrC.

2. Methods and Results

2.1 Strength measurement method using a crush test

Fig.1 illustrates the basic principles of the crush test and the simplified FEM model of the crush testing system. While each layer of the TRISO particle is ideally spherical, manufacturing processes result in ZrC shells which have aspheric geometries. In the investigation, a test specimen is represented as a uniform hemisphere shell. Compared to the existing conventional crush test of SiC coating on TRISO particle, the crush test of ZrC coating layer is more complex because buffer PyC and inner PyC layer are attached below ZrC coating layer[7,8]. In SiC TRISO particle, the PyC layer is easily removed through the heat treatment in the atmosphere environment. However,

in ZrC TRISO particle, the PyC layer cannot be removed by heat treatment due to the easy oxidation of ZrC in the atmosphere environment. Therefore, the crush test was carried out with a hemisphere shell consisting of three layers of buffer PyC, inner PyC, and ZrC coating, and the fracture stress applied was analyzed by FEA simulation. In the FEM model, the ram was high strength steel. The bottom of the hemisphere shell was constrained in six degrees of freedom (DOF). The outer radius of hemisphere shell was 350 μ m. The thickness of buffer PyC, inner PyC, and ZrC layer was 70 μ m, 30 μ m, and 30 μ m, respectively. Force was applied to the upper of the ram.

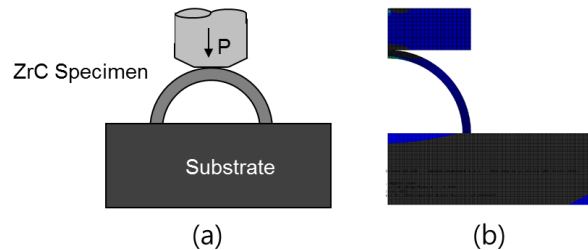


Fig. 1 (a) Schematic drawing of the crush test setup. (b) Simplified FEM model of crush testing system.

Fig. 2 shows in-plane principal stress distribution of the hemisphere shell in the crush test system when the applied load was 2 N. In the FEA results of crush test, the most induced stress caused by the load was applied to the ZrC layer so cracks occurs in the ZrC layer during crush testing. The maximum stress of ZrC layer was 1196 MPa. Based on the FEA results, the fracture strength of the ZrC coating layer can be calculated by measuring the load at which the ZrC coating layer is broken by the crush test.

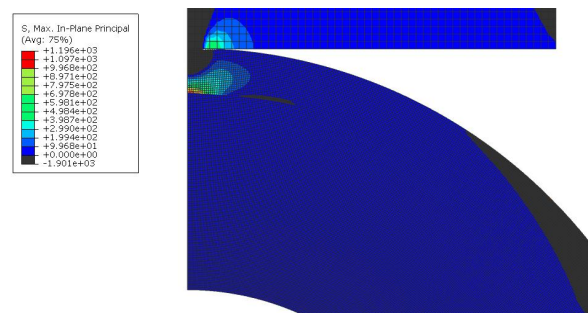


Fig. 2 The in-plane stress distribution of the hemisphere shell when the applied load was 2 N.

2.2 Evaluation of mechanical properties of ZrC coatings

ZrC TRISO coated particle was produced by a FBCVD (Fluidized-Bed Chemical Vapor Deposition) method. ZrC coating layer was deposited from $ZrCl_4$, CH_4 , H_2 and Ar at 1500 °C, $ZrCl_4$ powder were vaporized at 280 °C ~ 300 °C in the sublimation chamber and supplied into the reactor with an Ar carrier gas. In order to control the stoichiometry of the ZrC coating, the supply amount of the Zr source was adjusted by the sublimation temperature of $ZrCl_4$. The ratio of source gases and the ratio of dilution gases were 0.8 and 0.5 respectively.

Fig. 3 shows the cross-section images of the ZrC TRISO particles and Raman spectrum analysis results. Thickness of ZrC-1 sample which was deposited at 290 °C was 40 μm . Thickness of ZrC-2 sample which was deposited at 280 °C was 13 μm , Thickness of ZrC-3 sample which was deposited at 300 °C was 20 μm . Raman spectrum of ZrC-1 shows almost no peak. Because a NaCl structure like ZrC has o vibration modes because all atoms lie at a site of inversion symmetry, Raman peak of ZrC is not observed for the perfect stoichiometric ZrC[9]. Raman spectrum of ZrC-3 have several peak, which present acoustic branches at about $A_1 = 210\text{ cm}^{-1}$, $A_2 = 280\text{ cm}^{-1}$ and optical branches at about 540 cm^{-1} and 600 cm^{-1} contributed to by carbon vacancy. Raman spectrum of ZrC-2 have a weak carbon vacancy related peak and another two peaks induced by free carbon such as a disorder-induced (D) peak (1350 cm^{-1}) and Graphite peak (1582 cm^{-1}).

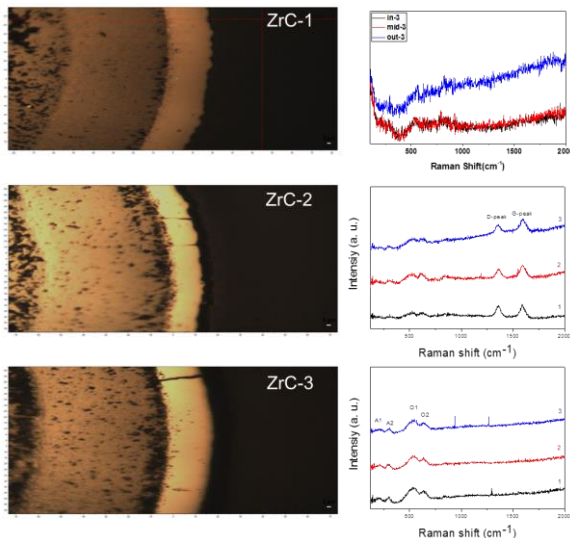


Fig. 3. Optical microscope image and Raman spectra of ZrC coating layers

The hardness of the ZrC coating layers was measured by nano indentation. Hardness of ZrC-1, ZrC-2, and ZrC-3 were 18.5, 29.0 and 35 GPa. In order to measure the strength of ZrC coating layer, ZrC hemisphere shells were prepared. ZrC TRISO particles were mounted using thermoplastic epoxy. The particles were ground

and polished to near midplane with 6 μm diamond suspensions. Kernels that had not already dropped out due to polishing to midplane were removed using an ultrasonic treatment. The particles were then removed from the epoxy by reheating the mount and washing off residual epoxy with acetone and yielded ZrC hemispherical shell with inner and buffer PyC. An instron with 1000 N load cell was used to load the specimens.

Fig. 4 shows the load-displacement curves of the ZrC-1 sample. In load-displacement curves, a small load drop was observed during the crush test, indicating that the ZrC coating layer where the maximum stress was concentrated was broken. The strength value of the ZrC coating layer was calculated by substituting the maximum stress value obtained through FEA simulations under the same conditions into the measured load. As a result, the strength value of ZrC-1 was measured at 255 MPa. It was confirmed that the strength of the ZrC coating layer can be measured by crush test using ZrC hemisphere shell with inner and buffer PyC layers. However, in the ZrC-2 and ZrC-3 sample, it was difficult to find a clear load drop due to the low load resolution of the equipment. The next step of the study is to measure and analyze the strength of the ZrC coating layer by observing a clear load drop using a equipment with a low load cell of 10 N or less.

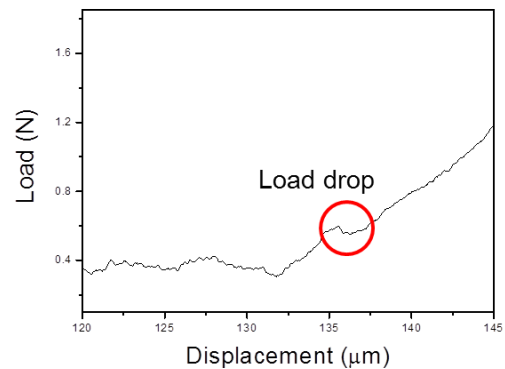


Fig. 4. Load-displacement curve of ZrC coating layer during crush test.

3. Summary

The crush test was performed to measure the strength of the ZrC coating layer and the stress analysis of crush test system was calculated by FEA simulation. As a result of FEA simulation using ZrC hemisphere shell with inner and buffer PyC layers, the maximum stress was applied to the ZrC layer. ZrC TRISO were fabricated by FBCD method. ZrC hemisphere shell were prepared by several treatments. In the load-displacement curves of ZrC hemisphere shell, a small load drop was observed when the ZrC coating layer was broken during crush test. From the fracture load, the strength value of

ZrC coating layer was evaluated by using FEA simulation results.

REFERENCES

- [1] K. Minato and K. Fukuda, "Chemical Vapor Deposition of Silicon Carbide for Coated Fuel Particles," *J. Nucl. Mater.*, Vol. 149 p. 233-46 1987.
- [2] S. Kouadri-Mostefa, P. Serp, M. Hemati, and B. Caussat, "Silicon Chemical Vapor Deposition (CVD) on Microporous Powders in a Fluidized Bed," *Powder Technol.*, Vol. 120 p. 82-87 2001.
- [3] K. Minato, T. Ogawa, K. Sawa, A. Ishikawa, T. Tomita, S. Iida, and H. Sekino, "Irradiation Experiment on ZrC-Coated Fuel Particles for High-Temperature Gas-Cooled Reactors," *Nucl. Technol.*, Vol. 130 p. 272-81 2000.
- [4] K. Minato, T. Ogawa, K. Fukuda, H. Nabielek, H. Sekino, Y. Nozawa, and I. Takahashi, "Fission Product Release from ZrC Coated Fuel Particles during Postirradiation Heating at 1600 oC," *J. Nucl. Mater.*, Vol. 224 p. 85-92 1995.
- [5] P. Wagner, L. A. Wahman, R. W. White, C. M. Hollaraugh, and R. D. Reiswig, "Factors Influencing the Chemical Vapor Deposition of ZrC," *J. Nucl. Mater.*, Vol. 62 p. 221-28 1976.
- [6] T. Ogawa, K. Ikawa, and K. Iwamoto, "Effect of Gas Composition on the Deposition of ZrC-C Mixtures: The Bromide Process," *J. Mater. Sci.*, Vol. 14 p. 125-32 1976.
- [7] S. -G. Hong, T. -S. Byun, R. A. Lowden, L. L. Snead, Y. Katoh, *J. Am. Ceram. Soc.* Vol. 90 p. 184-191, 2007.
- [8] B. C. Davis, L. Ward, D. P. Butt, B. Fillery, I. Reimanis, *J. Nucl. Mater.* Vol. 477, p. 263-272 2016.
- [9] H. Wipe, M. V. Klein W. S. Williams, *Phys. Status Solidi* Vol. B108 p. 489-500 (1981).