Microstructure and fracture analysis of fully ceramic microencapsulated fuel

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1. Introduction

After Fukushima accident(2011), the interest for the safety of nuclear plant has been increased. The concept of enhancing stability nuclear fuel itself is main issues to increase safety of nuclear power plant. Fully ceramic microencapsulated(FCM) nuclear fuel is one of the recently suggested concept as the accident tolerance nuclear fuel system[1-2]. Nuclear fuel enhancing the accident tolerance is satisfied two parts. First, the performance has to be retained compared to the existing UO_2 nuclear fuel and zircaloy cladding system under the normal operation condition. Second, under the severe accident condition, the high temperature structural integrity has to be kept and the generation rate of hydrogen has to be reduced largely. FCM nuclear fuel is composed of tristructural isotropic(TRISO) fuel particle and SiC ceramic matrix. SiC ceramic matrix play an essential part in protecting fission product. In the FCM fuel concept, fission product is doubly protected by TRISO coating layer and SiC ceramic matrix compared to the current commercial UO_2 fuel system. SiC ceramic has excellent properties for fuel application. SiC ceramic has low neutron absorption cross-section, excellent irradiation resistivity and high thermal conductivity[3-4]. Additionally, the relative thermal conductivity of the SiC ceramic as compared to UO_2 is quite good, reducing operational release of fission products form the fuel. TRISO coating layer which is deposited on UO_2 kernel is consists of PyC/SiC/PyC trialyer and buffer PyC layer.

Generally SiC ceramics are produced by liquid phase sintering using sintering additives due to strong covalent bonding property and low self-diffusivity coefficient. Hot press sintering is very effective to fabricate SiC ceramic including different second phase[5-6]. In the FCM fuel concept, SiC ceramic matrix has to be fabricated with oxide sintering additives. Oxide sintering additives are able to react with outer PyC coating layer during sintering.

In this study, SiC matrix composite including TRISO particles were sintered by hot pressing with Al_2O_3-Y_2O_3 additive system. In order to investigate the reaction between sintering additives with PyC, the contents of sintering additive increased from 3 wt.% to 20 wt.%. The thermal shock resistance of SiC matrix composite was studied and the fracture feature after thermal shock according to reaction between sintering additives and PyC was observed.

2. Methods and Results

β-SiC powder TRISO particle deposited by chemical vapor deposition method were used to fabricate SiC matrix composite. Al_2O_3 and Y_2O_3 sintering additives was used to liquid phase sintering of pellet. Nano sized SiC powder was added in order to enhance the sintering of SiC matrix. The starting powders were ball-milled with ethanol. TRISO particle was overcoated with the mixed powder in order to place TRISO particle uniformly embedded in the SiC matrix. The thickness of the coating layer was several micro-meters. A spherical mixed body with about diameters of 1.5 ~ 2 mm consists of the kernel of TRISO particle and the outer mixed powder overcoating layer. The mixed powder and overcoated TRISO particles were inserted to a graphite sleeve and hot pressing was carried out under a pressure of 20 MPa at 1800 °C for 1 hr in an Ar atmosphere. The hot pressing conditions were shown with Table 1.

Table 1. Hot pressing condition of SiC matrix composite with TRISO particle

	TRISO particle	Contents of sintering additives
FM01	30 vol.%	3 wt.%
FM02	30 vol.%	10 wt.%
FM03	40 vol.%	10 wt.%
FM04	30 vol.%	20 wt.%
FM05	40 vol.%	20 wt.%

The density of the sintered body was measured by Archimedes method. The as-sintered specimens were observed by scanning electron micropy after mirror-polishing and subsequent etching with CF_4 plasma containing 10% oxygen. The distribution map of TRISO coated particles within SiC matrix composite was observed by X-ray μCT with a sample size of 10 mm diameter and 4 ~ 6 mm height.

The relative density of SiC matrix composite was calculated by considering the weight proportion of TRISO particles. Relative densities of 40 vol.% specimens are above 92%, the relative densities of specimens with 30 vol.% TRISO particle are above 95%.

The X-ray radiographic image of the specimen sintered at 30% volume fraction of TRISO particle with 3 wt.% sintering additives is shown in Fig. 1. It is shown that the spherical shade of the zirconia kernel within TRISO particle embedded in the SiC matrix. The other

coating layer: the OPyC, the SiC, the IPyC layer of TRISO particle are not resolvable in the radiographic image as they interact the same as the SiC matrix with the incident photons[7]. In the distribution map of the zirconia kernel, which is representative TRISO particle embedded FCM pallet, TRISO particles are shown uniformly distributed well. It is thought that SiC powder overcoating on TRISO particle play a role to keep a distance between TRISO particles. However, an empty space without TRISO particle in SiC matrix composite is still observed, the improvement of overcoated TRISO particle loading method in graphite sleeve which is used for hot press sintering is required.
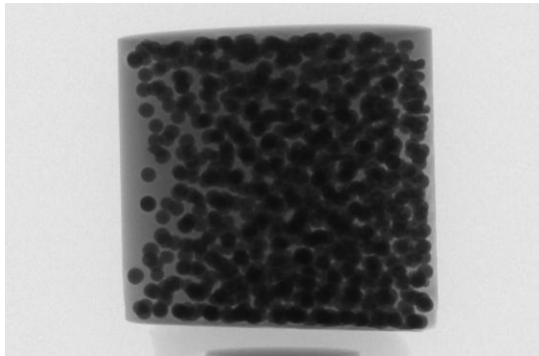

Fig. 1. X-ray radiography of SiC matrix composite with 30 vol.% TRISO particles.

The microstructure of specimens with different sintering additives contents are shown in Fig. 2. The liquid phases are well observed at grain boundary, the densification is promoted by the liquid phase through liquid phase sintering. In the specimen with 10, 20 wt.% sintering additives, large amounts of liquid phases are observed due to too large doses of sintering additives. In the specimen with above 20 wt.% sintering additives, the reaction between sintering additives and outside of PyC coating layer is observed. The thickness of reactants shows about ~10 micrometers, the reactants layer are located along entire outside of TRISO particle. EDS analysis was used to investigate the reaction mechanism.
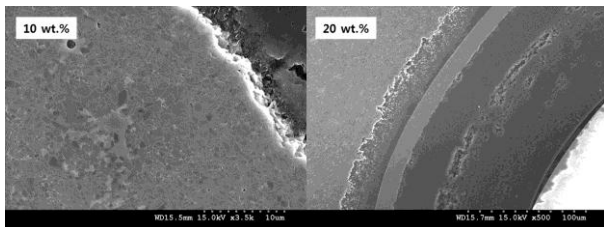

Fig. 2. SEM micrograph of SiC matrix and TRISO particle with different amounts of sintering additives.

The fractured specimen fabricated with 20 wt.% sintering additives and 40 vol.% TRISO particles is shown in Fig. 3. It is known that crack propagates along the outside of TRISO particles and TRISO coating layer did not break[7]. For the investigation of thermal shock resistance in SiC matrix composite water quench method was used. The samples were heated in a vertical tube furnace in argon gas to 1200 ^{o}C and held there for 10 min. Then the specimens were rapidly immersed into a ~20 ^{o}C water bath. SiC ceramic without TRISO particles breaks in two after thermal shock experiments by comparison with SiC matrix composite with TRISO particle does not break. It is thought that toughening mechanism arise from TRISO particle occurs. The cross-section microstructure analysis is conducted for detailed analysis. From the microstructure, crack propagation mechanism was investigated

Fig. 3. Fracture surface of SiC matrix composite with 40 vol.% TRISO particles

3. Summary

SiC matrix composite with TRISO particle was fabricated by hot pressing. 3 to 20 wt.% of sintering additives were added to investigate reaction between sintering additives and outer PyC layer of TRISO coating layer. The relative densities of all specimens show above 92%. The reaction between sintering additives and PyC is observed in most TRISO particles, the thickness of reactants shows about ten micrometers. The thermal shock resistance of SiC matrix composite was investigated. SiC matrix composite did not break after 1200 ^{o}C water quench experiments, it is known that toughening mechanism by TRISO particle occurs.

Acknowledgements

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