Fabrication of fully ceramic microencapsulated fuel by hot pressing

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1. Introduction

Fully ceramic microencapsulated(FCM) nuclear fuel is one of the recently suggested concept to enhance stability nuclear fuel itself[1-2]. The requirements to increase the accident tolerance of nuclear fuel are mainly two parts: First, the performance has to be maintained 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 decrease largely. FCM nuclear fuel consists of tristructural isotropic(TRISO) fuel particle and SiC matrix. TRISO fuel particle is composed of UO_2 kernel and PyC/SiC/PyC trilyer which protects kernel, SiC matrix is fabricated by sintering. 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 attracted for fuel application owing to low neutron absorption cross-section, excellent irradiation resistivity, and high thermal conductivity[3-4]. Additionally, the relative thermal conductivity of the SiC matrix as compared to UO_2 is quite good, yielding as-irradiated fuel centerline temperature compared to high temperature for the existing fuel leading to reduced stored energy in the core and reduced operational release of fission products from the fuel[2].

Generally SiC ceramics are fabricated via liquid phase sintering due to strong covalent bonding property and low self-diffusivity coefficient. Hot pressing is very effective method to conduct sintering of SiC powder including different second phase[5-6]. In this study, SiC-matrix composite including TRISO particles were sintered by hot pressing with Al_2O_3-Y_2O_3 additive system. Various sintering condition were investigated to obtain high relative density above 95%. The internal distribution of TRISO particles within SiC-matrix composite was observed by x-ray radiograph. From the analysis of the cross-section of SiC-matrix composite, the fracture of TRISO particles was investigated. In order to uniform distribution of TRISO particle embedded in the SiC matrix, SiC powder overcoating is considered.

2. Methods and Results

TRISO particle deposited by chemical vapor deposition method and β-SiC powder were used to fabricate SiC matrix composite. Liquid phase sintering with Al_2O_3 and Y_2O_3 sintering additives was used. Nano sized SiC powder was added in order to enhance the sintering of SiC matrix. The starting powders were ball-milled with ethanol and the coarsening powders were mixed with TRISO coated particle after ball milling. The mixed powder was inserted to a graphite sleeve and hot pressing was carried out under a pressure of 20 MPa at 1700 ~ 1800 °C 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

	Temperature	Time	TRISO Particle
FM01	1700 ℃	1 hr	34%
FM02	1800 ℃	1 hr	34%
FM03	1800 ℃	3 hr	28%
FM04	1800 ℃	5 hr	28%
FM05	1750 ℃	1 hr	28%

The density of the sintered body was measured by Archimedes method. The as-sintered specimens were observed by scanning electron micocopy 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 12 mm diameter and 4 ~ 6 mm height.

The density of SiC matrix composite with different sintering temperature and time are shown at Fig. 1. The relative density of the composite specimen was calculated by considering the weight proportion of TRISO particles. The specimens with 1 wt.% sintering additive show low density below 80% at a temperature of 1800 °C and the specimens containing above 3 wt.% sintering additive show high density above 90% at a temperature of 1750 °C. As increasing sintering time and temperature, the density of specimen increase. The specimen sintered at 1800 °C for 3 hr with 7 wt.% sintering additive is fully densified. The density of specimens sintered at 1750 °C with 28% volume fraction of TRISO particle is generally higher than that of specimens sintered at 1800 °C with 34% volume fraction of TRISO particle. It is thought that the densification would be more difficult as increasing contents of TRISO particle.
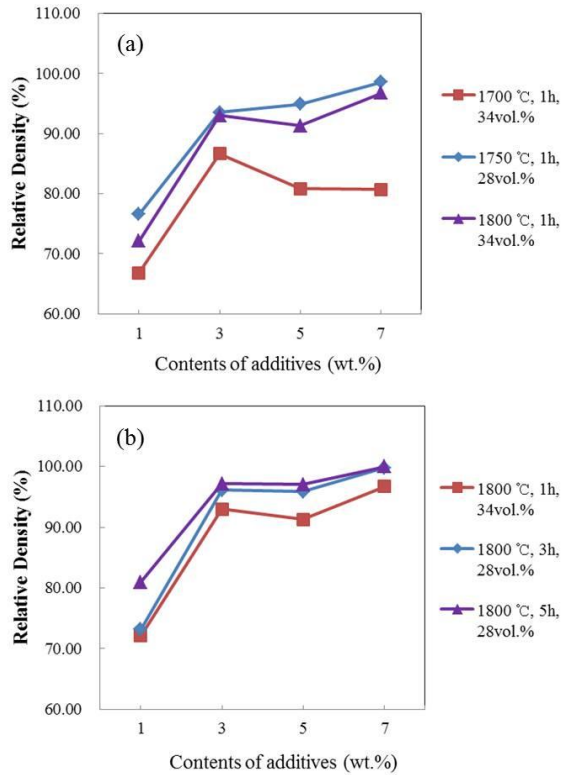
Fig. 1. Relative density of SiC matrix composite with different sintering additives contents: (a) with different temperature, (b) with different time

The microstructure of the specimen with different sintering additives contents are shown at Fig. 2. SiC matrix composites were fabricated at 1800 ^{o}C during 3 hr with 28% volume fraction of TRISO particle. In the specimen sintered with above 3 wt.% sintering additive, the liquid phase is well observed at grain boundary. It is thought that the densification is promoted by the liquid phase through liquid phase sintering. The narrow and long grains were partially observed in the microstructure at the fully densified specimen. It is thought that phase transformation was occurred from β-SiC to α-SiC and it is well-known at previous study[7].

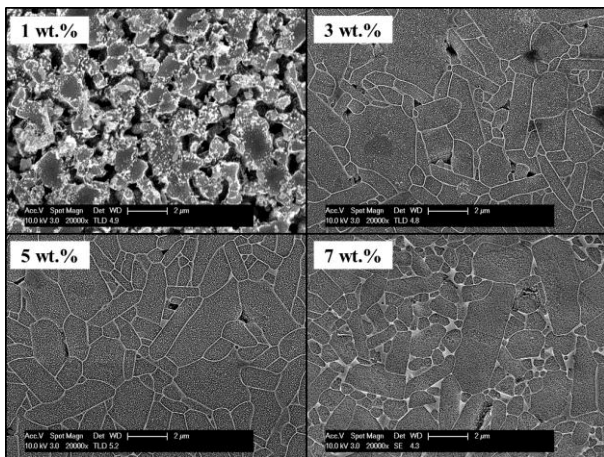Fig. 2. SEM micrograph of SiC matrix sintered with different sintering additives

The X-ray radiographic image of the specimen sintered at 1800 ^{o}C for 3 hr with 28% volume fraction of TRISO particle is shown at Fig. 3. 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. In the distribution map of the zirconia kernel, which is representative TRISO particle embedded FCM pallet, TRISO particles are crowded in the lower part of the pallet and are broadly distributed in the upper part of the pallet. It is because an experimental method of the mixed powder insertion into graphite sleeve. When the mixed powder including TRISO particle is inserted and flow down into graphite sleeve, TRISO particle is located at the lower part of the pallet because the density of TRISO particle is higher than that of agglomerate starting powders. The densification of the lower part of the pallet became different because of the narrow distribution of TRISO particle. It is important that the uniform distribution of TRISO particle for full densification of SiC matrix composite.

SiC powder overcoating on TRISO particle was considered in order to place TRISO particle embedded in the SiC matrix. The thickness of the coating layer is several micro-meters in the general overcoating method. In this study, several millimeters overcoating technique was used. A spherical mixed body with about diameters of 3 ~ 5 mm consists of the kernel of TRISO particle and the outer SiC powder overcoating layer. FCM pallets with uniformly distributed TRISO particle were fabricated using the spherical mixed body.
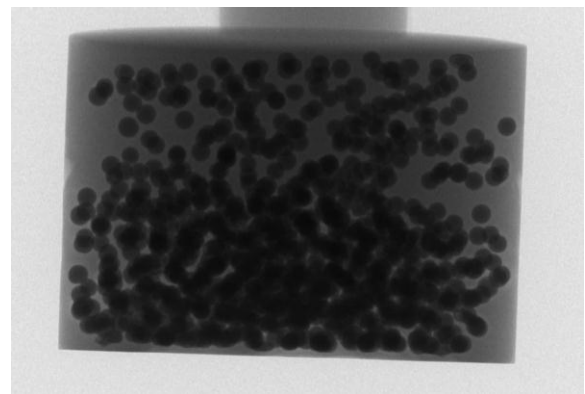

Fig. 3. X-ray radiography of SiC matrix composite

3. Summary

SiC matrix composite including TRISO was fabricated by hot pressing. FCM pallets with full density were obtained with Al_2O_3-Y_2O_3 additive system. From the microstructure image, the effect of the sintering additive contents and sintering mechanism was analyzed. The distribution of TRISO particle within the SiC matrix is observed by x-ray radiography. TRISO particle is located non-uniformly due to the mixed

powder insertion method into a graphite sleeve. SiC powder overcoating on TRISO particle is investigated in order to place TRISO particle uniformly within the SiC matrix.

Acknowledgements

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