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Effect of hydrothermal corrosion on the fracture strength of SiC layer in TRISO fuel particles

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Abstract

The hydrothermal corrosion behavior of SiC layer in tristructural-isotropic (TRISO) fuel particles and its effect on the fracture strength were investigated. The corrosion test was performed using the static autoclave at 400 °C/10.3 MPa. The SiC layer exhibited a thickness loss and the corrosion rate followed a linear law. During corrosion, carbon was formed on the SiC surface due to the loss of Si. The corrosion was found preferentially occurred at the grain boundary of SiC, leading to the grain detachment and pit formation. The rate determining step of the corrosion was SiO₂ formation rather than SiO₂ dissolution in the hydrothermal environment. The fracture strength of SiC shell after corrosion was evaluated using the crush test. It showed a slight decrease with an increase of corrosion time, due to the thickness reduction of SiC layer. The results of this study demonstrated that the SiC in TRISO particles has good corrosion resistance in the hydrothermal environment.

Keywords

silicon carbide, fuel particle, hydrothermal corrosion, fracture strength

1 Introduction

The tristructural-isotropic (TRISO) fuel particles developed for high temperature reactors (HTRs) are known for their superior high-temperature tolerance and extraordinary stability under neutron irradiation.^{1, 2} These advantages also render TRISO particles a high potential for use in light water reactors (LWRs). Particularly after the Fukushima Dai-ichi nuclear power plant accident, the current Zr alloy claddings do not have sufficient accident tolerance to guarantee fuel safety in a typical loss of coolant accident scenario, which has sparked renewed interest in exploring the fuel system to improve the safety of LWRs under accident conditions.³ Consequently, the research efforts to use TRISO particles in LWRs are significantly increasing over the past few years.⁴⁻⁶

Currently, a new fuel system for LWRs called fully-ceramic microencapsulated fuel (FCM) has been proposed.⁵ It consists of TRISO particles dispersed in SiC matrix to form the fuel compact, which can then be loaded in the nuclear cladding. Under the normal operation condition of LWRs, the TRISO particles are isolated from water coolant by several protective barriers. However, once the cladding breakage should happen, the TRISO particles might be exposed to the water coolant. The hydrothermal corrosion could cause degradation of the structural integrity and the mechanical properties of the outer layers, which would result in fuel damage and disastrous safety problems. Additionally, interest in using SiC as the nuclear fuel cladding material in LWRs has increased considerably.⁷ Therefore, the hydrothermal corrosion of SiC layers

of TRISO particles in water coolant at high temperature and pressure should be investigated before this material can be deployed in LWRs.

In recent years, there have been many reports on the hydrothermal corrosion of bulk SiC in relation to its application as fuel cladding in LWRs.⁸⁻¹³ However, the SiC layer on spherical substrate which is the case for TRISO particles exhibited different microstructure and mechanical properties from that of SiC bulk.¹⁴ Until now, the corrosion and mechanical behaviors of SiC layers in TRISO particles in the hydrothermal environment have not been elucidated. Therefore, in this study, the corrosion behavior of SiC layers of TRISO particles was investigated in the static autoclave. The fracture strength of SiC layers after corrosion was also studied, which is essential for the design and performance analysis of TRISO particles.

2 Experimental procedure

The TRISO particles were provided by the University of Manchester made by fluidized bed chemical vapor deposition using zirconia particles with a diameter about 500 μm as simulated kernels.¹⁵ They were prepared without the OPyC layers, the average thickness of the buffer, IPyC, and SiC layers were 200, 40, and 21 μm , respectively.

2.1 Hydrothermal corrosion

The corrosion test was carried out in the static autoclave. The TRISO particles were exposed to the high temperature and pressure water (400 °C/10.3 MPa) with a pH = 7.0-7.2 for up to twelve days. No boric acid and LiOH were added to the water chemistry condition and the water environment was deaerated to a low oxygen content.

Due to the variations in the shape and size of TRISO particles, large errors involve in measuring the weight change of TRISO particles. In order to evaluate the corrosion kinetics more accurately, the hemispherical SiC shell obtained by burning off the PyC layer was also added during the corrosion test and the thickness change of the separated SiC shell was used as a measure of the corrosion degree. The thickness measurement was performed every 3 days of corrosion using an optical microscope.

2.2 Crush testing

After corrosion, the TRISO particles were embedded in epoxy and grinded to the mid-plane of the particles using 240, 600, 1200, and 2000 grit SiC papers, then polished using increasingly fine diamond suspensions. The hemisphere particles were heated at 700 °C for 10 hours in air to burn off the PyC layers and obtained free-standing hemispherical SiC shells whose strength represent the strength of the whole particles.¹⁶

A modified crush testing was carried out for the hemispherical shells in a universal mechanical testing machine (Z020; Zwick/Roell, Ulm-Einsingen, Germany). For each condition, 30 hemispherical shells were selected and tested at room temperature with a crosshead speed of 0.005 mm/s. Figure 1 illustrates the process of the test. A SiC shell (out radius R , thickness δ) was diametrically loaded between a plunger and a testing bed by an external load F . An annealed brass foil was inserted between the SiC shell and plunger which ensured a more uniform stress distribution on the circular contact area (radius r_0). According to the result of finite element analysis, it was essential to have the contact area with a reasonable size for an accurate evaluation of the fracture strength.¹⁷ If the contact area was too small, measurement errors would be prohibitively

Figure 1 (A) SEM image of a SiC hemispherical shell and (B) schematic sketch of the crush test system

large. However, if the contact area was too large, the maximum principal stress failed to reach the inner apex of SiC shell, which would greatly affect the accuracy of the result. It was required that the angle between the vertical direction and radius direction should not be more than 10° (as shown in Figure 1). Therefore, by using the brass foil that has a proper hardness (71 HV_{0.05}) after a heat treatment (425 °C, 1 h), the contact area can reach a reasonable size (r_0 : 60 μm -85 μm).

The fracture strength of SiC shell can be calculated from the testing results according to the approach as reported in the previous study.¹⁸ A brief summary is given here. The maximum local fracture strength (σ_f^L) at the inner apex of SiC shell is a combination of the maximum membrane stress and bending stress, which can be calculated using the following equations^{18, 19}:

$$\sigma_f^L = \sigma_{\text{membrane}} - \sigma_{\text{bending}} \quad (1)$$

$$\sigma_{\text{membrane}} = -C_1 \frac{F\sqrt{1-v^2}}{\delta^2} \quad (2)$$

$$\sigma_{\text{bending}} = -C_2 \frac{F\sqrt{1+\nu}}{\delta^2} \quad (3)$$

$$C_1 = 0.2205 - 0.04\mu - 0.0115\mu^2 \quad (4)$$

$$C_2 = 1.2044 e^{(-1.2703\mu)} \quad (5)$$

$$\mu = r_0 \left[\frac{12(1-\nu^2)}{R^2 \delta^2} \right]^{\frac{1}{4}} \quad (6)$$

where ν is the Poisson ratio, δ is the thickness of shell, F is the applied load, C_1 and C_2 are the intermediate coefficients, μ is the intermediate variable, r_0 is the radius of the circular contact area, and R is the outer radius of the shell.

Weibull distribution function is the most widely used method to analyze the distribution of the local fracture strength. Using Weibull's two-parameter distribution, the cumulative probability of failure (P) is presented by^{18, 19}:

$$P = 1 - \exp \left[-S_E \left(\frac{\sigma_f}{\sigma_0} \right)^m \right] \quad (7)$$

where σ_f , m , σ_0 , and S_E are the local fracture strength, the Weibull modulus, the characteristic strength and the size effect factor, respectively. The size effect factor S_E can be calculated using following expression^{18, 19}:

$$S_E = \pi r_0^2 \quad (8)$$

Equation (7) can be rewritten in a linear form^{18, 19}:

$$\ln \left[\ln \left(\frac{1}{1-P} \right) \right] = m \ln \sigma_f^L + \ln \left(\frac{S_E}{\sigma_0^m} \right) \quad (9)$$

The Weibull modulus and characteristic strength can be obtained from the slope and intercept terms of the Equation (9) respectively. For each specimen, the value of P_i is a prescribed probability estimator rather than a true value. Several probability estimators and their merits have been reported in the previous study.²⁰ A conservative probability estimator is given here¹⁹:

$$P_i = \frac{i}{N+1} \quad (10)$$

where P_i is the probability of failure for the i th-ranked stress datum and N is the sample size.

Compared to the local fracture strength, the fracture strength for the full spherical shell (σ_f^F) is best described as the fracture strength representing the whole shell under a uniform internal stress. Combining the effective surface area and the Weibull modulus, the fracture strength for full spherical shell can be calculated using the following equation^{18, 19}:

$$\sigma_f^F = \sigma_f^L \left(\frac{S_E^L}{S_E^F} \right)^{\frac{1}{m}} = \sigma_f^L \left[\frac{\pi r_0^2}{4\pi(R-\delta)^2} \right]^{\frac{1}{m}} \quad (11)$$

where σ_f^L , S_E^L , σ_f^F , and S_E^F are the fracture strength, and the effective surface for the local loaded area, the fracture strength and the effective surface for the full spherical shell, respectively. Specifically, the coefficient $\pi r_0^2 / 4\pi(R-\delta)^2$ is a scaling factor accounting for the size difference between the localized loading and the uniform internal stress.

2.3 Characterization

The thickness of SiC layer after corrosion was measured at about twenty points using an optical microscope (BX51M, Olympus, Japan). The microstructure of SiC layer was studied by scanning electron microscopy (Inspect F50; FEI, Hillsboro, OR) and transmission electron microscope (TEM) (TALOS F200X, FEI, USA). TEM specimen was prepared using focused ion beam (FIB) (GAIA3, Tescan, Czech Republic). The phase and chemical information of the samples was characterized using X-ray diffraction (XRD) (Ultima IV, Rigaku, Japan), Raman spectroscopy (LabRAM

HR Evolution, Horiba, France) with 532 nm argon laser and X-ray photoelectron spectroscopy (XPS) (250XI, Thermo ESCALAB, America).

3 Results

3.1 Hydrothermal corrosion kinetics

Figure 2 shows the thickness change of SiC shell as a function of corrosion time at 400 °C/10.3 MPa. The measurement was performed on the same hemisphere shell at different time of corrosion. At least 10 hemispheres were used for the thickness measurement. The data suggested that the thickness reduction increased slightly with the increase of exposure time and the corrosion of SiC proceeds at a constant rate following the linear rate law. As shown from the optical images, the SiC shell was hardly corroded and almost kept its integrity, implying that the SiC layers of TRISO particles had good corrosion resistance in the hydrothermal environment.

Figure 2 The thickness of SiC shell as a function of corrosion time

3.2 Phase analysis of SiC layers after corrosion

The phases of the SiC surface after corrosion were analyzed using XRD and Raman spectroscopy, respectively. Figure 3A shows the XRD patterns for the TRISO particles. Three typical phases could be identified for all the samples, i.e., the ZrO_2 kernel, the PyC layer, and SiC layer, respectively. Meanwhile, Raman spectroscopy of the TRISO particles is shown in Figure 3B. The first-order peaks at 796 and 972 cm^{-1} and second-order peaks at 1540 and 1720 cm^{-1} correspond to 3C-SiC. In addition, the D (1350 cm^{-1}) and G (1600 cm^{-1}) bands in Figure 3C, which belong to the first-order peaks of carbon also appeared after corrosion. With an increase of corrosion time, the amount of carbon increased, as manifested by the intensity ratio between carbon and SiC peaks. This result is inconsistent with the previous studies, in which no carbon was formed on the surface of the corroded SiC bulk.^{8-12, 21, 22} The formation of carbon could suggested that the SiC layers of TRISO particles were corroded due to the loss of Si from SiC layer.

Figure 3 Phase analysis of TRISO particles before and after corrosion, (A) XRD patterns, (B) Raman spectra and (C) detailed spectra from 1200 cm^{-1} to 1800 cm^{-1}

However, taking into account the thickness of SiC layer and the penetration depth

of the Raman laser, the appearance and increase intensity of D and G bands may be originated from the inner PyC layers when the thickness of the SiC layer is reduced after corrosion. Therefore, more detailed examination using XPS was employed to analyze whether carbon was indeed formed on the surface of the corroded specimens.

Figure 4 XPS spectra of TRISO particles before and after corrosion, (A) Si 2p peak for the as-deposited specimen, (B) Si 2p peaks for all the specimens and (C) C 1s peaks for all the specimens

Figure 4 shows the XPS spectra from the surface of SiC layer before and after corrosion. For Si 2p peaks (Figure 4A), the line of as-deposited specimen was fitted to three chemical states at 100.54, 102.2, and 103.2 eV, which were attributed to SiC, SiO_xC_y and SiO₂, respectively.^{10, 23} The Si 2p peaks corresponding to the SiO_xC_y and SiO₂ completely disappeared after corrosion of 3 days, as shown in Figure 4B. The

disappearance of SiO_2 peak after corrosion is in agreement with the results reported for the water corrosion of bulk SiC .^{9-11, 13} For C 1s peaks (Figure 4C), significant differences between the as deposited specimen and the corroded specimens were observed. The C 1s peaks of the as-deposited specimen can be distinguished by three peaks, which were observed at 282.5, 284.08 and 286.48 eV correspond to the existence of SiC , SiOxCy and surface absorption impurity(C-O/C-H), respectively.^{24, 25} After 3 days of corrosion, the peak belonging to the C-O-Si bond of SiOxCy centered at 284.08 eV disappeared while a new peak was formed at 284.6 eV, which can be attributed to the C-C bond of carbon. The results suggested the formation of a carbon-rich surface due to the loss of Si from the SiC layer, which is consistent with the Raman analyses. In addition, the C 1s peaks were similar for all the corroded specimens, regardless of the corrosion time. As concluded from Figure 3 and Figure 4, carbon was formed but no sign of any silicon oxides (e.g., SiO_2) was detected on the surface of the corroded specimens.

3.3 The microstructure of the SiC layers

Figure 5 shows the surface and cross-sectional microstructure of SiC layers of TRISO particles after corrosion at 400 °C/10.3 MPa for different time. The surface of as-deposited SiC layer exhibited both a faceted structure with large grains and a globular-like structure with small grains (Figure 5A). After 3 days of corrosion, the surface morphology was still similar to the as-deposited sample (Figure 5B). After 6 days, a slight corrosion occurred and some small grains started being attacked (Figure 5C). In contrast, after 9 days, the small grains were severely attacked and delaminated

from the surface (Figure 5D). After 12 days, individual large SiC grains could be distinguished on the surface and a considerable amount of large SiC grains detached from the surface (Figure 5E). Furthermore, pits were also observed on the surface.

Figure 5 Figure 5 SEM images (A-E) correspond to the SiC surface microstructure after corrosion for different time, (F) and (G) correspond to the SiC fractured cross sectional microstructure for the as deposited sample and 12 days corroded sample, respectively. (H) the FIB section and (I), (J) the bright-field TEM images for the 12 days corroded sample

However, no significant variation was observed between the fractured cross-sections of the as deposited sample and the 12 days corroded sample, as can be seen from Figure 5F and Figure 5G. Therefore, TEM was employed to further characterize the microstructure of the sample corroded for 12 days. Figure 5 (H) shows the FIB section, which was taken on the surface of the sample corroded for 12 days. In Figure 5 (I), an obvious corroded layer at the surface of SiC layer was observed. The source of this corroded layer is that portion of the SiC grains have undergone corrosion and dissolution by the high temperature and pressure water.¹³ In addition, many small pores were also observed at the interface between the corroded layer and the SiC layer. In Figure 5 (J), the corroded grain can be distinguished. These images confirm that the grain boundaries are preferentially attacked.

3.4 Fracture strength of SiC shells

Figure 6 and 7 show the Weibull plots of the local fracture strength (σ_f^L) and the full fracture strength (σ_f^F) of the SiC layer before and after corrosion. Each dataset was linearly fitted to obtain the Weibull modulus m (the slope of the linear fit), which shows the strength distribution. All the results of the crush tests were summarized in Table 1. Both the local fracture strength (σ_f^L) and the full fracture strength (σ_f^F) tended to decrease slightly with an increasing corrosion time. The average values of the local fracture strength reduced from 1356 MPa to 1281 MPa, while those for the full fracture strength reduced from 609 MPa to 590 MPa. However, the Weibull modulus revealed similar values from 6.21 to 6.72 for the local fracture strength and 7.13 to 7.77 for the full fracture strength.

Figure 6 The local fracture strength of SiC shells in the form of Weibull distribution after corrosion for different time

Figure 7 The full fracture strength of SiC shells in the form of Weibull distribution after corrosion for different time

Table 1 Summarized results of the crush tests (with the Weibull modulus given in brackets) of SiC shells after corrosion for different time

Samples	$2r_0$ (μm)	F (N)	σ_f^L (MPa)	σ_f^F (MPa)
As deposited	143.46	5.43	1356 (6.45)	609 (7.13)
3 days	144.12	5.37	1352 (6.21)	604 (7.37)
6 days	147.53	5.35	1290 (6.47)	593 (7.77)
9 days	153.46	5.39	1279 (6.72)	594 (7.37)
12 days	152.53	5.41	1281 (6.64)	590 (7.49)

4 Discussion

4.1 Corrosion kinetics of SiC layers

The hydrothermal corrosion behavior of SiC has been investigated previously.^{11-13, 22, 26} Thermodynamic calculations show that the corrosion of SiC in the hydrothermal environment could occur through the following reactions (12a)-(12d)²⁰:

In the current study, the formation of carbon suggests that the dominant reaction is Equation (12d). The Gibbs free energy change of reaction (12d) at 400°C is -320.02 kJ/mol. Therefore, the rate of SiO₂ formation via reaction (12d), r_I , can be written as:

$$r_I = k_I(a_{\text{SiC}})(a_{\text{H}_2\text{O}})^2 \quad (13)$$

where k_I , a_{SiC} and $a_{\text{H}_2\text{O}}$ are the reaction rate constant, the activity of SiC and the activity of water, respectively. At the reaction interface, SiC and water are abundantly available, the activities are assumed to be unity. Therefore, the rate of silica formation (r_I) can be substituted by the reaction rate constant (k_I).

Meanwhile, the SiO₂ can be dissolved into water via the following reaction:

In this case, the rate of SiO₂ dissolution depends on the pH of water. As reported in previous studies, the rate of SiO₂ dissolution via reaction (14), r_{net} , will have the general form²⁷⁻³¹:

$$r_{net} = k_f \frac{A}{M} \left(1 - \frac{m}{m_{sat}}\right) \quad (15)$$

where k_f (mol/kg-s), A (m^2), M (kg), m , and m_{sat} are the forward reaction rate constant, the surface area, the mass of water, the concentration of SiO₂ in water, and the solubility limit of SiO₂ in water, respectively. It is possible to assume that the concentration of silica in water (m) is insignificant during test.¹¹ Therefore, the net rate of silica dissolution (r_{net}) can be substituted by the forward reaction rate constant (k_f).

Table 2 summarizes the reaction rate constants of SiO₂ formation (k_f) and SiO₂ dissolution (k_f). It can be seen that the SiO₂ dissolution rate constant is 4 magnitude higher than the formation rate constant, indicating that the SiO₂ formation is the rate determining step for the corrosion of SiC layer in the hydrothermal environment, which gives rise to the observed linear corrosion kinetics. This also means that once SiO₂ formed, it dissolves immediately into water. This could explain why no SiO₂ was observed on the surface of the corroded SiC layer.

Table 2 Summary of reaction rate constant of SiO₂ formation and dissolution

Reaction rate constant (mol/m ² -s)	SiO ₂ formation (k_f) ¹¹	SiO ₂ dissolution (k_f) ²⁷
Parameters	2.49×10 ⁻⁹	3.55×10 ⁻⁵

4.2 Microstructure of SiC layers upon hydrothermal corrosion

In the hydrothermal environment, SiO₂ dissolves readily in water and does not maintain an effective protection. Therefore, further corrosion depends on the features of the grain size and grain boundary. For sintered SiC, sintering agents segregate in the grain boundaries and these sintering agents dissolve faster than SiC grains, resulting in preferential dissolution at the grain boundary.³² Previous studies have reported that

preferential intergranular dissolution was a typical phenomenon in the corrosion behavior of sintered SiC under hydrothermal conditions, which depended on the manufacturing routes and the sintering agents.^{8, 21, 33} Compared with sintered bulk SiC, the corrosion of CVD-SiC tends to proceed relatively more uniform because of the higher purity at the grain boundary.³⁴ In this study, small SiC grains were observed to be dissolved faster than large ones. This could be related to the larger fractions of grain boundary areas for smaller grains, which makes them to be more susceptible to the reaction with water.⁹ Therefore, large SiC grains have better corrosion resistance than small ones in the hydrothermal environment. In this manner, the formation of pits can be attributed to the detachment of SiC grains from the surface. On the other hand, previous investigation had proposed that the formation of pits could be caused by local galvanic cell formation due to hydrolysis reactions involving dissolution of SiO₂ and formation of carbon.²² This is in agreement with the current work in terms of the carbon formation.

4.3 Effect of hydrothermal corrosion on the fracture strength of SiC layers

As reported in previous studies, the microstructure such as the roughness of IPyC/SiC interface, the porosity, the grain size and the thickness of SiC layer have effects on the fracture strength of the SiC layers in TRISO particles.^{16, 18, 32, 35-42} Byun et al. have evaluated the fracture strength of the SiC layer using the modified crush test method and concluded that the variation in fracture strength is linked to the inner surface roughness and porosity.¹⁸ Cromarty and Cao et al both reported that the fracture strength of TRISO particles was proportional to the thickness of SiC layers.^{16, 35}

In this study, the roughness of IPyC/SiC interface and, the porosity of SiC layer do not show any change after corrosion. The slight decrease in strength may be due to the reduction in the thickness of the SiC layer. When being tested, the SiC shell is highly stressed under a circular contact area. Since the fracture strength depends sensitively on the contact area of material stressed, thinner specimen has a smaller effective tested area and tends to have higher stress concentration at the inner surface of the SiC shell, where crack would initiate, and propagate through the whole SiC layer resulting in a smaller fracture strength and a smaller Weibull modulus.^{16, 18} However, it has to be noted that the crush test is expected to contain considerable experimental errors due to the irregularities and variations in the shape and size of specimens, as pointed out by previous studies.^{17, 18} Taking into account the experimental errors, the changes in the full fracture strength (609 MPa to 590 MPa) and the Weibull modulus (7.13 to 7.77) suggest that the mechanical strength of the samples decrease slightly or almost unchanged after hydrothermal corrosion.

5 Conclusions

The hydrothermal corrosion of SiC layers in TRISO particles was investigated in the static autoclave with the high temperature and pressure water (400 °C/10.3 MPa). No SiO₂ was detected but carbon was formed on the surface of corroded specimens. The thickness of SiC layer decreased slightly with the increase of the exposure time. The formation of SiO₂ was analyzed to be the rate determining step for the corrosion of SiC layer, leading to the observed linear corrosion kinetics. The corrosion mechanism of SiC layer was the preferential corrosion at the grain boundary, and the formation of

pits can be attributed to the detachment of SiC grains from the surface. The slight decrease in strength values with the increase of corrosion time may be due to the reduction in the thickness of the SiC layer. These results indicate that the SiC layers of TRISO particles have exceptionally good corrosion resistance in the hydrothermal environment.

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