On the Dimensional Change of Coated Fuel Particles upon Irradiation Testing

Young-Woo Lee[1*], Jihae Yoon[1], Yeon-Ku Kim[2], Seunghwan Yeo[1],
Youngjun Kim[3], Heemoon Kim[3], Moon Sung Cho[4], Eung Seon Kim[2]
[1]*Advanced Nuclear Fuel Technology Development Div.,* [2]*Material PIE & Radwaste Disposal Div.,*
[3]*VHTR Technology Development Div.,* [4]*Nuclear Education & Training Center*
Korea Atomic Energy Research Institute
1199 Deokjin-Daero, Yuseong-gu, Daejeon, Republic of Korea
[*]*Corresponding author: ywlee@kaeri.re.kr*

1. Introduction

KAERI has performed its first HTR fuel irradiation test utilizing its Hanaro research reactor from August 2013 to March 2014 for 132 EFPD. A detailed description of the irradiation test has been presented. [1] In this test, 2 test rods were encapsulated in the test capsule, in which one rod was loaded with 9 fuel compact specimens and the other rod with 5 fuel compacts and 4 sets of graphite specimens. One set of graphite specimen consists of a pair of 1 matrix graphite disc and 1 IG-110 structural graphite disc.

After the irradiation test, a series of post irradiation examinations (PIEs) were carried out on the selected irradiated compact specimens and the graphite matrix specimens, in order to understand the irradiation behavior and performance of the coated particle fuel as well as the variation of the properties of A3-3 matrix graphite and IG-110 structural graphite. In this study, the dimensional change of coated fuel particles after irradiation test was investigated on the UO_2 kernel and three coating layers surrounding the kernel (buffer layer, inner pyrolytic carbon (PyC) and SiC layers) by measurement with X-ray micro-radiography by comparison with non-irradiated coated fuel particles.

2. Experimental

2.1. Irradiation test conditions in Hanaro reactor [1]

Fig. 1 shows a schematic construction layout of encapsulated two fuel test rods in the irradiation capsule, which was a non-instrumented device. The fuel compact and graphite specimens were loaded in graphite sleeves, clad in the stainless steel 316L tubes and end-cap welded. The cladding tubes were approximately 16 mm in diameter and 150 mm in height including the plenum space. The temperature of the fuel test rod specimens was controlled by filling mixtures of He and Ne gases when fabricating fuel test rods: a mixture of 30% He and 70% Ne for the fuel rod 1 and a mixture of 10% He and 90% Ne for the fuel rod 2, respectively.

During the BOC of the irradiation testing, the calculated peak temperature of the fuel compact, which was axially located in the middle of test rod 1, was calculated to be about 1306K under a mixed gas atmosphere of 30% He and 70% Ne, and the calculated peak temperature in test rod 2 was about 1045K under a mixed gas atmosphere of 10% He and 90% Ne.

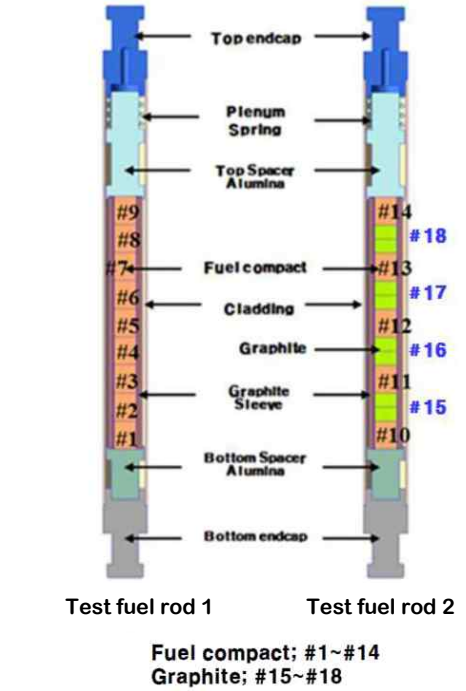


Fig. 1. Schematic construction layout of the 2 fuel rod specimens in the test capsule [1]

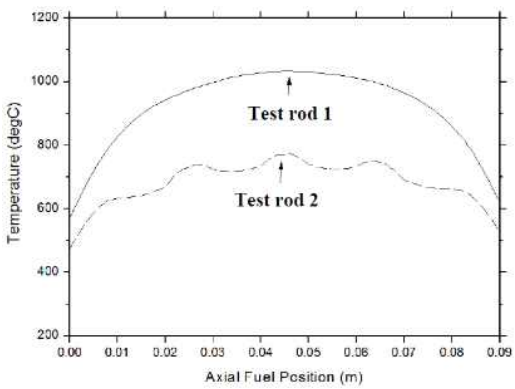


Fig. 2. The estimated axial temperature distribution of the fuel test rod 1 and 2 before irradiation test [1]

The maximum power of fuel compact is estimated to be 56 W at 25.06 EFPD, and the maximum power of particle is 215.4 mW in rod 1. The maximum discharged burn-up is about 37,344 MWD/MTU (3.99 FIMA (Fissions per Initial Metal Atom)). For the rod 2, the maximum power of fuel compact is 50 W, and the maximum power of particle is 197.5 mW. The maximum discharged burn-up was about 35,698 MWD/MTU (3.81% FIMA). The maximum fluence of graphite specimen was 2.99×10^{20} n/cm^2 (E > 0.18 MeV).

2.2. Selection and preparation of coated fuel particle specimens for X-ray radiographic analysis

The number of specimens selected for the analysis was 4 compacts, two from the rod 1 and the other two from the rod 2, which had been located in the positions of 4 and 5 in the rod 1 and 11 and 14 in the rod 2 numerated in Fig. 2, which, respectively, are designated as compacts A5, B15, A3 and B2 hereafter. For the radiographic analysis, the above-mentioned four compacts were deconsolidated (particles separated from the graphite matrix of the compacts) by burning the compacts in a furnace at 1123K in an air atmosphere for 5 up to 8.5 hrs depending on the specimens to convert graphite material of the compacts into CO_2 gases. In this way, only coated fuel particle specimens without outer PyC layer could remain, as the outer PyC also burnt away as CO_2 gas together with graphite material during burning the graphite matrix.

2.3. X-ray radiography dimensional measurements

For the measurement of kernel diameter and coating thicknesses of buffer layer, inner PyC layer and SiC layer, radiographic images were obtained by using a software of an personal computer attached to the main X-ray radiography equipment, purchased from YXLON Co., Germany of which the capacity is 160kV and the technical details are given in Table 1. Specimen images were obtained at 70kV and 70μA, four images from each coated particle specimen at the position in different angles by turning the particle specimen holder to 0, 90, 180 and 270 degrees. 10 irradiated coated particles were sampled out of 263 particles separated from each irradiated compact specimen numbered here above, i.e., A5, B15, A3 and B2. The method of sampling applied for this measurement led to the measurement on 40 images. For comparison, un-irradiated particles were also measured for kernel diameter, and thicknesses of buffer, inner PyC and SiC layers. An image analysis system software was used to measure the dimensions of particle on the particle images. An example is given for a typical particle image in Fig. 3.

Table 1. Technical details of X-ray radiographic equipment

X-ray Tube	Tube type	Open micro-focus tube
	Target material	Tungsten
	Voltage range	25 – 160 kV
	Current range	0.01 – 10 mA
	Detectability	< 1 μm
Image Chain	Detector type	Panel 1313 high speed
	Number of pixel	1,004 x 1,004 pixel
	Pixel size	127 μm
	Total magnification	17,000x

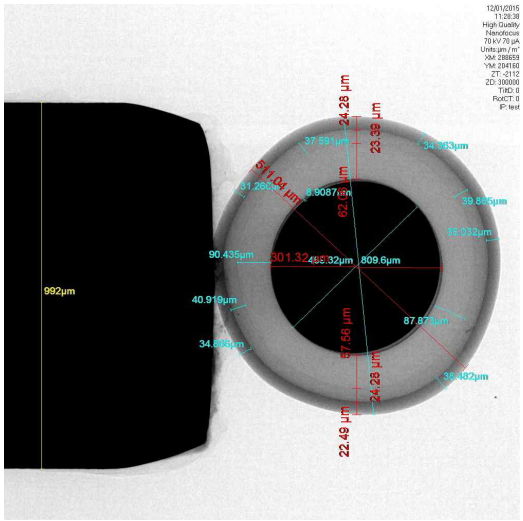


Fig. 3. An example of dimensional measurement on a coated particle X-ray image (a separated irradiated particle from deconsolidation of B2 compact)

3. Results and Discussions

For the un-irradiated particle specimens, values are compared in Table 2 between the average values measured on X-ray images and those by ceramographic measurements on the optical micrographs. The standard deviations for kernel diameter and thicknesses of buffer, inner PyC and SiC layers range 8.0-20.0(μm), 4.0-8.0(μm), 0.8-1.5(μm), and 0.8-1.05(μm), respectively. Except measurements for kernel diameter of batch B and buffer layer thickness of batch A, results of measurements obtained by both methods agree fairly well.

Table 3 shows the average values of dimensional changes and change rates (in percentage) of kernel diameter and thicknesses of buffer, inner PyC and SiC layers after irradiation test. Here, dimensional change and change rate are defined as; $(X_{bi}\text{-}X_{ai})$ and $(X_{bi}\text{-}X_{ai})/X_{bi}x100$, respectively, where, X_{bi} and X_{ai} denote

dimension before irradiation and dimension after irradiation, respectively.

Table 2. Comparison of dimensional measurements of between X-ray and ceramographic measurements of un-irradiated coated particle specimens

	Kernel diameter (μm)	Buffer thickness (μm)	IPyC thickness (μm)	SiC thickness (μm)
Batch A, X-ray measured	**468.60**	**103.86**	**39.11**	**35.58**
Batch A, ceramography	**469.80**	**93.00**	**40.02**	**35.93**
Batch B, X-ray measured	**480.77**	**105.55**	**39.86**	**34.98**
Batch B, ceramography	**470.30**	**112.82**	**41.08**	**36.23**

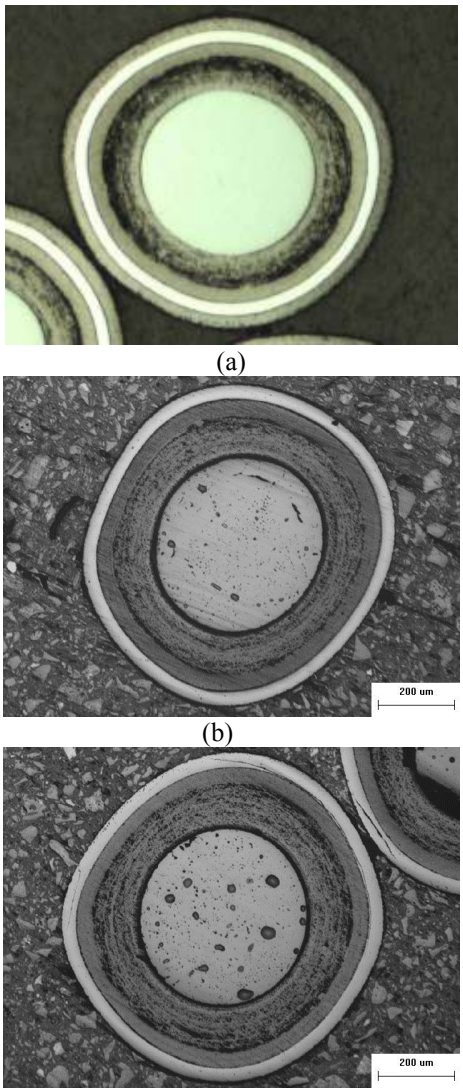
Table 3. Dimensional change of coated particle specimens after irradiation.

	kernel diameter	buffer thickness	IPyC thickness	SiC thickness
A3 change average, μm	**8.09**	**3.65**	**-2.23**	**-1.05**
A3 change rate, %	**1.72**	**3.92**	**-5.58**	**-2.94**
A5 change average, μm	**12.96**	**7.93**	**-5.05**	**-2.83**
A5 change rate, %	**2.76**	**8.52**	**-12.62**	**-7.87**
B2 change average, μm	**11.86**	**-14.54**	**-1.84**	**-0.29**
B2 change rate, %	**2.52**	**-12.89**	**-4.49**	**-0.81**
B15 change average, μm	**9.00**	**-12.95**	**-2.43**	**-3.16**
B15 change rate, %	**1.91**	**-11.48**	**-5.92**	**-8.71**

3.1. Dimensional change of kernel in the coated particle specimens after irradiation

The values in the measurements seem indicate, at a first glance, that the kernel swelled and the inner PyC and SiC layers shrank. The changes in buffer layers do not show a consistent change; some indicate + and others -. However, for the kernel dimensional change which indicates a swelling by their values shown in Table 2, if the micrographs of the un-irradiated particle (Fig 4(a)), of the irradiated particles (Fig. 4(b) from A3 compact and Fig. 4(c) from compact A5) in Fig. 4, are compared, it is most likely that the UO_2 kernels shrank after irradiation. This confusion can be attributed to the dimensional measurement method of X-ray radiography. The gap (or de-bonded) region between outer periphery of the kernel and inner periphery of the buffer layer cannot be distinguished in the dark area on the particle image, and hence, this gap is included when measuring the diameter of the kernel. In fact, the ceramographic measurement result estimated about 448μm and 440μm for A3 and for A5 of irradiated particle kernel, respectively, compared with 469.8μm for A batch of un-irradiated particle kernel, given in Table 2.

Recently, Bower et al. [2] obtained a kernel volume increase of about 26% in UCO up to high burnup of 16.1% FIMA (fission per initial metal atoms). The average burnup estimated in Hanaro irradiation test in this work is, as mentioned in *2.1*, 3.81% FIMA, very low compared with the work carried out by Bower et al. Also, as discussed [2], fuel swelling can be dependent on a number of fuel properties and irradiation conditions, including different fuel kernel composition (UO_2 vs. UCO).



(a)

(b)

(c)

Fig. 4. Ceramographic micrographs of an un-irradiated particle (a), an irradiated particle from A3 (b) and an irradiated particle from A5 (c)

3.2. Dimensional change of PyC layers in the coated particle specimens after irradiation

It is known that the CVD PyC shrinks upon neutron irradiation, if it is isotropic and of high density. [3] The dimensional changes of inner PyC layers shrank after

irradiation in this work, as shown in Table 3. The irradiation-induced shrinkage seems also to depend on the sample position in Rod 1 and 2 (temperature) and at low level of fluence dose of about 2.99×10^{24} n/m^2 (E > 0.18 MeV).

The buffer PyC layer does not behave as isotropic inner PyC in this work. Bower et al. reports a densification of buffer layer of the UCO coated particle irradiated up to 16.1% FIMA. [2] The values shown in Table 3 for the buffer layer are not consistent: The buffer layers of the coated particle specimens from batch A swelled while those from batch B shrank. The difference in material properties of these two batches was essentially from the density of inner PyC layers. The density of the inner PyC layer of batch A is 1.75 g/cm^3 while that of batch B is 2.01 g/cm^3. In fact, the inner PyC layers of coated particle from batch A (A3 and A5) showed a little larger shrinkage than those from batch B, which is generally true on the assumption that the shrinkage is dependent on density; the lower the density, the higher the shrinkage. Now, on the assumption that the buffer layer is bonded with the adjacent inner PyC layer the shrinkage of inner PyC layer causes pulling the buffer layer, hence, the latter swells, i.e., the inner PyC with a lower density in the coated particle from the batch A pulls the bonded buffer layer by its higher shrinkage. However, for the batch B particles the shrinkage of the inner PyC is less and the buffer layer remained shrunk. These discussions are absolutely qualitative, and there can be always measurement uncertainties which can be an important factor to be taken into account, which is particularly true for the dimensional measurement on the buffer layer with fairly un-distinctive boundaries after irradiation.

3.3. Dimensional change of SiC layer in the coated particle specimens after irradiation

It is generally known that CVD-SiC swells upon neutron irradiation with a sufficient dose at high temperatures. Fig. 5 illustrates a general swelling behavior of SiC plotted with historical compiled data accumulated over thirty years. [4] Table 3 shows that the thickness of SiC of the coated particle specimens is become thinner after irradiation testing. This is contradictory to the observation on Fig. 5. Unfortunately the irradiation condition in this work cannot be included in the range covered by this figure: with such a low dose of neutron irradiation, it can be assumed that the SiC layer first be amorphized and then this amorphized SiC phase with newly formed material defects such as voids by neutrons be densified. Or, it is possible that the some mechanical interaction or stress state between the SiC layer and the bonded inner PyC layer may affect the thickness of the SiC layer.There is another factor that could influence the final characteristics of the irradiated particle specimens: To get particle specimens out of compacts, the compact specimens had been burnt at 1123K in air. This heat treatment could also modify the coating layers, the SiC layer in particular, as well as its microstructure. However, at this point, neither ways can explain such a different observation from the previously published works, and further quantitative examination must be carried out on this observation.

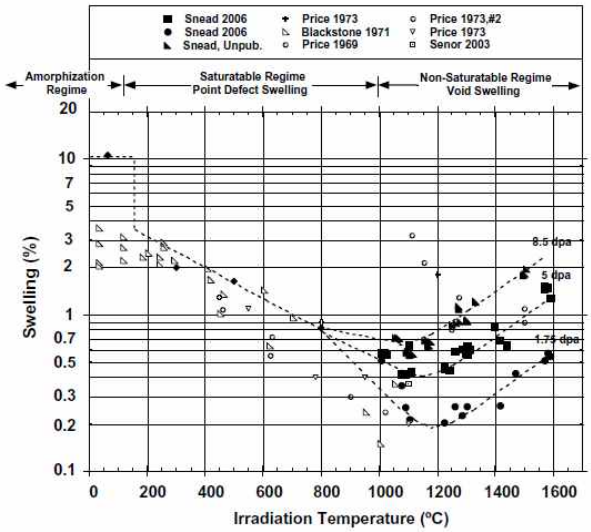


Fig. 5. Irradiation-induced swelling of SiC to high temperatures, reproduced from [4]

4. Summary

In this study, the dimensional change of coated fuel particles after irradiation test was investigated on the UO_2 kernel and three coating layers surrounding the kernel (buffer layer, inner pyrolytic carbon (PyC) and SiC layers) by measurement with X-ray micro-radiography and comparison with non-irradiated coated fuel particles. The irradiation test carried out in this work achieved a maximum burnup of 3.81% FIMA and was with fairly low neutron dose of about $0.3\text{x}10^{25}$ n/m^2. The estimated maximum temperature was below 1073K. The results obtained in this work are quite different from those by recent work with high burnup of 16.1% FIMA for UCO for kernel and buffer [3] and by other works for SiC [4] with rather high dose up to 8 dpa (for SiC) and high temperatures up to 1600K. There are other possibilities of causing different results are; measurement methods, preparation methods of irradiation specimens and others. In this respect, some further precise and quantitative examinations and analyses are needed for the logical explanations.

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