Diffusivity of Xe-133 in Irradiated Uranium Nitride with a Low Burnup(< 0.1 MWd/t-U)

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1. Introduction

Nitride fuels have been studied as a fuel of a FBR with the advantages of high density and short conversion time. Those fuels have a compatibility with sodium as a coolant of the FBR. High melting point and thermal conductivity of nitride fuel are better than those of oxide fuel. Usually, (U,Pu)N has been studied for blanket materials rather than UN. So many papers referred to the fuel performance of (U,Pu)N. But, UN must be evaluated firstly to know the thermal properties of nitride fuels. Especially, the diffusion behavior of xenon in UN has not been studied sufficiently, so it is difficult to find those data. In this study, we tried to obtain xenon diffusion behavior by a post irradiation annealing test.

2. Experimental

2.1 Specimen Preparation

Four UN and two UO_2 samples were made with 45.2% and 47.6% of porosity, respectively. Those samples were disk shape with below 300 mg as shown in Fig.1.

As the results of BET, 0.2434 m^2/g for UN and 0.3132 m^2/g for UO_2 were obtained.

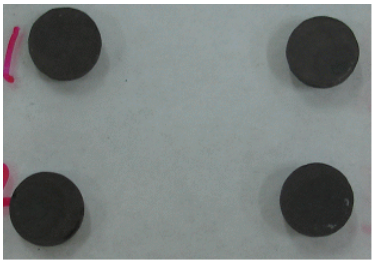

Fig. 1 UN samples

SEM of samples was carried out to obtain the microstructure and grain size.

2.2 Apparatus

Annealing system consisted of a furnace and a filtration. The chamber of the furnace was made by high purity alumina. Two thermocouples were installed; one is to measure the sample temperature and the other is to measure an arbitrary temperature(<1,000℃) for the oxygen partial pressure. The furnace can be raised up to 1650 ℃. Filtration material was granular charcoal and it was placed in liquid nitrogen to catch the xenon gas under a solidification. Carrier gas was helium(10% of hydrogen) and flowed at 0.1 ml/min. during an annealing.

2.3 Irradiation and annealing

Irradiation and annealing for each fuel sample were carried out once a week. 6 fuel samples were contained in each aluminum capsule and were irradiated in the HANARO research reactor for 16 minutes. After a week for a cooling time, samples were loaded into the furnace and heated up at 1200℃~1400℃ for UN and at 1400℃ ~ 1600℃ for UO_2. Annealing time was all most 10~11 hours. Xenon released out from a sample was carried to the filtration system with helium gas and trapped in the charcoal surrounded by liquid nitrogen. This accumulated xenon(Xe-133) in the filtration system was measured by a HPGE detector with every set time continuously. Annealing time, temperature and activity of released Xe-133 were applied to a diffusion equation to obtain the diffusion coefficients.

3. Results

4 UN samples(UN-1,UN-2,UN-3,UN-4) were annealed at 1400℃ ~ 1600℃ for UN-1 and UN-2 and at 1200℃~1400℃ for UN-3 and UN-4. Fig.2 shows the fractional release of Xe-133 with time for UN-3 and UN-4. Annealing tests for the UO_2 samples were performed in the same way as the UN samples for a comparison. But, the temperature range was higher due to a low release rate at 1200℃. Fig.3 shows the data of UO_2. When we calculated the fractional release, the ORIGEN-2 code was used to calculate the radioactivity of Xe-133 produced and released, following a gamma detection with a Ba-133(81keV) reference source in the same geometry for a Xe-133 detection. To obtain the diffusion coefficient(D) graph of f^2.vs. t was needed as in Fig.1 and Fig.2. The slopes at each temperature in the graph were obtained. From the following equations, D was calculated.

$$S = \frac{36D}{\pi a^2} \qquad \text{Eq.(1)}$$

Where, 'a' is the equivalent radius which can be obtained by the volume and surface area of a sample. 'S' is the slope of f^2.vs.t at each temperature.

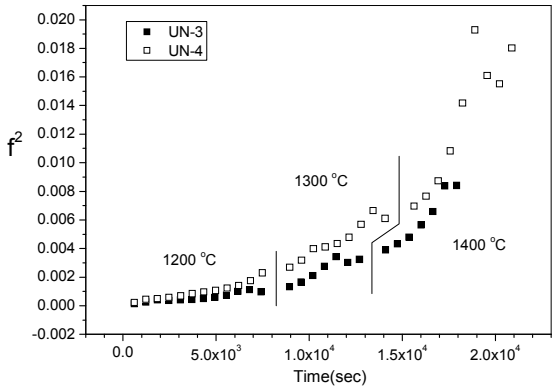

Fig. 2 Released fractions for UN-3 and UN-4

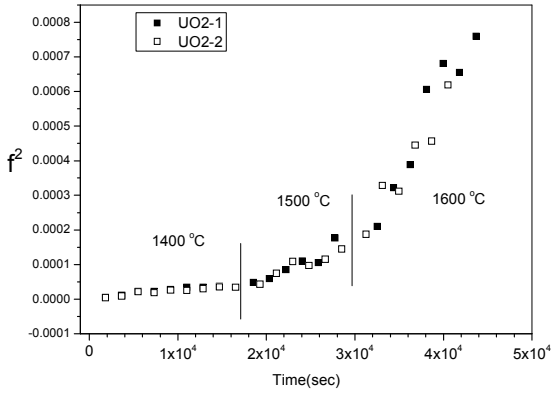

Fig. 3 Release fractions for UO_2 samples

Using the values of slope and 'a', D can be obtained as shown in Fig.4 for UN samples and Fig.5 for UO_2 samples. The fractional release in UN samples was higher than that in UO_2 samples.

General diffusion coefficient equations for UN and UO_2 are as bellows;

$$D(mm^2/s) = 8.765\times10^{-6}\exp\left(-\frac{230(kJ/mol)}{RT}\right) \text{ for UN}$$

$$D(mm^2/s) = 1.437\times10^{-3}\exp\left(-\frac{393(kJ/mol)}{RT}\right) \text{ for } UO_2$$

4. Conclusion

To study of the diffusivity of Xe-133 in UN fuel, we carried out a post-irradiation annealing test. The samples were made with a disk type and high porosity. UO_2 samples were made as well to compare to the UN sample. Those samples were irradiated in the HANARO research reactor for 16 minutes to prevent a bubble creation. Released Xe-133 activity was detected while a sample was annealed at a set temperature. From the results of the Xe-133 activity produced and released, the fractional release rate was obtained and then, the diffusion coefficients were calculated. UN fuel showed higher release rate and diffusivity than UO_2 fuel. Activation energy for UN is lower than that for UO_2.

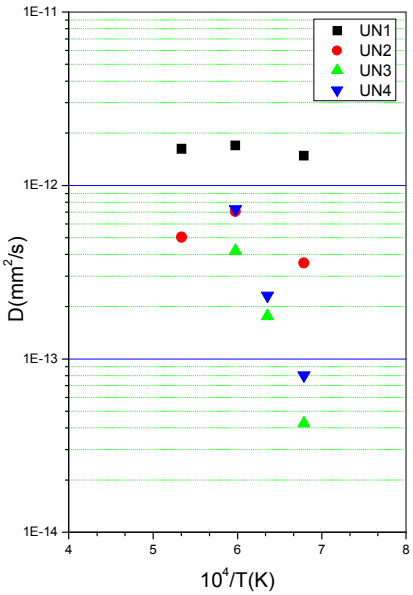

Fig. 4 Diffusion coefficients of UN samples at each temperature

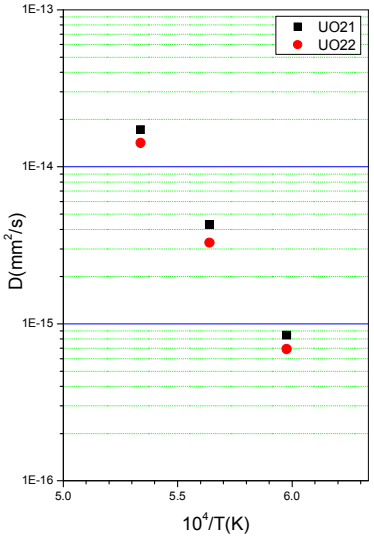

Fig. 5 Diffusion coefficients of UO2 samples at each temperature

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[2] Hj. Matzke, Radiation Effects, "Radiation enhanced diffusion in UO_2 and $(U,Pu)O_2$", 75 , 317 (1983)