# Measurement of Fission Gas Release with Additives in UO<sub>2</sub> at Low Burnup

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### **1. Introduction**

The fission gas release is one of important behaviors in the fuel performance. It makes thermal conductivity of gas in a gap of fuel rod low then, the fuel temperature and the internal pressure are increased, which cause fuel life to be short. The fission gas diffusion in  $UO_2$  is the dominant behavior and must be the factor obtained by experiments. Especially, the diffusion coefficient is fundamental parameter to establish a diffusion equation. It is obtained by two types of the tests; one is in-pile test which is simulated reactor and obtained the apparent diffusion coefficient, the other is post-irradiation annealing test which is basic system to observe several diffusion mechanisms with step by step as sample preparation. Generally, the diffusion coefficient is affected by temperature, stoichiometry, irradiation effect and valence of fission products.

In this study, the PIA test(post-irradiation annealing test) with the UO<sub>2</sub> + additives(+3, +4, +5 valence) was carried out to observe the valence effect.

#### 2. Experimental

## 2.1 Annealing apparatus

The system for the PIA test was installed in IMEF(Irradiated Materials Examination Facility) and consists furnace, filtration and detector as shown in Fig.1. The furnace is electric resistance type with a super kanthal heat source. It can be raised up to 1600 °C and the zirconia oxygen sensor was installed. Helium gas as the carrier is good to transport xenon gas into the cryogenic filtration.



Fig. 1 The apparatus for PIA test (IMEF)

The filtration was made with the charcoal contained in the pyrex glass chamber. It was placed into liquid nitrogen to catch the xenon gas as the solid adsorption. The lead box covered the filtration system and gamma detector was attached on it to count the released Xe-133 gamma rays. The detector is the HPGe(high purity germanium) semi-conductor type and cooled down by the liquid nitrogen.

#### 2.2 Sample preparations

The fuel samples were made with the fresh UO<sub>2</sub> and additives. The additives were Nd(+3), Ce(+4) and Nb(+5). Those were added in UO2 with contents of 1.6 w/o, 3.2 w/o and 6.5w/o. All samples were disk type with 4.5~4.7 mm of the dia. and 1.4~1.6 mm of the thickness as well as 95%~97% of the average density. BET of the samples were measured and the values were  $1.0 \times 10^{-7} \text{ m}^2/\text{g} \sim 9.68 \times 10^{-8} \text{ m}^2/\text{g}.$ 



Fig. 2 UO<sub>2</sub> fuel sample



Fig. 3 The zry-4 tube contained a fuel sample

The samples were contained into zry-4 tube and welded in helium atmosphere. After irradiation for 16 minutes in HANARO reactor, the sample tube was dismantled and the sample was contained in Al<sub>2</sub>O<sub>3</sub> crucible to be loaded in furnace for the annealing test.

### 2.3 The annealing procedure

To obtain radioactivity of the released and the generated Xe-133, Ba-133, standard source, was

detected at the same as geometry of Xe-133 due to gamma energy of Ba-133(81 keV) which is same as Xe-133. Before the annealing, total gamma scanning for  $Al_2O_3$  crucible was carried out to obtain the radioactivity of Xe-133 in the irradiated fuel sample.

ORIGEN-2 code was used to obtain the burnup and the generation of Xe-133.

Annealing was performed at 1400 °C for 15 hours, 1500 °C for 9 hours and 1600 °C for 6 hours with continuous. The gamma detector was activated in filtration with every 3600 sec. and the released Xe-133 peaks were obtained in all temperature ranges. The carrier gas(He+H<sub>2</sub>(10%)) was flowed with 0.1 l/min. and transferred the released Xe-133 to the filtration system.

#### 3. Results

The fuel samples were moved to irradiation facility and annealing system was set up so far. Three samples of UO<sub>2</sub>+Nd(+3) will be irradiated from April  $8^{th}$  to April  $22^{th}$  with one sample per a week. Annealing test will begin from April  $8^{th}$ . The results of UO<sub>2</sub>+Nd(+3) will be came out before the KNS Spring Conference.

In the case of the uranium vacancy controlled diffusion, +3 valent dopants reduce the cat-ion vacancy concentration, therefore xenon mobility is slower. It means the released fraction is lower than that of pure UO<sub>2</sub>. The other hand, +5 valent dopants make the xenon mobility faster, so the released fraction is higher due to the higher concentration of cat-ion vacancy as shown in fig.4[1]. But,  $Cr_2O_3$  in Fig.5 was different behavior due that  $Cr^{3+}$  would be interstitial site[2].

Mazke referred that no difference between dopants was observed as shown in Fig.6, so xenon does not diffuse via uranium vacancy[3]. He introduced the tri-vacancy cluster which consists of a uranium vacancy and two oxygen vacancies.

#### 4. Conclusions

This study is to observe whether the xenon diffusion is affected by various valent additives. In addition, the difference of xenon diffusion will be checked with amount of additives.

## REFERENCES

- K.Une, I.Tanabe, M.Oguma, Effects of Additives and The Oxygen Potential on The Fission Gas Diffusion in UO<sub>2</sub> Fuel.', J.Nucl.Mater.150, 93p.(1987)
- [2] S.Kashibe, K.Une, 'Effect of additives (Cr<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, MgO) on diffusional release of Xe-133 from UO<sub>2</sub> fuels.', J.Nucl.Mater **254**, 234p.-242p.(1998)
- [3] Hj.Matzke, 'Diffusion in Doped UO<sub>2</sub>.', Nuclear applications. 2, 131p.(1966)

[3] Mutsumi Hirai, "Thermal diffusivity of UO<sub>2</sub>-Gd<sub>2</sub>O<sub>3</sub> pellets", J.Nucl.Mater., **173**, 247p.(1990).



Fig. 4 Diffusion coefficients for  ${\rm Ti}^{+4}$  and  ${\rm Nb}^{+5}$  additives in  $UO_2[1].$ 



Fig. 5 Diffusion coefficients for  $Cr^{+3}$ ,  $Al^{+3}$ ,  $Mg^{+2}$  and  $Si^{+4}$  in UO<sub>2</sub> [2].



Fig. 6 Diffusion coefficients for Nb<sup>+5</sup>, Zr<sup>+4</sup>, La<sup>+3</sup> and Y<sup>+3</sup> in  $UO_2[3]$ .