

Behavior of Fission Gas Release with Valence Effect in irradiated UO₂

Heemoon Kim^{a*}, Kwangheon Park^b, Sang-Ho Na^a, Chang-Je Park^a, Kweon Ho Kang^a, Man Soon Cho^a,
Ung Sup Song^a, Ki Ha Kim^a, Sang Bok Ahn^a

^aKorea Atomic Energy Research Institute, Yuseong, Daejeon, 305-353 Rep. of Korea

^bDept. of Nuclear Eng., Kyunghee Univ., Yongin, Kyunggi., Rep. of Korea

*Corresponding author: hkim1211@kaeri.re.kr

1. Introduction

Fission gas release is one of the most important behaviors in fuel performance, which is related to the fuel temperature and the internal pressure. The diffusion coefficient of fission gas is a fundamental parameter to establish a diffusion equation. This was obtained by the post-irradiation annealing test, which is a basic system to observe several diffusion mechanisms with a multiple steps as the sample preparation conditions. Generally, the diffusion coefficient is affected by temperature, stoichiometry, the irradiation effect, and the valence of fission products.

In this study, the PIA test(post-irradiation annealing test) with UO₂ + additives(+3, +4, +5 valence) was carried out to observe the valence effect[1,2].

2. Experimental

2.1 Annealing apparatus

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



Fig. 1 The apparatus for PIA test (IMEF)

The filtration system was made with the charcoal contained in a pyrex glass chamber. It was placed into liquid nitrogen to catch xenon gas as a solid adsorption. A lead box covered the filtration system and a gamma detector was attached on it to count the released Xe-133

gamma rays. The detector was a HPGe(high purity germanium) semi-conductor type and cooled down by liquid nitrogen.

2.2 Sample preparations

The fuel samples were made with fresh UO₂ and additives. The additives were Nd(+3), Ce(+4) and Nb(+5). These were added in UO₂ with contents of 1.6 w/o, 3.2 w/o and 6.5w/o as shown in Table.1. All samples were a disk type 4.5~4.7 mm in dia. and 1.4~1.6 mm thickness, as well as 95%~97% of the average density as shown in Fig.2. BET of the samples were measured and the values were $1.0 \times 10^{-7} \text{ m}^2/\text{g} \sim 9.68 \times 10^{-8} \text{ m}^2/\text{g}$.

Table.1 Sample properties

Index	Additive	Contents(w/o)
10F-03U	Nd ₂ O ₃	1.6
10F-04U	Nd ₂ O ₃	3.2
10F-05U	Nd ₂ O ₃	6.5
10F-06U	CeO ₂	1.6
10F-07U	CeO ₂	3.2
10F-08U	CeO ₂	6.5
10F-09U	Nb ₂ O ₅	1.6
10F-10U	Nb ₂ O ₅	3.2
10F-11U	Nb ₂ O ₅	6.5



Fig. 2 UO₂ fuel sample

The samples were contained in a zry-4 tube and welded in a helium atmosphere as shown in Fig.3.

After irradiation for 16 minutes in the HANARO reactor, the sample tube was dismantled and the sample was contained in a Al₂O₃ crucible to be loaded in furnace for the annealing test.



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

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 the gamma energy of Ba-133(81 keV) which was the same as Xe-133. Before the annealing, a total gamma scanning for Al₂O₃ 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 13 hours, 1500 °C for 9 hours, and 1600 °C for 6 hours continuously. The gamma detector was activated in filtration every 3600 sec. and the released Xe-133 peaks were obtained in all temperature ranges. The carrier gas(He+H₂(10%)) flowed with 0.1 l/min. and transferred the released Xe-133 to the filtration system.

3. Results

Fractional release of Xe-133(f^2) was plotted with time in every sample. The slope of each temperature in each plot was used to obtain diffusion coefficient of Xe-133. Tri-valent additive(Nd) reduced the amount released while penta-valent additive(Nb) increased. With contents of each additive, the tests of Nd and Ce showed irregular behavior but the tests of Nb were shown in good trend. Fig.4 shows diffusion coefficients of all samples and shows a significant difference between Nd and Nb data. Three orders of difference were observed in tri and penta valence effects in the same contents. In the case of Nd and Ce, one order of difference would be showed between contents even it was not followed the trend. The case of Nb showed a narrow difference with its contents.

4. Conclusions

In the case of uranium vacancy controlled diffusion, +3 valent dopants reduced the cat-ion vacancy concentration, therefore xenon mobility was slower. This means the released fraction was lower than that of pure UO₂. On the other hand, +5 valent dopants made the xenon mobility faster, so the released fraction was higher due to the higher concentration of cat-ion vacancy as shown in fig.4. Mazke referenced that no

difference between dopants was observed, 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. Whether xenon moves via uranium vacancy or tri-vacancy cluster, xenon mobility is controlled by concentration of uranium vacancy.

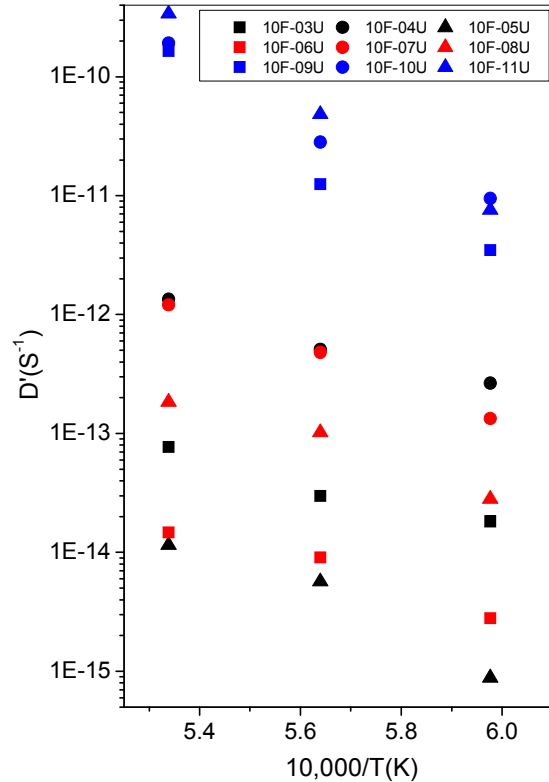


Fig. 4 Diffusion coefficients($D'=D/a^2$) in every sample.

REFERENCES

- [1] K.Une, I.Tanabe, M.Oguma, 'Effects of Additives and The Oxygen Potential on The Fission Gas Diffusion in UO₂ Fuel.', J.Nucl.Mater. **150**, 93p.(1987)
- [2] S.Kashibe, K.Une, 'Effect of additives (Cr₂O₃, Al₂O₃, SiO₂, MgO) on diffusional release of Xe-133 from UO₂ fuels.', J.Nucl.Mater **254**, 234p.-242p.(1998)
- [3] Hj.Matzke, 'Diffusion in Doped UO₂.', Nuclear applications. **2**, 131p.(1966)