# Diffusivity Measurement of Xenon Atom in Irradiated Doped $UO_2$

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## 1. Introduction

Xenon which is generated by fission in $UO_2$ is released out from a $UO_2$ pellet. The release of xenon has been studied with the diffusion in solid. The diffusion of xenon is controlled by stoichiometry of $UO_2$, the irradiation effect and the valence of fission products as well as temperature.

In the case of fission products, mobility of xenon is different with their valences. In this study, the PIA test(post-irradiation annealing test) with $UO_2$ + dopants(+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 $^oC$ and a zirconia oxygen sensor was installed. Helium gas, as the carrier, was good to transport xenon gas into the cryogenic filtration system.

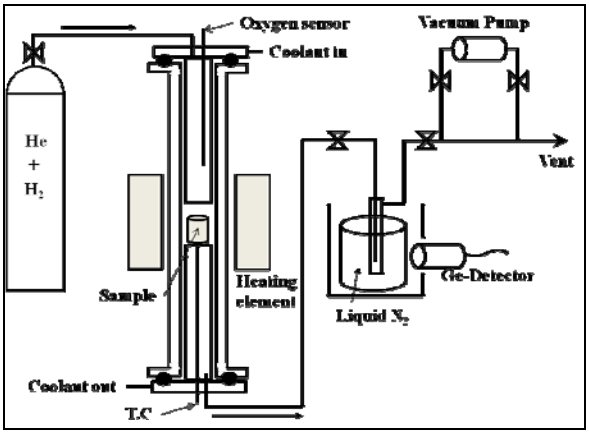


Fig. 1 The apparatus for PIA test (IMEF)

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 $UO_2$ and dopants. The dopants were Nd(+3), Ce(+4) and Nb(+5). These were added in $UO_2$ with contents of 1.6 w/o, 3.2 w/o and 6.5 w/o as shown in Table.1. Fig.2 shows grain size of each sample. Grain size of two pure UO2 samples was 30±2 μm.

Table.1 Sample properties

| *Index* | *Additive* | *Contents(w/o)* |
|---|---|---|
| 10F-03U | $Nd_2O_3$ | 1.6 |
| 10F-04U | $Nd_2O_3$ | 3.2 |
| 10F-05U | $Nd_2O_3$ | 6.5 |
| 10F-06U | $CeO_2$ | 1.6 |
| 10F-07U | $CeO_2$ | 3.2 |
| 10F-08U | $CeO_2$ | 6.5 |
| 10F-09U | $Nb_2O_5$ | 1.6 |
| 10F-10U | $Nb_2O_5$ | 3.2 |
| 10F-11U | $Nb_2O_5$ | 6.5 |
| 10F-12U | Pure $UO_2$ | |
| 10F-13U | Pure $UO_2$ | |

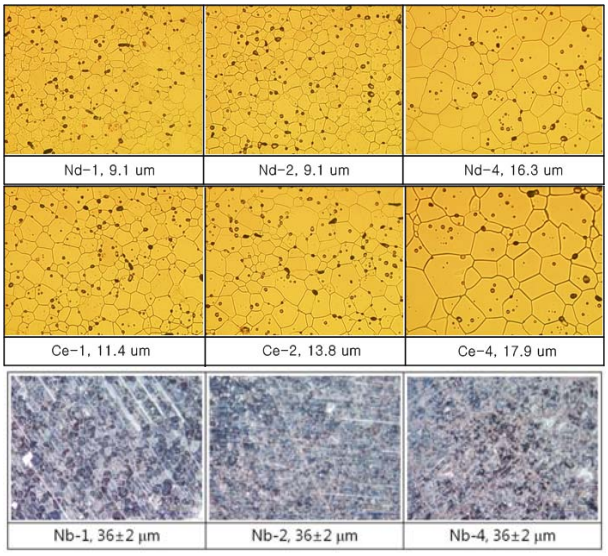


Fig. 2 Grain size of each sample

All samples including two pure $UO_2$ 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. BET of the samples were measured and the values were $1.0x10^{-7}$ $m^2/g$ ~ $9.68\ x10^{-8}$ $m^2/g$.

After irradiation for 16 minutes in the HANARO reactor, the sample tube was dismantled and the sample was contained in a $Al_2O_3$ 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 the gamma energy of Ba-133(81 keV) which was the same as Xe-133. Before the annealing, a 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 $^oC$ for 13 hours, 1500 $^oC$ for 9 hours, and 1600 $^oC$ 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+$H_2$(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 dopant(Nd) reduced the amount released while penta-valent dopant(Nb) increased. With increase of amount of dopant, the tests of Nd, Ce and Nb showed higher diffusivity behavior. But samples of highest Nd and Ce were lower than others due to large grain. The tests of Nb were shown in good trend with independence of grain size. Fig.3 shows diffusion coefficients of all samples and shows a significant difference between Nd and Nb data. Generally, the tri-valence effect reduces xenon mobility but Nd increased the xenon mobility in some extent of content. Ce(+4) also showed higher mobility. The case of Nb showed a highest diffusivity with narrow difference even though large grain size. Pure $UO_2$ with large grain showed low diffusivity. Grain size effect seems to be stronger than dopant effect.

## 4. Conclusions

In the case of uranium vacancy controlled diffusion, Nd(+3) dopants reduced the cat-ion vacancy concentration, therefore xenon mobility was low.

This means the released fraction was lower than that of pure $UO_2$. 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.3. 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.

But a valence effect would be in some extent of concentration. Higher content of dopant seems to increase diffusivity regardless of valence effect.

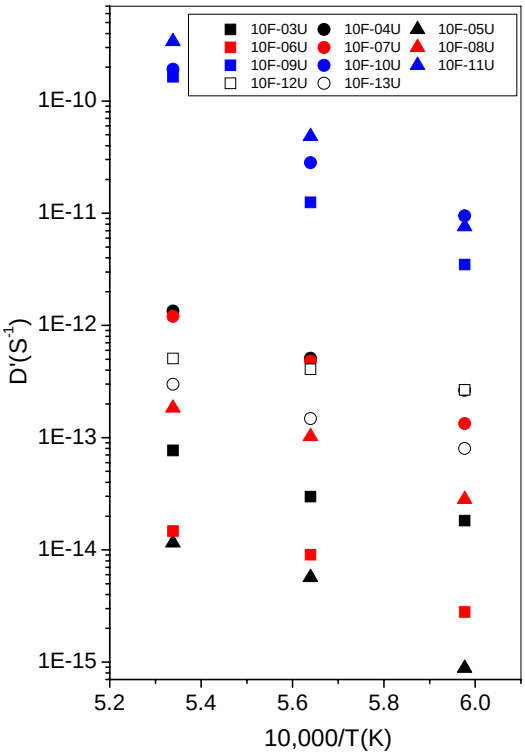


Fig. 3 Diffusion coefficients($D'=D/a^2$) in every sample.

## REFERENCES

[1] K.Une, I.Tanabe, M.Oguma,'Effects of Additives and The Oxygen Potential on The Fission Gas Diffusion in $UO_2$ Fuel.', J.Nucl.Mater.**150**, 93p.(1987)

[2] S.Kashibe, K.Une, 'Effect of additives ($Cr_2O_3$, $Al_2O_3$, $SiO_2$, MgO) on diffusional release of Xe-133 from $UO_2$ fuels.', J.Nucl.Mater **254,** 234p.-242p.(1998)

[3] Hj.Matzke, 'Diffusion in Doped $UO_2$.', Nuclear applications. **2**, 131p.(1966)