The Development of Xenon Diffusivity Measurement for Irradiated Ceramic Fuels with Low Burn-up

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Abstract. The fission gas diffusion coefficient is an important factor to study fission gas release, which is a common occurrence for fuel performance. Especially, Xe-133 is a good tracer and recommended in a post-irradiation annealing test. Annealing equipments for fuel was setup in IMEF in KAERI and several tests for various oxide fuels have been carried out. To obtain the atomic diffusivity of Xe-133, all fuel samples were irradiated with very low burn-up (< 0.1 $MW \cdot d \cdot t^{-1}$ U) to prohibit creation of fission bubbles. Therefore, these tests were not performed in a hotcell but a service area due to low radiation doses. The amount of production and release of Xe-133 were calculated by using ORIGEN-2 code. Based on the Booth theory, diffusion coefficients at each temperature were obtained. Diffusion coefficients of Xe-133 in all fuel samples were compared and analyzed by a published data. Additionally, we consider installation of equipments in a hotcell for a high burn-up fuel sample.

1. INTRODUCTION

Fission gas release occurs in nuclear fuel during reactor operation. Gaseous and volatile elements, fission products, are released from fuel pellets by temperature. It causes a fuel rod to have shorter life span due to higher internal pressure and pellet temperatures. Thus, the research of the fission gas release has been studied with diffusion mechanism. To setup a diffusion model, diffusion coefficient is important factor, which is obtained by experiments; one is an in-pile test and the other is a PIA(post-irradiation annealing) test. The former is effective in observing the apparent release behavior(diffusion and mechanical release), while the latter is the thermal diffusion. The thermal diffusion in fuel pellets is considered as lattice diffusion, surface diffusion and grain boundary diffusion. The lattice diffusion was controlled by several factors, e.g. oxidation or reduction state in high temperature, valence of additives and burn-up.

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In this study, lattice diffusion of Xe-133 ($T_{1/2}$ = 5.2 d, 81 keV) has been observed by PIA test with various ceramic fuel samples and conditions. To observe atomic diffusion in lattice, the Booth model was adopted, thus, a very low burn-up was decided to reduce traps which interrupt the atom mobility. Most samples were poly-crystal, accordingly grain boundary diffusion was considered.

2. EXPERIMENTAL

2.1. Sample preparations

Sixty samples were made, irradiated and annealed for the Xe-133 diffusion observation since 2000 as shown in Table 2.1. Most samples were made with 0.3 g and natural enrichment. Initially, UO_2 single grain powders were made and all following samples were poly-crystals. In Fig.2.1, the cubic and disk shapes were available to calculate the volume. A BET measurement was carried out in some samples but it was not reasonable. $(U,Th)O_2$ samples were made with 35% of ThO_2 and 65% of UO_2. SIMFUEL were made with natural elements based on the 27500 $MW \cdot d \cdot t^{-1}$ U as shown in Table 2.2. In 2008, Nitride fuel samples were made with disk type and high porosity to compare with oxide fuel. To observe the valence effect of additives, Nd_2O_3, CeO_2 and Nb_2O_5 were mixed with pure UO_2 in 2010. Moreover, two different grain sizes of UO_2 were made and, currently, two additional different grain sizes were added.

TABLE 2.1 CERAMIC SAMPLE SUMMARY FOR PIA TEST

Year	Samples	No.	Grain size [μm]	TD [%]	Enrichment
2000	UO_2 (powder)	1	23	N/A	Natural U
2001	UO_2 (powder)	4	23	N/A	
2002	[a]UO_2, [b]$(Th,U)O_2$ (powder, 3cubes)	13	[a]8.1±0.5 [b]7.5±0.5	95–97	
2003	SIMFUEL (3cubes)	1	10±2	95–97	
2004	SIMFUEL (3cubes)	12	10±2	95–97	
2006	SIMFUEL (3cubes)	2	10±2	95–97	
2007	SIMFUEL (3cubes)	4	10±2	95–97	
2008	UN (disk)	4	N/A	45.2	
	UO_2 (disk)	2	N/A	47.6	
2010	UO_2+additives (disk)	11	9–17	95–97	
2010	UO_2 (two grain size)	6	6, 13	95–97	

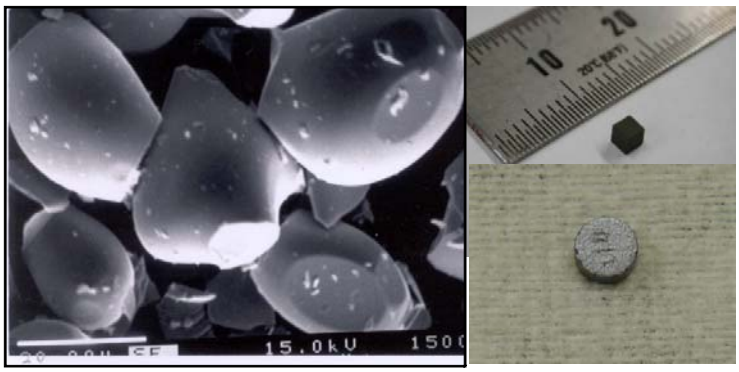

FIG. 2.1. Sample shapes (left: powder; right (top): cube; right (bottom): disk).

TABLE II. CONTENTS OF SIMFUEL

Contents	mg	Contents	mg
Rb	0.095	PdO	0.34
SrO	0.171	TeO_2	0.126
Y_2O_3	0.12	$BaCO_3$	0.57
ZrO_2	1.04	La_2O_3	0.42
MoO_3	1.055	CeO_2	1.99
RuO_2	0.825	Nd_2O_3	1.46
Rh_2O_3	0.11	UO_2	300
Total	308.35		

2.2. Irradiation

Those samples were contained a quarts tube at first, but was changed to a zry-4 tube for safety. The container was filled with helium of 1.2 bar and inserted in 3 irradiation capsules as shown in Fig. 2.2. The capsules were placed in the IP4(C) hole in the HANARO research reactor for irradiation. Twenty minutes of irradiation time was available for 24 MW power of the reactor, but the irradiation time was reduced to 16 minutes for 30 MW of operation power, which was equivalent to same burn-up. After irradiation and cooling time (7 days), the capsule was dismantled and contained in an alumina crucible at IMEF (Irradiated Materials Examination Facility).

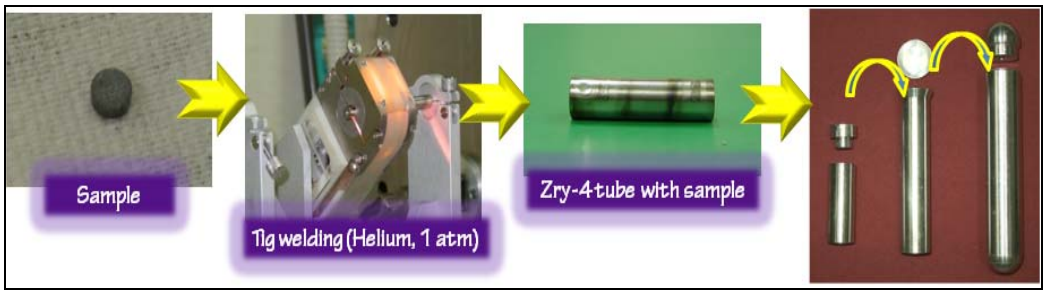

FIG. 2.2. Sample container (welding) for irradiation.

2.3. PIA system and procedures

For PIA test, the system was designed as shown in Fig. 2.3. It consisted of a furnace, filtration and gamma detection. The furnace was an electric resistance type(super kanthal) and withstand heat up to 1600°C. The chamber and internal structures were made of pure alumina. A B-type thermocouple was installed to measure the sample temperature. Oxygen sensor, ZrO_2 tube, measured the oxygen partial pressure in chamber. Helium gas was flowed with 100 $ml \cdot min^{-1}$ as carrier.

The filtration was designed as a cryogenic trap system to catch the gaseous xenon by solidification. The filter media was charcoal with a glass casing. It was placed into liquid nitrogen. The helium, as carrier, remained gas during filtration. The gamma detector was a semi-conductor type with high pure germanium crystal (HPGe). It was activated to obtain gamma-rays of Xe-133 released from the sample.

The gamma scan for 3600 sec was carried out for the sample in crucible to obtain the radioactivity of Xe-133 generated before the annealing test. After the sample was loaded into the furnace chamber, the temperature was controlled incrementally at 1400°C, 1500°C and 1600°C for UO_2, $(Th,U)O_2$ and SIMFUEL at 1200°C, 1300°C and 1400°C for UN. The annealing time for each temperature was decided according to the amount of xenon release. Generally, a disk sample with 95% TD would be kept for 15 hours, 9 hours and 6 hours, respectively, but a shorter annealing time was applied in the

case of oxidation atmosphere or low TD of sample. After annealing, a gamma scan was performed again before disposal.

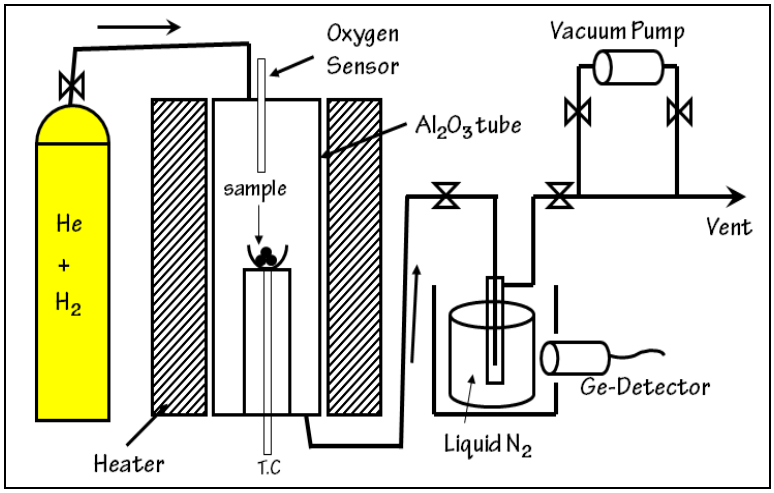

FIG. 2.3. Diagram of PIA system.

3. RESULTS

Fractional release was obtained by means of Ba-133 of reference source which emit 81 keV of gamma ray equal to Xe-133. Plot of f^2 vs. t is valuable to obtain slope at each temperature range. The slope is compared to $(36D) \times (\pi a^2)^{-1}$. Therefore, if '*a*' value is decided, diffusion coefficient is obtained. ThO_2 and fission products effects show retardation of release as shown in Fig. 3.1 -370 $kJ \cdot mol^{-1}$. of the oxygen potential is shown as 'x' value of UO_{2+x} was 0.0005 by means of oxygen sensor. Fig. 3.2 showed

Xe-133 release in uranium nitride and grain size effect of UO_2. A number of fractional release data were generated with various samples based on the same procedure.

Based on the plot of f^2 vs. t, the slope of each temperature range was calculated by a linear fitting. Finally, Diffusion coefficients were obtained by means of slope. Fig. 3.3 shows diffusion coefficients of Xe-133 in the single grained and the poly-crystal UO_2 with different oxygen potential, and they were compared to SIMFUEL. Diffusion coefficients of all samples were higher in increased oxygen potential and ThO_2 showed a retardation of Xe-133 release. SIMFUEL showed a tri-valence effect due to lower diffusion coefficients.

Conversely, uranium nitride (UN) was showed in Fig. 3.4 as similar diffusion coefficients with UO_2 even though the structure was different, and a tri-, quad- and penta-valence effects show a difference of diffusivity. However, large grained samples were lower in diffusivity in the same valence samples even though higher contents (right graph on Fig. 3.4).

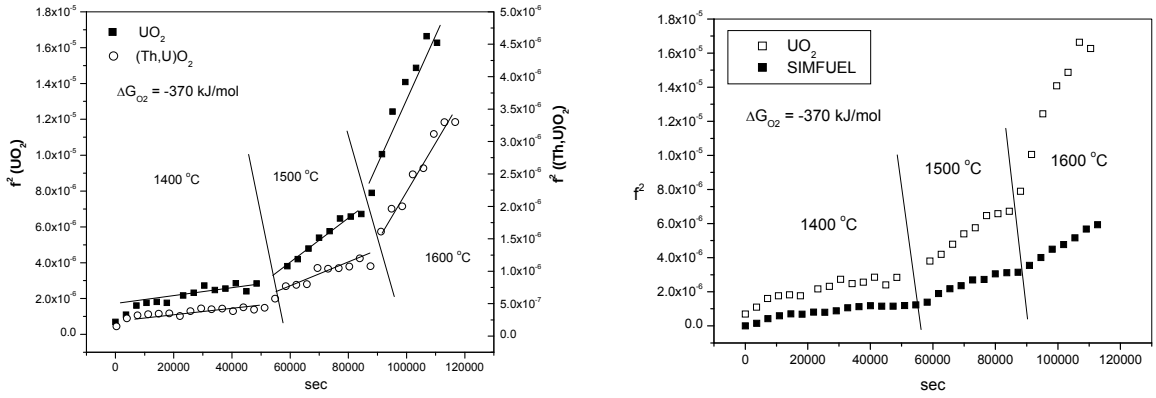

FIG. 3.1. Fractional release of UO_2, $(Th,U)O_2$ and SIMFUEL.

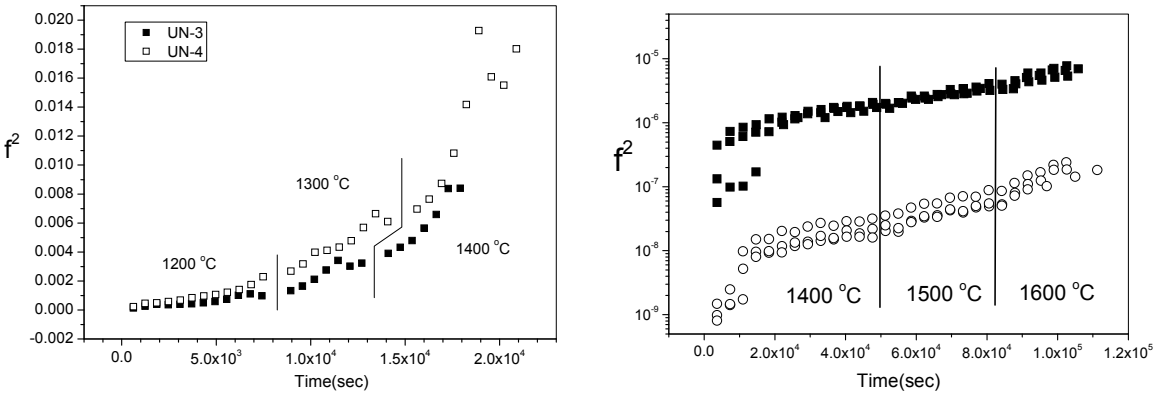

FIG. 3.2. Fractional release of UN and grain size effect of UO_2.

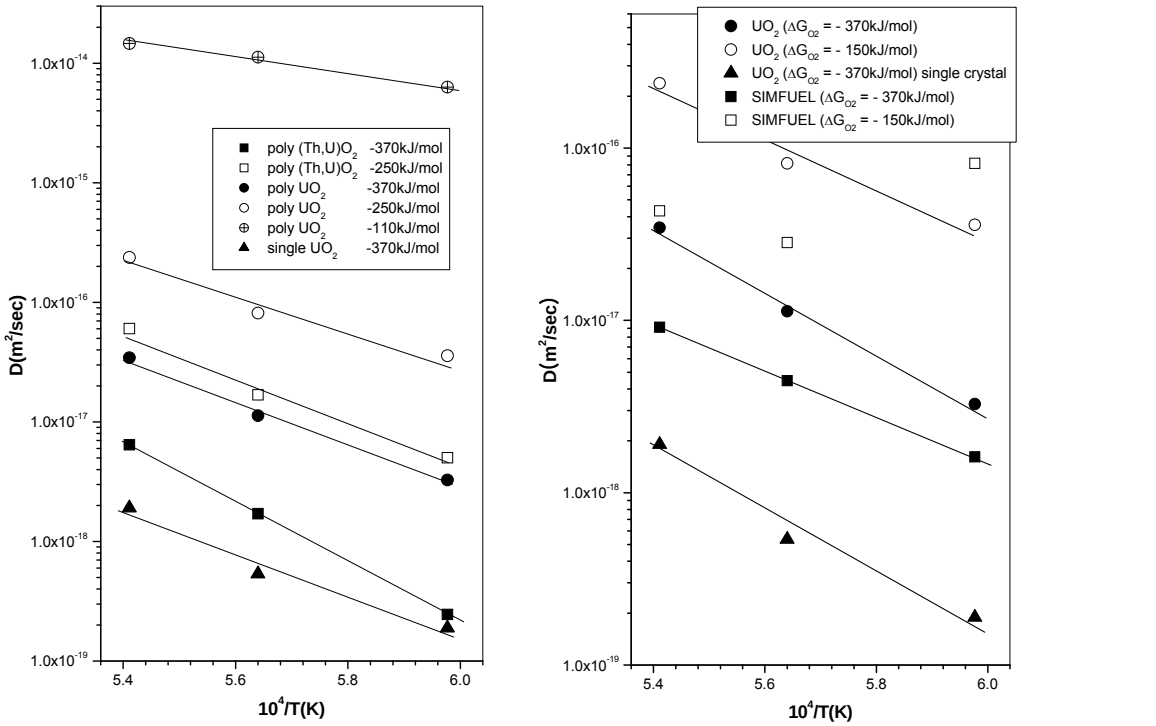

FIG. 3.3. Diffusion coefficients of UO_2(single and poly crystal) and $(Th,U)O_2$, and those of UO_2 were compared to SIMFUEL.

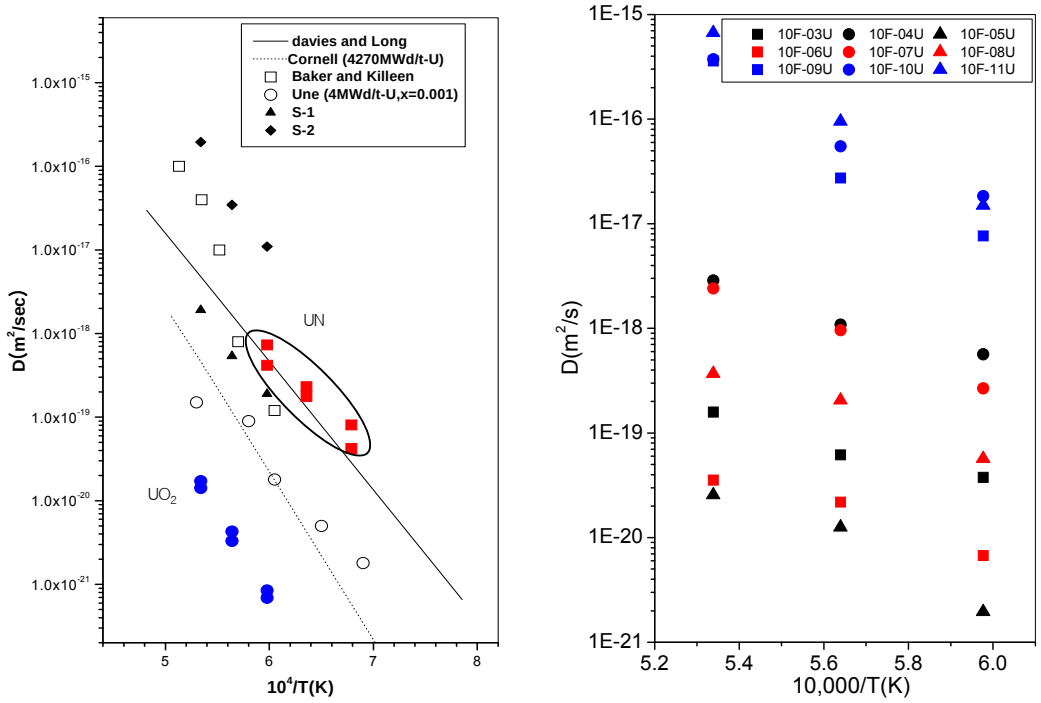

FIG. 3.4. Diffusion coefficients of UN with compared to UO_2 data and diffusion effects of doped UO_2 with different valence (10F-03U–10F-05U : Nd_2O_3; 10F-06U–10F-08U : CeO_2; 10F-09U–10F-11U : Nb_2O_5).

4. DISCUSSION

To measure diffusion coefficients of noble gases or volatile isotopes in fuel, a PIA and a in-pile test were used with each advantage [1]. To observe diffusion mechanism, a PIA test is useful with long lived tracer such as Xe-133.

PIA tests were performed based on the Booth theory [2] in this study as shown in Eq. 1:

$$f^2 = (36D) \times (\pi a^2)^{-1} \times t \quad (1)$$

Where
f is fractional release;
D is diffusion coefficient [$m^2 \cdot s^{-1}$];
a is equivalent sphere radius [m];
t is annealing time [s].

Eq. (1) is available in the case of f<0.3, and $(36D) \times (\pi a^2)^{-1}$ is almost linear. Fractional release was obtained by Xe-133 radio activities of production and release using a standard source (Ba-133) and ORIGEN-2 code [3]. Based on the Ref. [4], the linear fitting on plots in Figs 3.1–3.2 were needed to obtain slope which is equal to that of Eq. (1). If 'a' value was measured, finally, D can be obtained.

Diffusion coefficient of Xe-133 is controlled by temperature, oxidation condition, fission products and burn-up. Lattice diffusion with no traps is carried out with low burn-up to satisfy Booth theory, which was referred in Ref. [5]. Thus, 0.1MW·d·t^{-1} U of burn-up was decided in this study. Matzke suggested xenon mobility via tri-vacancy cluster [6]. Some calculations showed xenon moved via uranium vacancy in hyper-stoichiometric UO_2. Both of them are related to uranium vacancy concentration. This concentration is controlled by an oxidation condition and valence effect of fission products.

Generally, fission gas release is higher in an oxidized sample , e,g, UO_{2+x} , and a higher valence of elements such as niobium (+5) which is one of fission products [7–8]. Even though larger grains such as 10F-09U–10F-11U in Fig. 3.4, Nb_2O_5 enhanced the xenon release. The bonding energy of ThO_2 is stronger than UO_2, so it seems to disturb the xenon by causing it to jump to another site [9]. Therefore, diffusivity is lower than that of pure UO_2.

The xenon diffuses via tri-vacancy cluster in UO_2 based on ref. [4, 6]. However, it diffuses via uranium vacancy in non-stoichiometric UO_2. Thus, the uranium vacancy concentration is an important factor. Higher oxygen potential and valence elements are higher in uranium vacancy concentration, so it enhances the release of xenon.

On the other hand, uranium nitride (UN) has been studied as the fuel of a fast reactor. With the same condition of PIA test for UO_2, the results of UN were similar to UO_2 even though the structure was different. Mobility mechanism has not been found so far.

5. CONCLUSIONS

PIA tests for ceramic fuels were carried out by IMEF (Irradiated Materials Examination Facility) at KAERI (Korea Atomic Energy Research Institute). Sixty samples were used to obtain the diffusion coefficient of Xe-133 with various conditions. Lattice diffusion of xenon satisfying Booth theory have been studied since 2000. UO_2, $(Th,U)O_2$, SIMFUEL, doped UO_2, UN were used as a disk or cube type. Analyses of diffusion behaviors were performed and the data was accumulated. Even these data were useful to calculate fission gas release in fuel performance calculations. Diffusion coefficients of xenon in UO_2 were controlled by the oxygen potential, the valence of doped elements and the burn-up as well as the annealing temperature. Xenon diffusion data in UN were similar to those in UO_2 to some extent of temperature despite the different structure. It is needed to be studied to observe the diffusion mechanism. Recently, U-Zr metallic fuels were prepared as PIA sample. To observe a high burn-up effect, a PIA system is planned to be installed in a hotcell.

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