Quantitative Analysis of Kr-85 Fission Gas Release from Dry Process for the Treatment of Spent PWR Fuel

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1. Introduction

As spent UO_2 fuel oxidizes to U_3O_8 by air oxidation, a corresponding volume expansion separate grains, releasing the grain-boundary inventory of fission gases. Fission products in spent UO_2 fuel can be distributed in three major regions : the inventory in fuel-sheath gap, the inventory on grain boundaries and the inventory in UO_2 matrix. Release characteristic of fission gases depends on its distribution amount in three regions as well as spent fuel burn-up. Oxidation experiments of spent fuel at 500°C gives the information of fission gases inventory in spent fuel, and further annealing experiments at higher temperature produces matrix inventory of fission gases on segregated grain. In previous study, fractional release characteristics of Kr-85 during OREOX (Oxidation and REduction of Oxide fuel) treatment as principal key process for recycling spent PWR fuel via DUPIC cycle have already evaluated as a function of fuel burn-up with 27.3, 35 and 65 MWd/tU[1-3]. In this paper, new release experiment results of Kr-85 using spent fuel with burn-up of 58 GWd/tU are included to evaluate the fission gas release behavior. As a point of summary in fission gases release behavior, the quantitative analysis of Kr-85 release characteristics from various spent fuels with different burn-up during voloxidation and OREOX process were reviewed.

2. Methodology of Fission Gas Release Experiment

The spent PWR fuel rods with different burn-up were used in this experiment. Table 1 shows the spent fuel characteristics in this study. G23 series ranging from 27 to 35 GWd/tU were discharged from Gori #1 in 1986, and K23 series of high burn-up spent fuel from Uljin #2 in 2001.

Table 1. Spent fuel characteristics

Rod No.	G23 Assembly		K23 Assembly	
	5B	K10A	B16	M03
Initial Enrichment	3.21%		4.2%	
Burn-up (GWd/tU)	27.3	35	58	65
Decay time (yrs) at experiment	16	18	5	3
NPP	Gori # 1		Uljin # 2	

Table 2 represents initial inventory of Kr-85 fission gas in various spent fuels calculated by ORIGEN code. Measurement system of Kr-85 released from each process was well described in Reference [4]. Spent fuel is oxidized at 500 °C for 5 hrs in first, and U_3O_8 powder is treated by the OREOX process which is composed of 3 cycles of an oxidation at 450 °C in air and a reduction at 700 °C in 4% H_2/Ar. Release fraction of Kr-85 was obtained by comparing the cumulative activities during the specified experiment interval with an initial inventory shown in Table 2.

Table 2. Initial inventory of Kr-85 in spent fuel

Rod No.	G23 Assembly		K23 Assembly	
	5B	K10A	B16	M03
Activity (Ci/Kg-HM)	2.94	3.55	125	142
Mass (g/MTHM)	7.5	10.5	31.9	36.1

3. Release Percent of Kr-85 from spent fuel

3.1 Estimation of Grain Boundaries Inventory of Kr-85 from Release Percent during Voloxidation step

Typical release kinetics of Kr-85 with time as a function of spent fuel burn-up during voloxidation at 500 °C are represented in Fig. 1. Quantitative data of total Kr-85 release percent in voloxidation process are summarized in Table 3.

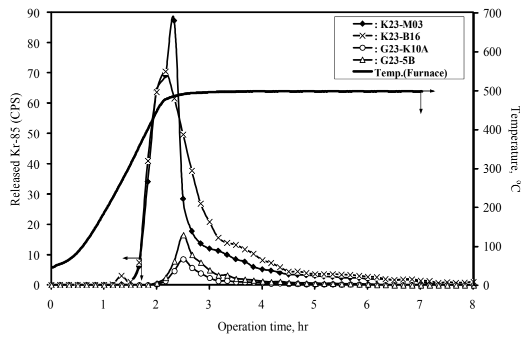

Figure 1. Release pattern of Kr-85 fission gas with spent fuel burn-up during voloxidation.

Table 3. Total release percent of Kr-85 in voloxidation step

Rod No.	G23 Assembly		K23 Assembly	
	5B	K10A	B16	M03
Release %	7.7	6.6	12.7	17.6

It was re-confirmed from new release experiment that as the spent fuel burn-up increases, Kr-85 release percent becomes higher. Quantitative release percent data of Kr-85 during voloxidation would be closely connected to grain boundary inventory of fission gases. It is well established that spent UO_2 fuel is oxidized from grain boundary attack of oxygen, and exposed grain begin to swell by formation of U_3O_8. Conversion of U_3O_8 and fracture of grain matrix produces a smaller size powder than original grain size in spent fuel [5]. Therefore, all fission gases distributed in grain boundaries are released during voloxidation step. Based on the total release percent shown in Table 3, grain boundaries inventory of Kr-85 on low burn-up of spent fuel would be about 7 %, and high burn-up fuel ranging from 58 to 65 GWd/tU represents 13 % ~ 18 %. It would be also interpreted that interconnection of fission gas bubbles along with grain boundaries on high burn-up spent fuel becomes more pronounced.

3.2 Release Percent of Kr-85 during OREOX step

Variation of Kr-85 activity in gas stream released from OREOX process in terms of spent fuel burn-up is shown in Fig. 2. Total release percent of Kr-85 with spent fuel burn-up in OREOX step is summarized in Table 4. Those data are based on the sum of each release percent obtained on specified time interval, and cumulative release percent with operation time is shown in Fig. 3.

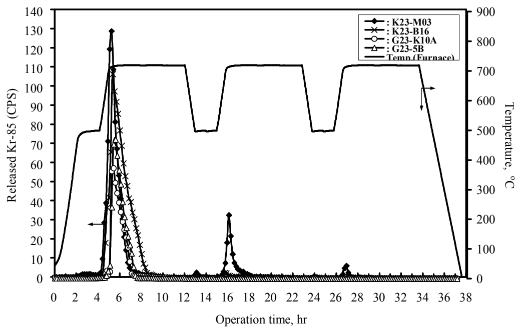

Figure 2. Release pattern of Kr-85 fission gas with spent fuel burn-up during OREOX process.

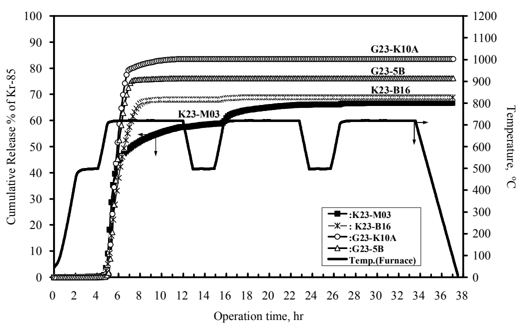

Figure 3. Cumulative release percent of Kr-85 fission gas with operation time in terms of spent fuel burn-up during OREOX process.

Table 4. Total release percent of Kr-85 in OREOX step

Rod No.	G23 Assembly		K23 Assembly	
	5B	K10A	B16	M03
Release %	79.2	83.5	68.8	66.7

Additional release experiment using K23-B16 spent fuel revealed that high burn-up spent fuel has similar distribution characteristic of Kr-85 fission gas on matrix, when compared to result of K23-M03. No release of Kr-85 during oxidation step in OREOX process was observed, and high release fraction was obtained in 1st reduction step. Because OREOX treatment of oxidized U_3O_8 powder makes a smaller particle size than initial powder, high release fraction of Kr-85 during OREOX step would be contributed to grain fracture of powder as well as increase of thermal diffusion with phase change of U_3O_8 to UO_2 at reduction step [5].

4. Conclusion

Additional release experiment using high burn-up spent fuel of 58 GWd/tU was performed to confirm the fission gases release characteristics with fuel burn-up during voloxidation at 500^oC and OREOX process. From the Kr-85 release behavior during voloxidation treatment of spent fuel, grain boundaries inventory of Kr-85 on low burn-up of spent fuel is about 7 %, and high burn-up fuel over 58 GWd/tU represents about 15 %. Thermal diffusion of fission gas with phase change of U_3O_8 at reduction step would be main factor for obtaining high release fraction.

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