Initial Release of Nuclides from Spent PWR Fuels

S. S. Kim, K.S. Chun Y. B. Kim and J.W. Choi

Korea Atomic Energy Research Institute

150 Dukjin-dong, Yusung-gu Daejeon 305-353, Korea

nsskim@kaeri.re.kr

SUMMARY

The relationship between the leaching and gap inventory of spent fuel has been studied. When a specimen of J44H08 spent PWR fuel with 38 GWD/MTU has been leached in the synthetic granitic groundwater in Ar atmosphere, the released fraction of cesium was increased rapidly up to 0.7% at around 500 days and stayed below 0.8% until 3years. This 0.7% of cesium might be released from the gap in this fuel.

The measurement of gap inventory with C15I08 spent PWR fuel, having 35 GWD/MTU and 0.22% of fission gas release, was also determined near 0.6% for the cesium, which is a similar fraction of cesium released from the leaching experiment with J44H08 fuel. Its gap inventories of strontium and iodine were about 0.03 and less than 0.2% respectively. Respective fractions of cesium and strontium in grain boundary of C15I08 were 0.78, 0.09%.

Key Words : leaching, spent fuel, gap inventory, grain boundary inventory

INTRODUCTION

The release mechanism of nuclides from a spent fuel is divided into three parts; the gap between uranium pellet and clad, including the large cracks in uranium matrix, grain boundary in uranium matrix and dissolution of uranium matrix itself [1].

Among the above three parts, when the spent fuel contacts with groundwater at the disposal site, the soluble nuclides in parts of gap and grain boundary can be dissolved relatively fast, and those in uranium matrix can be released very slowly, depending on the oxidation rate of uranium.

The initial release rate of cesium and iodine from spent fuel depend on the liner power rating (LPR) during the operation of a power plant. Though the dissolution of soluble elements in the gap by water is fast for the spent fuel with high LPR, their gap inventories for the spent fuel with low LPR are obtained the fraction of xenon inventory released from the puncturing of a fuel rod by the time of 90 days leaching [2~3].

For the spent PWR fuels, Johson et al. [4] insisted that the gap inventory of cesium is similar to released fraction of fission gas within the fuels with low discharge of fission gases. However, the direct relationship between fission and cesium inventory is difficult to be defined because the gap inventories of cesium are very different among spent fuels with similar release fraction of fission gases [5].

The gap and grain boundary inventories of cesium, strontium and iodine measured from a spent fuel and compared the fraction of cesium released from a leaching experiment with another spent fuel with similar burn-up in this study.

EXPERIMENTAL

1. Leaching experiment

A disc-shaped specimen of spent PWR fuel was prepared from a spent fuel rod by cutting with a diamond blade. The burn-up of the specimen was presumed to be 38 GWD/MTU for J44-H08 discharged from Kori-2 Nuclear Power Plant, and their initial enrichment of U-235 had been 3.52wt.%. After weighing of it, the specimen was put into 450 ml of synthetic granitic groundwater (Table 1) as leachant in a stainless steel 316L bath. The leachant was purged with Ar for over 15 minutes in order to remove oxygen, and the bath was kept in an Ar-filled box at a little higher pressure than that in the hot cell at the ambient hot cell temperature.

Table 1. Composition of synthetic granitic groundwater

Element	K	Sr	Mg	Li	Ca	Zn	Mn
Concentration (mg/L)	0.75	0.19	0.58	0.09	10.5	0.08	0.01
Element	HCO_3	Cl	SO_4	CO_3	Na	F	Fe
Concentration (mg/L)	74.9	18.9	25.3	9.9	50.8	7.69	0.04

2. Measurement of gap and grain boundary inventory

For the measurement of gap inventory of C15I08 with 35 GWD/MTU of burn-up and 3.20 wt.% of initial enrichment, a specimen was prepared by the same method for the leach test. After weighing it. its clad was separated from uranium matrix by pressing. Both of clad and uranium matrix were put in a bottle with 50 ml of distilled water for a certain period. About 5 ml of the solution from the bottle was collected periodically up to 149 days and filtered with

0.2㎛ filter to remove the uranium particles for the measure of gap inventory.

When the experiment of gap inventory was finished, the specimen was dried and powdered for the measurement of grain boundary inventory after removing a clad. All of the powder was put into 50 ml of 0.1M HCl solution and kept about 20 minutes. Then, about 5 ml of the solution was sampled by filtering with 0.2㎛ filter and the rest of solution was discarded. Fresh 50 ml of 0.1M HCl was added subsequently into the bottle with powder and sampling was performed by the previous method.

3. Analysis

The concentration of uranium and cesium in sample solution was analyzed by using inductively coupled plasma - mass spectrometer(ICP-MS) and γ-spectrometer, respectively. The activity of strontium was measured using liquid scintillation counter(LSC) after removing of cesium with ammonium molybdophosphate and separation of strontium with SR-resin [6], and the concentration of iodine was analyzed by neutron activation method after separation of iodine with a Stroes-Gascoyne's mehod [7].

RESULTS AND DISCUSSION

Total inventories of nuclides were calculated with ORIGEN II code. From the leaching experiment with J44-H08 by synthetic groundwater, the cumulative release fraction of cesium with leach time is shown Fig. 1. The released fraction of cesium is increased rapidly up to about 0.7% by the leach time of around 500 days and stayed below 0.8% until 3 years.

The concentration of uranium in 954-days leachate was $2x10^{-6}$ M (pH= ~8.0), which is the

similar value of $1.4x10^{-6}$ M from the calculation with EQ3/6 in the condition of near-field groundwater with pH=8.6 and Eh=-200mV [8]. However, this corresponds to 1.2 mg/m^2-day and is higher than 0.5 mg/m^2-day of CEA's result [9] obtained in reducing condition for the release rate of uranium

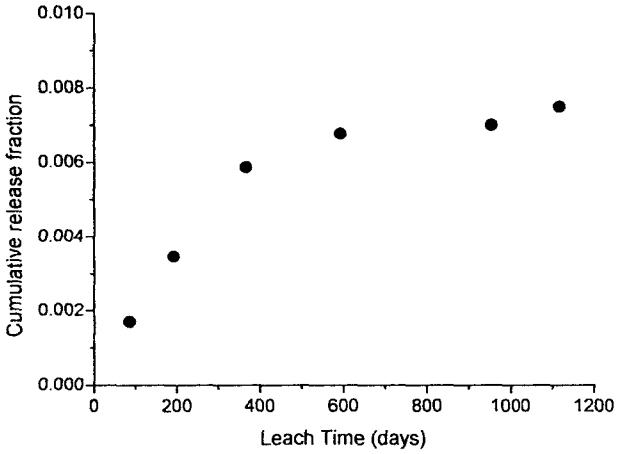

Fig. 1. Cumulated fraction of cesium released from J44-H08 spent fuel by synthetic granitic groundwater in Ar atmosphere.

From the experiment for the measurement of gap inventory with C15I08 by distilled water, the release fraction of cesium with leach time is shown Fig. 2. This graph also indicates that cumulative fraction of cesium is increased rapidly up to about 0.6% and thereafter increased slowly. The gap inventory of cesium of this spent fuel is assumed 0.6% that is larger than release fraction of fission gases unusually.

The comparison of Fig, 1 and Fig. 2 suggests that the released cesium in leaching experiment is released from the gap of J44-H08.

The gap inventory of strontium is determined 0.03%. However, because the concentration

of iodine in solution was too low to detect using γ-spectrometer and LSC, less than 0.2% was determined by neutron activation method. Fractions of cesium and strontium in grain boundary of C15I08 released into 0.1M HCl were 0.78 and 0.09%, respectively.

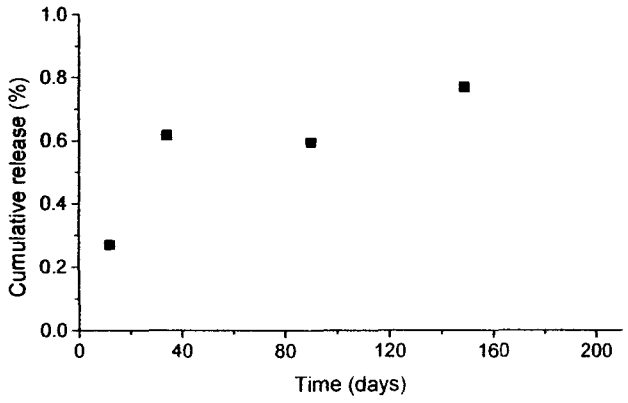

Fig. 2. Cumulated fraction of cesium released from C15I08 spent fuel by distilled water.

CONCLUSION

Compare to measurement of gap inventory with similar spent fuel, the amount of cesium released from a spent PWR fuel for 2~3 years in Ar atmosphere is corresponded to the gap inventory. The concentration of uranium in a leachate was similar to the value obtained from EQ3/6 code in near-field condition of repository site. The gap and grain boundary inventories of some elements in C15I08 were measured and found a gap inventory of cesium larger than released fraction of fission gases unusually.

REFERENCES

1. W.J. Gray and D.M. Strachan, Mat. Res. Soc. Symp. Proc., **212**, p.205 (1991).
2. R.S. Forsyth, J. Nucl. Mat., **190**, p.3 (1992).
3. S. Stroes-Gascoyne et al., Nucl. Tech., **77**, p.320, (1987).
4. L.H. Johson and J.C. Tait, "Release of segregated nuclides from spent fuel", SKB-TR-97-18, (1997).
5. W.J. Gray and C.N. Wilson, "Spent fuel dissolution studies (FY 1991 to 1994)", PNL-10540, (1995).
6. S.S. Kim, K.S. Chun and C.H. Kang, Analytical Science & Technology, **15(1)**, 1-6, (2002).
7. S. Stroes-Gascoyne, et al., Mat. Res. Soc. Symp. Proc., **353**, 625-631, (1995).
8. Kaija Ollial and Lasse Ahonen, "Solubility of uranium for TILA-99", POSIVA 98-13, p 22, (1988).
9. C. Poinssot, Proceeding of spent fuel workshop'2002, held Avignon in France, 23-25, (2002).