Proceeding of the Korean Radioactive Waste Society
Vol. 2(1), June 2004

Long-term Dissolution Behavior of Cesium from Spent PWR Fuel in Contact with Compacted Bentonite under Synthetic Granitic Groundwater

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Abstract

The amount of cesium released from the leaching of spent fuels in contact with and without the compacted bentonite block, which was compacted as the density of 1.4g/cm^3, up to 5.7 years were measured and the empirical formula of the fractional release rate of cesium were derived from these measured values. The empirical formulas show that the long-term release rate of cesium under a repository would become a constant, as about $3x10^{-6}$ fraction/day, after a certain period. The cumulative fractions of cesium released from the spent fuel with bentonite and with copper and stainless steel sheets were steadily increased, but the fraction from bare fuel was rapidly increased and then sluggishly increased. However, the remained value except its gap inventory from the cumulative fraction of cesium released from bare fuel was almost very close to the others. This suggests that the initial release of cesium from bare fuel might be dependant on its gap inventory.

1. Introduction

The R&D for the disposal of spent fuels accumulated up to our NPPs' lifetime at the Korea Atomic Energy Research Institute (KAERI) has been performed since the early 1997 in order to develop a reference repository system in Korea up to the year 2006. This program is in the early stage of the second year R&D in the third phase (2003-2006). The results up to the present recommend that the repository system would be located in the plutonic rock and consisted of vertical emplacement boreholes in separate disposal panels for spent CANDU and PWR fuels, corrosion resistant containers, which their PWR and CANDU spent fuels are encapsulated into, and domestic Ca-bentonite buffer layers[1].

Many experimental results have been reported for the dissolution behavior of spent nuclear fuel and unirradiated UO_2 in water under various conditions [2-7]. Most of the studies on the dissolution of unirradiated UO_2 pellet with bentonite have been carried out and only a few experiments have been tried for the identification of dissolution mechanism of spent nuclear fuel in a repository condition [8-10], which is not well known with the lack of real data. In

KAERI, the long-term dissolution experiment of spent PWR fuel has been carried out under the compacted bentonite and synthetic granitic groundwater since June of 1998. The purpose of this experiment is to get the information on corrosion behavior of spent fuel and release rate of radionuclides from the fuel within our domestic bentonite and synthetic granitic groundwater in order to support the development of radionuclide release model from spent fuel in the near field and the identification of the release mechanism, and finally to support the performance assessment of our reference disposal concept. The results obtained up to the present are described in this paper.

2. Experimental

2.1. Preparation of specimens

The rod of spent PWR fuel cut to about 3mm thickness with a diamond blade. The burn-ups of the specimens were presumed to be 39,089 and 37,805 MWD/MTU for J44-H08 and J-44-A03 discharged from Kori-2 Nuclear Power Plant, respectively, and their initial enrichment of U-235 had been 3.4869wt.%. Then all the specimens were weighted and photographed by an optical microscope.

2.2. Leach test

Domestic Ca-bentonite, which was compacted to 1.4g/㎤, was filled into the under-part of a leach cell (43mmΦ x 33mm) except space to hold a specimen, and one specimen was put in the space. Then, the under-part was bolted with the upper-part in which only the compacted bentonite was filled. The leach cell was put into 450ml of synthetic granitic groundwater (Table 1) as leachant in a bath (135mmΦ x 180mm). The stainless steel 316L was used for the fabrication of the leach cells and the baths. And stainless steel filters with the pore size of 10μm were put at the top and bottom sides of the leach cell in order to prevent the release of the compacted bentonite into the leachant by its swelling. The leachant was purged with Ar for over 15 minutes in order to remove oxygen, and all baths were kept in an Ar-filled box at a little higher pressure than that in the hot cell. For the comparison of the effect of bentonite on the leaching of spent fuel, some specimens without bentonite were put into baths. And two bare spent fuel fragments were leaded into water baths with and without metal sheets. All the loaded specimens have been emplaced in a hot cell for their leaching test at ambient hot cell temperature (see Fig. 1).

Table 1. Composition of synthetic granitic ground water

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

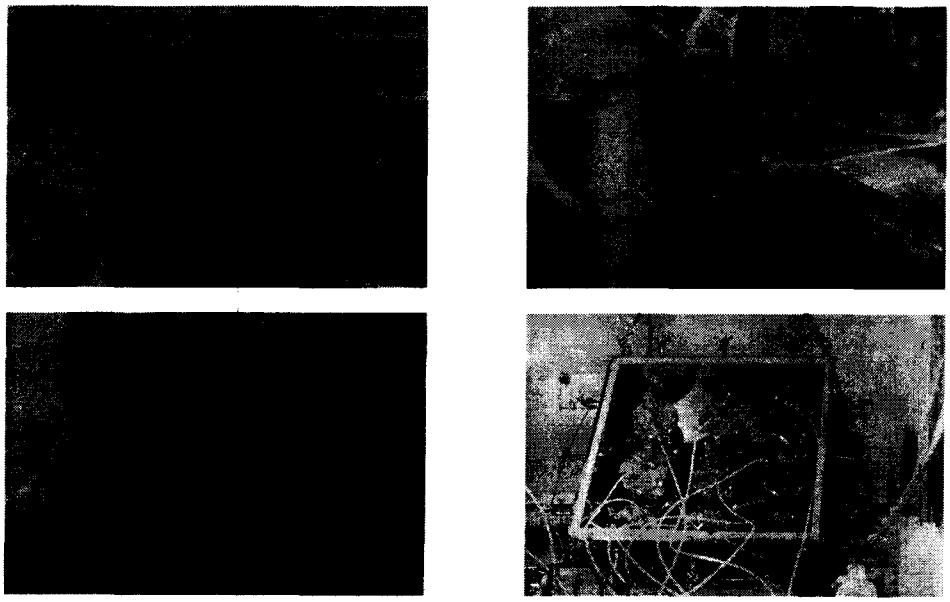

Fig. 1. Photograph of leach cells and water baths in the hot cell

2.3. Analysis

About 10ml of the leachates from each bath was sampled after a certain period of leaching and the activities of these leachates were measured by using γ-spectrometer (EG&G ORTEC ADCAM-100). The copper and stainless steel sheets from the leach cell for the specimen were taken out, settled into the 5M HNO_3 solution for one day, and then the activities of the solution has been measured. Some cells with the bentonite blocks were dismantled after a certain period of their leaching and then the bentonite blocks in contact with their specimens were sliced at a certain thickness as around 7 pieces. And the sliced bentonite layers were dissolved and the activities of cesium in the solutions were measured.

3. Results and discussion

The fractional release rates of cesium from spent fuel specimens up to 2092days are illustrated in Fig. 2. This figure shows that the release rates of cesium from the specimens are rapidly decreased up to a certain periods and then become a constant. This shows very similar

tendency to Forsyth's result[11]. In the presence of bentonite or metal sheets the constant rate would reach at around 400 to 500 days later, while the time when the rate from the bare spent fuel become a constant would require far more longer period than these.

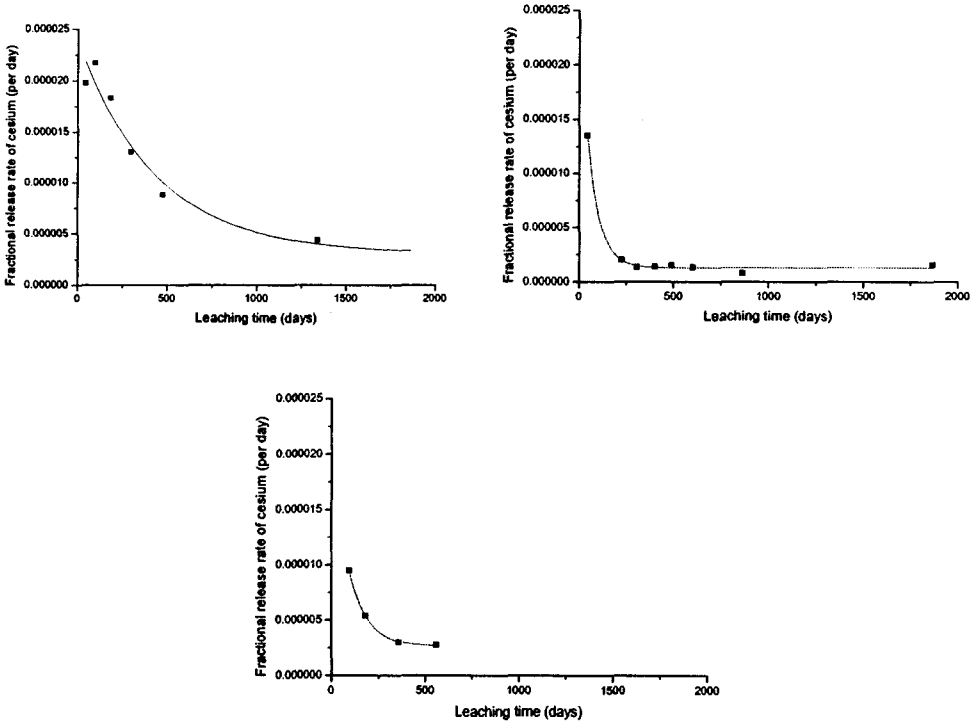

Figure 2. Fractional release rates of cesium as a function of leaching time

From the measured values the empirical formulas of the fractional release rates of cesium are derived as the following equations :

For bare fuel : $Ro = 3.05x10^{-6} + 2.08x10^{-5}\ Exp[-2.29x10^{-3} * T]$ .....(1) and

For only metals : $Rm = 1.29x10^{-6} + 2.38x10^{-5}\ Exp[-1.55x10^{-2} * T]$ .....(2) and

For bentonite : $Rb = 2.67x10^{-6} + 1.85x10^{-5}\ Exp[-1.05x10^{-2} * T]$ .....(3),

where Rs are the fractional release rates, as fraction per day, and T is the leaching time, as days. These equations indicate that the long-term release rate from the bare fuel specimen would be around $3.05x10^{-6}$ fraction/day, which is in the same range of Forsyth's results[11]. This rate is over two times higher than that without bentonite and with metal sheets, but a little higher than that from the specimen in contact with bentonite block. These lower rates may be due to the effects of waste package material(s) and bentonite material on the dissolution rate of cesium. This suggests that the long-term release rate of cesium under a repository condition would be not more than that from the spent fuel in contact with bentonite block.

The cumulative fractions of cesium released from the specimens up to around 1600 days

are shown in Figure 3. This figure shows that the fraction of cesium released from the bare fuel specimen is rapidly increased up to around 400days and then sluggishly increased, while the fractions from the others are steadily increased and these fractions are very close each other. The fraction from the bare fuel up to about 400days is about $7x10^{-3}$, which is very close to the gap inventory of cesium ($6x10^{-3}$)[12], and up to about 1560days is about $9.7x10^{-3}$, which is higher than the results of Finn et al.[13,14] but lower than that of Forsyth[15]. The cumulative fraction of cesium released from the bare fuel except its gap inventory is almost the same as the fractions from the others, $2.6x10^{-3}$ up to 1640days for only metal sheets and $3.1x10^{-3}$ up to 1118 days for bentonite. This suggests that the initial release of cesium from bare spent fuel would be dependant on its gap inventory, but the amount of the initial release could be reduced due to the presence of bentonite or waste package materials. This may be due to the formation of alteration products that could incorporate cesium[16].

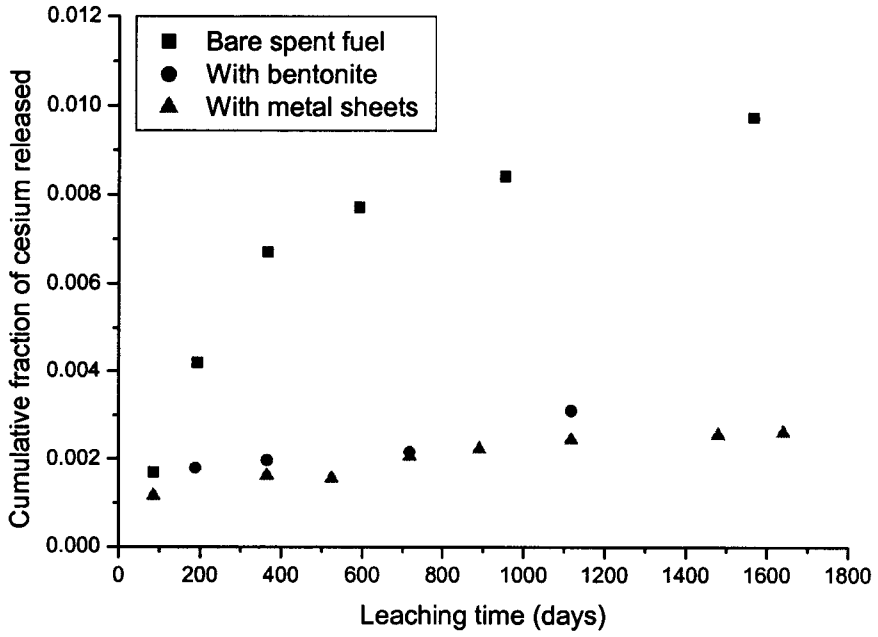

Fig. 3. Cumulative fractions of cesium released by leaching up to about 1600days

The specific activity of cesium contaminated on copper sheets was approx. $0.41\mu Ci/cm^2$, which was about ten times higher than that on stainless steel sheets. This means that copper could be more favorable material for the retardation of cesium than stainless steel. On the other hand, the total fraction of the cesium contaminated on the copper and stainless steel sheets was about $3x10^{-5}$, which was about two order lower than the amount of cesium in the leachate. This suggests that some other effect(s) by waste package material(s) could be affected on the dissolution of cesium from spent fuel.

The distribution of cesium in the bentonite block is illustrated in Figure 4. This shows that most of cesium released from the specimen is accumulated on the bentonite close to the specimen and the sorption band is getting wide with the increase of leaching time.

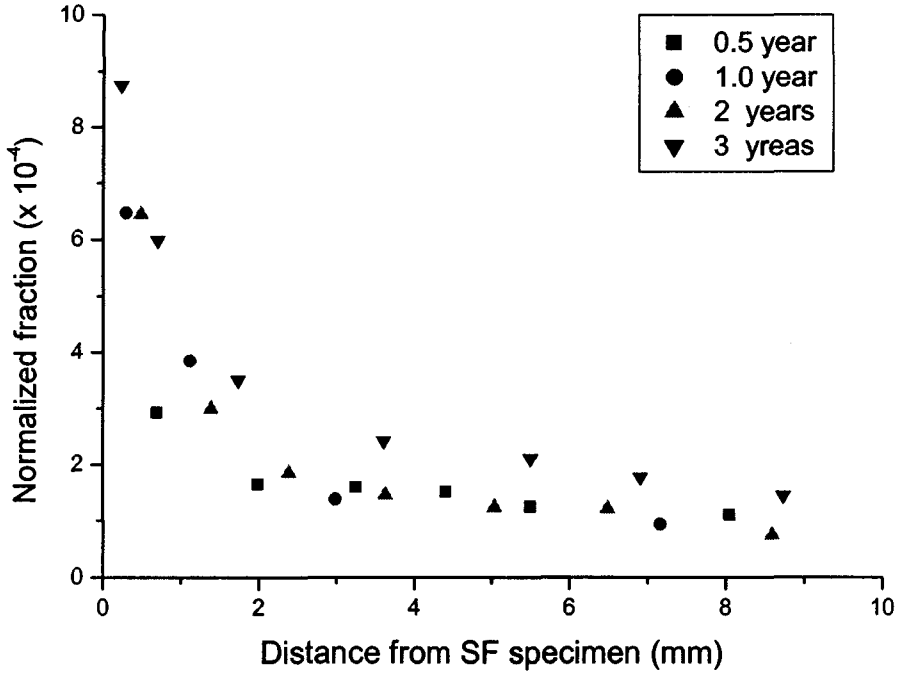

Figure 4. Distribution of cesium in bentonite block

To identify the effects of bentonite and waste package materials on the dissolution of cesium from spent fuel, the further research should be required.

4. Conclusions

To identify the long-term leaching behavior of spent PWR fuel under a repository, the spent fuel exposed in their leachant up to 5.7 years and these leachates were collected, and their cesium activities were analyzed.

The empirical formulas of the fractional release rates of cesium, which are derived from the current measured values, indicate that the long-term release rate of cesium from spent fuel under a repository condition would become a constant as about $3x10^{-6}$ fraction/day.

The experimental results suggest that the cumulative fraction of cesium released from spent fuel could be reduced by the presence of bentonite and/or waste package materials. However, the further research should be required for the identification of the incorporation of cesium into an alteration product of spent fuel under the presence of bentonite and/or metals.

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