Durability test on irradiated rock-like oxide fuels

K. Kuramoto *, N. Nitani, T. Yamashita

Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan

Abstract

For a profitable use of Pu, Japan Atomic Energy Research Institute has been promoting researches for once-through type fuels. The strategy consists of stable rock-like oxide fuel fabrication in conventional fuel facilities followed by almost complete Pu burning in LWR and disposal of chemically stable spent fuel without further processing. Because leach rates of hazardous nuclides, such as TRU and β-emitters, that have long half-lives, are very important for the evaluation of geological safety, leaching tests in deionized water at 363 K were performed with reference to the MCC-1 method. Five irradiated fuel pellets, a single phase fuel of a yttria-stabilized zirconia (YSZ) containing UO_2 (U-YSZ), two fuels of U-YSZ particle dispersed in $MgAl_2O_4$ (SPI) or Al_2O_3 (COR) matrix, two homogeneous-blended fuels of U-YSZ and SPI or COR powders, were submitted to the tests. Stainless steel containers with Au coating and ethylene propylene diene monomer were used as leaching vessels and packing, respectively. The evaluated normalized leach rates of Zr, U and Pu were obviously lower than those of the other important elements and nuclides. Americium, Np and especially Y showed unexpectedly high evaluated normalized leach rates. The volatile elements, Cs and I, showed enhanced leaching within particle-dispersed type fuels because of crack formation around the particle.
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1. Introduction

For a profitable use of Pu, Japan Atomic Energy Research Institute (JAERI) has been promoting researches for once-through type fuels. The concept consists of stable rock-like oxide (ROX) fuel fabrication in conventional fuel facilities followed by almost complete Pu burning in LWR safely, and disposal of chemically stable spent fuels without further processing. In our recent work we showed that ROX could be easily fabricated [1], that it would transmute Pu twice as effective than MOX in the case of reactor-grade Pu in LWR [2] and that it could be irradiated in LWR with comparable safety to UO_2 fuels [3–5]. The ROX concept can be completed successfully by showing good geological stability of spent ROX fuels.

The important radiotoxic nuclides are TRU, for example Np (^{237}Np, $\tau_{1/2}$: 2×10^6 a), Am (^{243}Am, $\tau_{1/2}$: 7×10^3 a) and Cm (^{245}Cm, $\tau_{1/2}$: 9×10^3 a and ^{246}Cm, $\tau_{1/2}$: 5×10^3 a), as well as β-emitters, such as ^{135}Cs ($\tau_{1/2}$: 3×10^6 a), ^{99}Tc ($\tau_{1/2}$: 2×10^5 a), ^{79}Se ($\tau_{1/2}$: 7×10^5 a), ^{129}I ($\tau_{1/2}$: 2×10^7 a), ^{81}Kr ($\tau_{1/2}$: 2×10^5 a), etc. Strontium-90, one of the significant nuclides for low level radioactive waste, can be ignored because of its short half-life compared to the geological time scale of order of one million years.

In the present study, leach tests were performed in a deionized water at 363 K for up to 173 days with reference to the MCC-1 method [6]. Nuclides leached from the irradiated ROX fuel may exist as ions and/or colloidal species in the leachate and as precipitates on a vessel and a sample holder. The leach rate of the important nuclides and elements must be estimated by the total amount existing in leachate and precipitates. In addition, because the colloidal species will influence the solution equilibrium and migration of the nuclides and the elements, the amount of colloids is estimated quantitatively. In the present paper, we described a detailed

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* Corresponding author. Tel.: +81 29 282 6129; fax: +81 29 282 6678.

E-mail address: kura1@nucef.tokai.jaeri.go.jp (K. Kuramoto).

procedure for the leaching experiment on the irradiated ROX fuels and showed the first leaching data for spent inert matrix fuel.

2. Experimental procedure

Five kinds of irradiated fuels, of which composition and irradiation conditions are listed in Table 1, were prepared using 19.57 wt% of enriched U instead of Pu and were irradiated in the Japan Research Reactor No. 3 for about 100 days at nominal power of 20 MW. An irradiated ROX pin was sliced to six specimens of 2 mm-thick disks including the stainless steel cladding using the diamond saw. The geometrical surface area of each specimen is about 0.45 cm^2.

Vessels used in the tests were made of stainless steel and their inside surfaces were coated with 20 μm-thick Au. The packing was made of ethylene propylene diene monomer, a kind of ethylene–propylene rubber. As specimen holders, vinyl chloride frames and Pt wires were used. These materials are well known to be highly resistant against irradiation damage.

Leaching tests were performed at the No. 4 hot cell of Waste Safety Testing Facility in JAERI. After polishing with #600 emery paper on each surface, specimens were rinsed with fresh water. Leachate was 50 ml deionized water and the leaching temperature was controlled at 363 K. Five leaching periods were selected as 8, 24, 60, 115 and 173 days. One specimen was submitted to each leaching period of 8, 24, 60 and 115 days, and two specimens to a leaching period of 173 days. Vessel weight including specimen, leachate and sample holder was measured every 14 days to check the leachate leakage. Weight loss of every leachate was less than 2% even after 173 days, which is within the MCC-1 regulation.

After the given leaching period, vessels were taken from the oven and pH measurements were made. The leachate was passed through a 0.20 μm-filter to remove small amounts of fuel fragments. The filtered leachate was transferred to a glove box and was distributed evenly into two bottles. A designed amount of nitric acid was added to one bottle to adjust its nitric acid concentration of leachate to 0.1 mol l^{-1} (Leachate A: LA). The other half part of the leachate was passed through a 1.8 nm-ultrafilter, and then the designed amount of nitric acid was also added to the leachate (Leachate B: LB). In order to quantify the precipitation on the vessel and sample holder, 0.1 mol l^{-1} of nitric acid was added to a used vessel containing a used holder, heated up at 363 K for 1 week and the solution was passed through a 0.20 μm-filter (Leachate C: LC). The total leaching amounts of the relevant elements and nuclides were obtained by concentration measurements in LA and LC. Colloid parts of them were obtained quantitatively by comparing the concentration in LA and LB. Additionally, a distribution factor between the precipitate and the solution could be estimated by comparison of concentrations in LA and LC.

Table 1
Sample composition and irradiation conditions of irradiated ROX fuels

	YSZ	YSZ + SPI (S-type)		YSZ + COR (C-type)	
	Z Single phase fuel	SH Homogeneously blended type fuel	SD Particle-dispersed type fuel	CH Homogeneously blended type fuel	CD Particle-dispersed type fuel
Sample composition in mol%					
YSZ[a]	81.75	11.90	11.90	11.93	11.93
UO$_2$	18.25	19.71	19.71	19.76	19.76
MgO	–	45.60	45.60	–	–
AlO$_{1.5}$	–	22.79	22.79	68.31	68.31
Sample irradiation conditions					
Linear power in kW m^{-1} (average/maximum)					
	13.9/15.2	23.4/26.0	23.0/25.4	20.7/22.7	24.9/27.4
Temperature at surface of pellet in K (average/maximum)					
	990/1030	1440/1510	1250/1310	1290/1350	1300/1360
Temperature at center of pellet in K (average/maximum)					
	1490/1580	1940/2080	1740/1850	1730/1830	1820/1930
Burn up in ^{235}U %/MWd cm^{-3}					
	21.01/0.059	24.15/0.103	23.28/0.100	20.87/0.088	23.89/0.105

[a] YSZ; 78.57 mol% ZrO$_2$ + 21.43 mol% YO$_{1.5}$.

Because Cs activity in every leachate was too high to handle and to analyze the leachate in this test, Cs must be removed from every leachate. Among the absorbents ammonium phosphomolybdate, $(NH_4)_3PO_4 \cdot 12MoO_3 \cdot nH_2O$ (AMP) [7,8], was found to be very effective to remove Cs from the leachate. The concentration of Cs in the leachate before Cs separation was measured by a Li-drifted type Ge semiconductor detector (Ge-detector). To remove Cs from LA, LB and LC, 0.020 g of AMP was added to each leachate. The mixture was shaken and settled for 10 min, and shaken again. After 10 min settlement, the mixture was passed through a 0.20 μm-filter. More than 99.9% of Cs could be removed from the leachate by these operations.

After Cs separation by AMP, the concentration of matrix elements, TRU and the β-emitters with long half-lives except for Cs in LA, LB and LC were determined by ICP-MS. The mass numbers 24–27, 50–172, 192–198 and 204–260 were measured using a standard solution by calibration curve method. The normalized leach rate of element k (NL_k, in $g\,cm^{-2}\,day^{-1}$) is given by the following equation:

$$NL_k = \frac{(C_{k\text{-LA}} + C_{k\text{-LC}}) \cdot V}{SA \cdot F_k \cdot t}, \tag{1}$$

where $C_{k\text{-LA}}$ and $C_{k\text{-LC}}$ are concentration in $g\,ml^{-1}$ of element k in LA or LC, respectively, V is the leachate or solution volume in ml, SA is the initial surface area of specimen in cm^2, F_k is the mass fraction of element k in a specimens and t is the leaching period in day. The normalized leach amount of element k ($NLA_{k\text{-LA}}$, $NLA_{k\text{-LB}}$ and $NLA_{k\text{-LC}}$, in $g\,cm^{-2}$) in LA, LB or LC is presented as the following equations, respectively:

$$NLA_{k\text{-LA}} = \frac{C_{k\text{-LA}} \cdot V}{SA \cdot F_k}, \tag{2}$$

$$NLA_{k\text{-LB}} = \frac{C_{k\text{-LB}} \cdot V}{SA \cdot F_k}, \tag{3}$$

$$NLA_{k\text{-LC}} = \frac{C_{k\text{-LC}} \cdot V}{SA \cdot F_k}, \tag{4}$$

where $C_{k\text{-LB}}$ is concentration of element k in LB in $g\,ml^{-1}$. The normalized total leached amount of element k (NLA_k, in $g\,cm^{-2}$) is also revealed from the following equation:

$$NLA_k = \frac{(C_{k\text{-LA}} + C_{k\text{-LC}}) \cdot V}{SA \cdot F_k}. \tag{5}$$

The leach rate and leach amounts of element k can be comparable directly with those of the other elements by normalizing with the factor F_k as in Eqs. (1)–(5). Table 2 shows F_ks of the important elements and nuclides in five irradiated ROX fuels, calculated by the ORIGEN2 code [9]. Cross-contamination from AMP and equipment in the hot-cell and gloveboxes hindered a meaningful Mg and Sn detection by ICP-MS. Because only stable [127]I and [82]Se nuclides were detected by ICP-MS, leach rate and leach amounts of [129]I and [79]Se were obtained by using the ratios of mass fraction $F_{\text{I-129}}/F_{\text{I-127}} = 4.24$ and $F_{\text{Se-79}}/F_{\text{Se-82}} = 0.16$. Measurement of Kr was not carried out because Kr is an inert gas and may escape during the procedure.

Leach rate and leach amount of [135]Cs were estimated by γ-ray measurement of ^{137}Cs in the leachate before Cs separation and in the LA, LB and LC solutions using

Table 2
F_ks of the important elements and nuclides calculated by the ORIGEN2 code

	Z-type	S-type	C-type
Matrix elements			
Zr[a]	5.4×10^{-1}	1.2×10^{-1}	1.1×10^{-1}
Y[a]	1.3×10^{-1}	2.9×10^{-2}	2.8×10^{-2}
U	3.3×10^{-1}	5.2×10^{-1}	5.1×10^{-1}
Al	–	2.3×10^{-1}	3.4×10^{-1}
Mg	–	9.1×10^{-2}	–
FPs			
Pu/[239]Pu[b]	3.7×10^{-4}/94.8%	5.8×10^{-4}/94.8%	5.7×10^{-4}/94.8%
Np/[237]Np	9.3×10^{-6}/100%	1.5×10^{-5}/100%	1.4×10^{-5}/100%
Am/[241]Am	2.1×10^{-7}/99.8%	3.3×10^{-7}/99.8%	3.2×10^{-7}/99.8%
Cs/[135]Cs	3.8×10^{-4}/14.8%	6.0×10^{-4}/14.8%	5.9×10^{-4}/14.8%
Tc/[99]Tc	1.1×10^{-4}/100%	1.7×10^{-4}/100%	1.7×10^{-4}/100%
I/[129]I	2.1×10^{-5}/80.9%	3.3×10^{-5}/80.9%	3.3×10^{-5}/80.9%
Se/[79]Se	1.1×10^{-5}/9.8%	1.8×10^{-5}/9.8%	1.7×10^{-5}/9.8%
Others	3.6×10^{-3}	5.8×10^{-3}	5.7×10^{-3}

[a] F_{Zr} and F_Y also contain fission products Zr and Y.
[b] /[239]Pu means ratio of [239]Pu to total Pu in %.

Li-drifted type Ge semiconductor detector. The counting time was 3600 s. Normalized leach rate and normalized leach amount of ^{135}Cs are given by the following equations, respectively:

$$NL_{Cs-135} = \frac{A_{Cs-137-LA} + A_{Cs-37-LC}}{10 \cdot \lambda} \cdot \frac{137}{N_{AV}} \cdot \frac{V}{SA \cdot F_{Cs-135} \cdot t}, \quad (6)$$

$$NLA_{Cs-135} = \frac{A_{Cs-137-LA} + A_{Cs-137-LC}}{10 \cdot \lambda} \cdot \frac{137}{N_{AV}} \cdot \frac{V}{SA \cdot F_{Cs-135}}, \quad (7)$$

where $A_{Cs-137-LA}$ and $A_{Cs-137-LC}$ are the radioactivities of ^{137}Cs in LA and LC for 10.0 ml, λ is decay constant of ^{137}Cs in s^{-1} and N_{AV} is Avogadro's constant.

3. Results and discussion

3.1. pH measurement

As a result of pH measurement, the leachate pH decreased rapidly from 5.9 to 4.0–4.3 in the early leaching period, and then pH of the leachates were increasing gradually with increasing leaching period. It was reported that leachate pH decreased suddenly by the water radiolysis in the high γ-irradiation field and as a result alkaline element leaching were also accelerated [10]. The pH increasing tendency is also well known in glass waste form leaching behavior because of dissolution of alkaline elements such as Na and Cs to leachate. The same phenomena occurred in this test, that is, rapid pH decrease by the water radiolysis and slow pH increase by harmonic Cs leaching.

3.2. Leaching of matrix elements

Fig. 1(a)–(c) shows NL_{Zr}, NL_Y and NL_U for five irradiated ROX fuels, and Fig. 1(d)–(f) indicates NLA_{Zr}, NLA_Y and NLA_U of the Z fuel. The NL_{Zr} decreased with increasing leaching time. The NL_{Zr} of the Z fuel was $\sim 3 \times 10^{-8}$ g cm^{-2} day^{-1} after 173 days, and those of the other fuels were 5–25 times higher. The NL_U of the Z fuel was also the lowest among the five fuels, however, NL_Y of the Z fuel was two or three orders of magnitude higher than those of the other fuels. This means that a part of Y did not dissolve with Zr and U congruently suggesting that a new phase of FP and a part of Y formed with low durability in water. In addition, NL_U of homogeneously blended (H-type) fuels were about one third of those of particle-dispersed (D-type) fuels.

The NL_Y and NL_U were also decreased with increasing leaching time as in the case of NL_{Zr}. These decreasing leach rates mean that rapid and large amount dissolution of these elements occurred in the early leaching period within the first 8 days, and then only a limiting amount of these elements had been leaching.

Therefore the measured NL_k was not the true normalized leaching rate in this case. The NLA_{Zr} of the Z fuel was increasing with increasing leaching time as shown in Fig. 1(d), and NLA_U and NLA_Y were also increasing slightly after 173 days. A similar tendency was observed for the other fuels. The required leach rate of element k can be obtained from the slope of NLA_k against leaching periods. The normalized leach rates of Zr, Y and U evaluated by the slope method were $\sim 2 \times 10^{-9}$, $\sim 5 \times 10^{-6}$ and $\sim 1 \times 10^{-9}$ g cm^{-2} day^{-1} (for the period from 8 to 173 days), respectively. Thus, it could be summarized that the leach rate order of matrix elements was as follows: Zr and U < Y.

As for the behavior of dissolved Zr, the precipitation fraction of Zr, NLA_{Zr}-LC, was very small, and the ratio of NLA_{Zr}-LA and NLA_{Zr}-LB were roughly 5:4. From these facts it can be concluded that 80% of leached Zr existed as ion species and the rest Zr formed colloidal species in the leachate. On the other hand, the dissolved U showed a quite different behavior from that of Zr. The normalized leach amounts of U in LA and LB were negligible, and NLA_U-LC was almost the same as the normalized total leaching amount of U, NLA_U, and this phenomenon became distinct with increasing leaching time. This means that dissolved U gradually formed precipitates on the vessel and the sample holder as leaching periods increased.

Fig. 2(a) and (b) shows NL_{Al} of four irradiated ROX fuels and leach amounts of Al for the CH fuel. The NL_{Al} gradually decreased with increasing leaching time, similar to the other matrix elements. The NLA_{Al} was almost constant, independent of leaching periods as well as fuel types. These findings clearly showed the evidence that leaching behavior of SPI and COR resembled each other in irradiated ROX fuels.

3.3. Leaching of TRU

Fig. 3(a) and (b) indicates NL_{Pu} of five irradiated ROX fuels, and NLA_{Np-237}, NLA_{Pu-239} and NLA_{Am-241} of the Z fuel. The leach rate difference caused by fuel type was not clear for TRU. As shown in Fig. 3(a), NL_{Pu} decreased with increasing leaching time due to rapid and excess fuel dissolution in the early leaching period. This tendency was also observed for Np and Am leaching. However, Fig. 3(b) obviously shows that not only NLA_{Np} (triangles) but also NLA_{Pu} (circles) and NLA_{Am} (squares) was increasing with increasing leaching time, as observed for those of matrix elements. Normalized leach rates of ^{237}Np, ^{239}Pu and ^{241}Am evaluated by the slope method for the Z fuel were 1×10^{-6}, 5×10^{-9} and 2×10^{-7} g cm^{-2} day^{-1} (8–173 days), respectively. The order of the leach amount in early leaching period and that of leach rate during 8–173 days are summarized as follows: ^{239}Pu < ^{237}Np < ^{241}Am and ^{239}Pu < ^{241}Am < ^{237}Np, respectively. As for the behavior of dissolved TRU, the
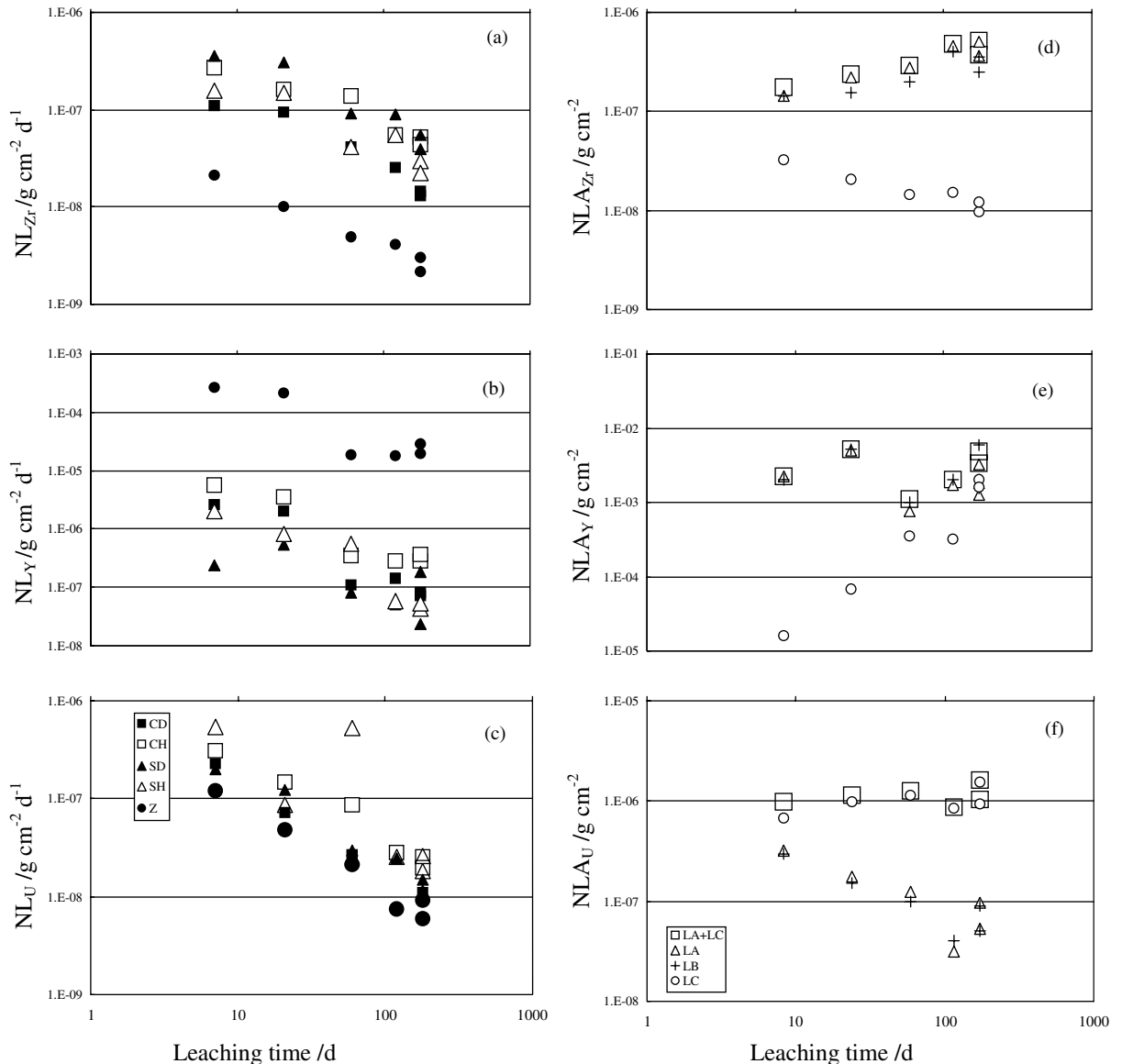
Fig. 1. Normalized leach rates (NL, in $g\,cm^{-2}\,day^{-1}$) and normalized leach amounts (NLA, in $g\,cm^{-2}$) of Zr, Y and U for the Z fuel: (a) NL_{Zr}, (b) NL_Y, (c) NL_U, (d) NLA_{Zr}, (e) NLA_Y, (f) NLA_U.

precipitation fraction of each TRU is almost the same as its total leach amounts as can be seen from Fig. 3(b) indicating that leached ions precipitated gradually on the vessel and the sample holder, and after 120 days almost dissolved TRU ions had precipitated. This is similar to the leaching behavior of U.

3.4. Leaching of β-emitters with long half-lives

The normalized leach amounts of ^{135}Cs, ^{99}Tc, ^{79}Se and ^{129}I in LA and LC for the Z fuel, and that of ^{79}Se for the SH fuel are shown in Fig. 4(a)–(d) and (f), respec-

tively. Additionally, that of Rh for the Z fuel is also illustrated as a reference in Fig. 4(e). Leaching behaviors of these nuclides are crucial and important for the safety assessment of the radioactive waste repository of ROX fuels.

The normalized leach amount of ^{135}Cs for the Z fuel is increasing slowly with increasing leaching time as shown in Fig. 4(a). The measured normalized leach rate of ^{135}Cs at 8 days was $\sim 2 \times 10^{-5}\ g\,cm^{-2}\,day^{-1}$ and that evaluated by the slope of leach amounts was $\sim 1 \times 10^{-6}$ $g\,cm^{-2}\,day^{-1}$ (8–173 days). The discrepancy of these rates also indicated large amount Cs dissolution in the
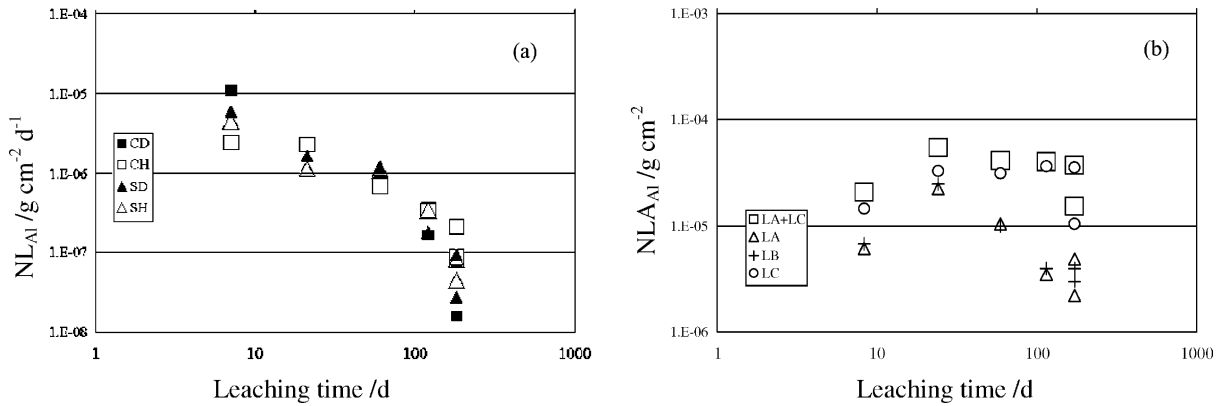
Fig. 2. Normalized leach rates (NL, in $g cm^{-2} day^{-1}$) and normalized leach amounts (NLA, in $g cm^{-2}$) of Al for the CH fuel: (a) NL_{Al}, (b) NLA_{Al}.
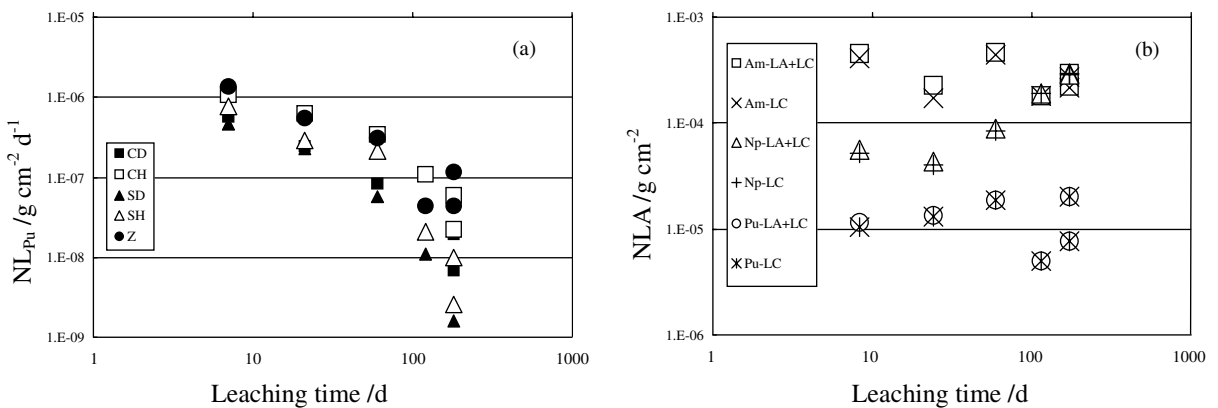

Fig. 3. Normalized leach rates (NL, in $g cm^{-2} day^{-1}$) of Pu-239 and normalized leach amounts (NLA, in $g cm^{-2}$) of ^{237}Np, ^{239}Pu and ^{241}Am for the Z fuel: (a) NL_{Pu-239}, (b) NLA_{Np-237}, NLA_{Pu-239} and NLA_{Am-241}.

early leaching period. The NL_{Cs-135} and evaluated normalized leach rate for H-type fuels were in the same order as those for the Z fuel, however, those for D-type fuels were roughly twice as high as those for the Z fuel. The post irradiation examination study for the irradiated ROX fuels showed that fission product gas release of D-type fuels were higher than those of the other fuels [4]. The microscopic cracks would form in the matrix around the particles, and the produced FP gas would escape through the cracks resulting in the high FP gas release rate of the D-type fuels. A similar mechanism can be applied to the different Cs leaching behavior between D-type and the other fuels: Cs volatilized, migrated and segregated in micro-cracks around the particles. This led to an increased opportunity to contact with the leachate and to the high leach rate of D-type fuels. Almost all of the dissolved ^{135}Cs exist as ion species through the leaching period, as clearly shown in Fig. 4(a).

The measured NL_{Tc-99} at 8 days and the evaluated leach rate for the Z fuel were $\sim 6 \times 10^{-6}$ and 6×10^{-8} (8

to 173 days) $g cm^{-2} day^{-1}$, respectively, and those for the other fuels were almost the same. A large dissolution of ^{99}Tc seemed to occur in the early leaching period. Dissolved ^{99}Tc in the leachate began to precipitate gradually and three quarters of ^{99}Tc had precipitated after 173 days. The leach rates and the leaching behavior of ^{99}Tc were similar to those of Rh, see Fig. 4(b) and (e). Therefore Tc-99 and Rh (possibly Ru and Pd) might form alloy phases and dissolve into the leachate congruently.

For ^{129}I leaching, the NL_{I-129} at 8 days and evaluated normalized leach rate for the Z fuel were $\sim 3 \times 10^{-7}$ and $\sim 6 \times 10^{-8}$ (8–173 days) $g cm^{-2} day^{-1}$, respectively. These values were similar to those of D-type fuels, but were 2–3 times as high as those of H-type fuels. This phenomenon could be explained by the same discussion as in the case of Cs leaching behavior: volatile I migrated to the cracks, and contact and dissolve to the leachate. As can be seen from Fig. 4(e), NLA_{I-129} in LA and LB leachates have the same values within the experimental errors
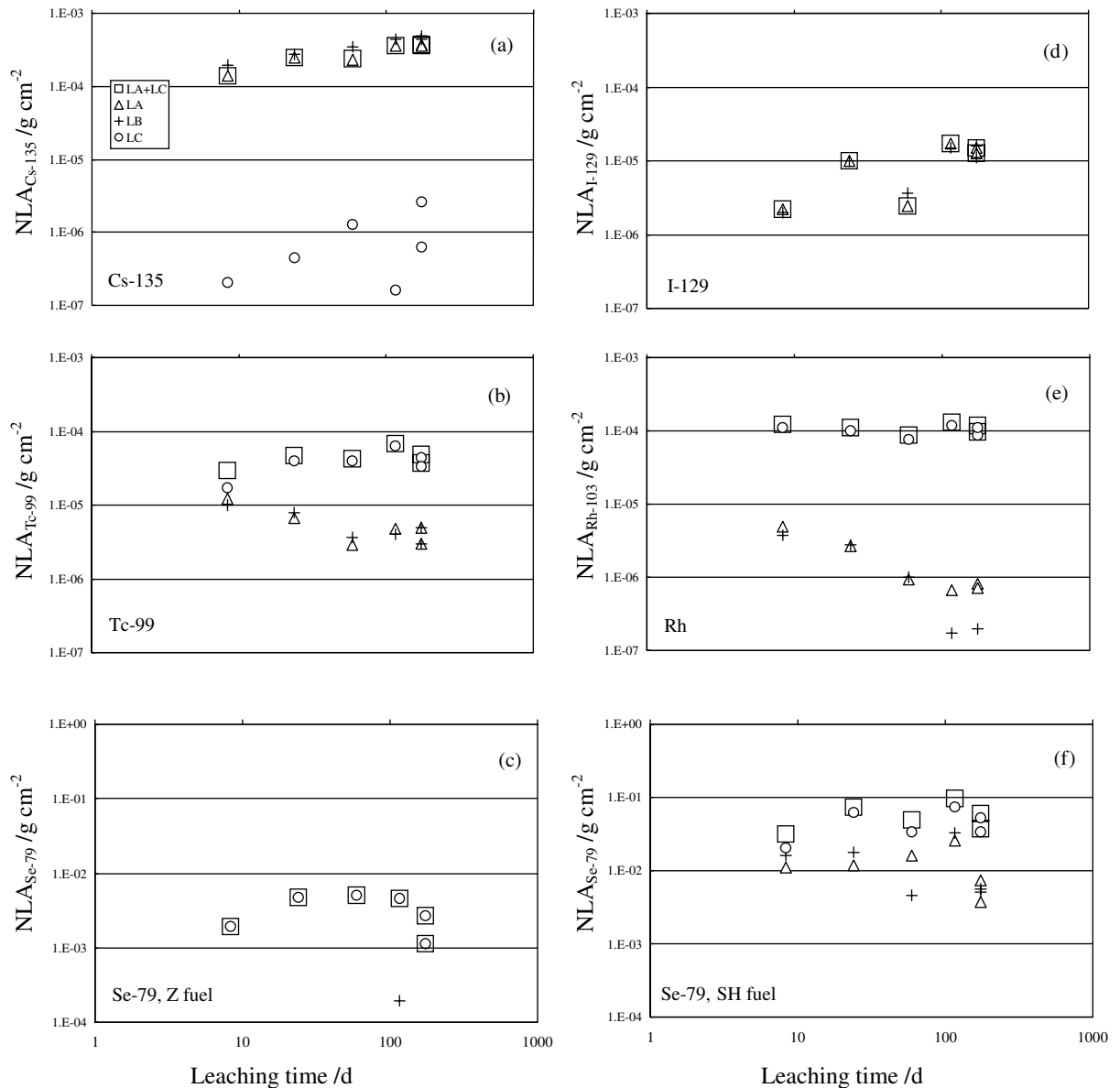
Fig. 4. Normalized leach amounts (NLA, in $g\,cm^{-2}$) of ^{135}Cs, ^{99}Tc, ^{129}I, ^{103}Rh and ^{79}Se for the Z fuel, and ^{79}Se for the SH fuel: (a) NLA_{Cs-135}, (b) NLA_{Tc-99}, (c) NLA_{Se-79} for the Z fuel, (d) NLA_{I-129}, (e) NLA_{Rh-103}, (f) NLA_{Se-79} for the SH fuel.

suggesting that almost all of dissolved I existed as ion species through the leaching period. The NLA_{I-129} and evaluated normalized leach rate of ^{129}I in irradiated ROX fuels were about two orders of magnitude lower than those of ^{135}Cs. According to the evaluation of FP chemical forms in irradiated ROX fuels, CsI was the major compound for I under excess Cs environment [11]. If CsI migrated to the surface of the leaching sample, CsI dissolved easily to the leachate, resulting in a high leaching rate of I. The fact that the normalized leach rate of I by the slope method was smaller slightly than those

of Zr and U means that CsI might be dissolved with the matrix elements meta-congruently.

The NL_{Se-79} at 8 days and evaluated normalize leach rate for the Z fuel were $\sim 2 \times 10^{-6}$ and $\sim 7 \times 10^{-6}$ (8–173 days) $g\,cm^{-2}\,day^{-1}$, respectively, and were one or two orders of magnitude lower than those for the other fuels. Almost all of the dissolved Se in leachate have precipitated in the case of the Z fuel as seen in Fig. 4(c). On the other hand, for the other fuels about two thirds of Se precipitated in the early leaching period and the precipitates fraction increased with increasing leaching

periods. Meta-stable soluble Se complexes with Al and/or Mg may form in leachates for S- and C-type fuels and the Se complexes may attribute to the different Se leaching behavior between the Z fuel and the other fuels. A detail reaction mechanism will be clarified when Se chemistry has been well established.

3.5. Comparison of evaluated normalized leach rates among the matrix elements, the TRU and the β-emitters

The order of evaluated normalized leach rates of the matrix elements, the TRU and the β-emitters was as follows:

$$Zr, U \text{ and } Pu < I \text{ and } Tc < Am < Cs \text{ and}$$
$$Np < Se \text{ and } Y$$

The first three elements, Zr, U and Pu, are well known as highly durable ones [12,13], and their durabilities are confirmed in this test. It was unexpected that Y showed a high solubility compared with the other important elements and nuclides. In addition, the evaluated normalized leach rates of Am and Np were two and three orders of magnitude higher than that of Pu, respectively, although chemical behavior of these elements were supposed to be similar to that of Pu. It was revealed in this test that the evaluated normalized leach rate of I, known as soluble element, was the same order to that of Tc, not of Cs.

4. Conclusions

Leach tests were performed in deionized water at 363 K for up to 173 days using five irradiated ROX fuels. The results are as follows:

(1) Normalized leach rates of the relevant elements and nuclides during 8–173 days were lower than those in early leaching period within the first 8 days because of rapid and large amount dissolution in this period.
(2) The evaluated normalized leach rates of Pu, Zr and U were obviously lower than those of the other relevant elements and nuclides.
(3) Americium and Np, and especially Y showed unexpectedly high normalized leach rates.
(4) Zirconium and soluble elements of Cs and I, existed in leachate as ion species. On the other hand, the other important elements and nuclides had precipi-

tated or had been precipitating in the leaching period.
(5) Volatile elements such as Cs and I showed large leach amount for D-type fuels due to the microcrack formation around the particle.

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