BOUNDARY DEPTH BETWEEN LLRW AND CLW AFFECTED BY FLUCTUATION OF ELEMENTAL COMPOSITION

Norikazu Kinoshita[1], Takuma Noto[1], Kazuaki Kosako[1], Motoyuki Asada[1],
Kohei Urabe[2], Akane Tada[2], Kazuyuki Torii[2]

[1] Institute of Technology, Shimizu Corporation, Etchujima Koto-ku, Tokyo, Japan

(norikazu.kinoshita@shimz.co.jp)

[2] Nuclear Projects Division, Shimizu Corporation, Kyobashi Chuo-ku, Tokyo, Japan

ABSTRACT

Activated materials discharged from decommissioning of nuclear power plants will be separated into low level radioactive waste (LLRW) and clearance level waste (CLW) based on their activity levels. Usually, the activity levels of the wastes are estimated with a computer simulation and measurement. The computer simulation requires elemental compositions of material, record of operation for a nuclear reactor, and detailed information of structure inside reactor and building. Concretes, main waste in the decommissioning, have a fluctuation in the elemental composition because of variation of distribution for aggregates. In the present work, we investigated the boundary depth between LLRW and CLW affected by the fluctuation of the elemental composition estimated from 200 g of samples. The fluctuations of the elemental compositions of Si, Ca, Fe, Co, Cs, and Eu were evaluated by analyzing 22 concrete samples with approximately 200 g each. The concentration of each element excepting Si was varied by 20 – 30% comparing to the average concentration of the samples. In addition, we examined estimation of distributions of radioactivities in a concrete using a monte-carlo simulation code PHITS. The 30% fluctuation of elemental composition gave the boundary depth between LLRW and CLW deeper by a few centimeters comparing with the depth estimated without the fluctuation.

1. INTRODUCTION

Decommissioning of decrepit nuclear power plants has been proceeded in the world. In Japan, shutdown of more than 10 nuclear power plants had decided so far. In the first stage of the decommissioning, activities inside reactor building are estimated using computer simulation and measurement. Activated concrete discharged from decommissioning of nuclear power plant is classified into low level radioactive waste (LLRW) and clearance level waste (CLW), based on the estimation of the activity level. Activities of ^{60}Co ($T_{1/2}$ = 5.27 yr), ^{134}Cs ($T_{1/2}$ = 2.06 yr), ^{152}Eu ($T_{1/2}$ = 13.33 yr), and ^{154}Eu ($T_{1/2}$ = 8.8 yr) produced by a neutron capture reaction on stable isotopes of Co, Cs, and Eu, are of dominant activities in the concrete, because of their large neutron capture cross section and half-life. However, concentrations of these elements are a ppm level in the concrete.

In Japan, Japan Power Demonstration Reactor (JPDR), a boiling water reactor operated for 17,110 hours with a thermal power of 45 – 90 MW, had completed demolishment of the reactor building and its relevant facilities although disposal of radioactive waste is being still under discussion. 3,770 tons of radioactive waste and 20,670 tons of non-radioactive waste were discharged (Shiraishi 1999). The radioactive wastes accounted for 1627 tons of metals etc. and 2,143 tons of radioactive concretes, which comprised of 74% for activated concretes and 26% for contaminated concretes. All of the concretes

discharged from core internals were treated as the radioactive waste. The ^{60}Co and $^{152,\ 154}$Eu were dominant activity even in the case of demolishment of JPDR. The amounts of the activity levels with > 4000 Bq/g, 40 – 4000 Bq/g, 0.4 – 40 Bq/g, and < 0.4 Bq/g accounted for 3%, 4%, 13%, and 80%, respectively.

The CLW can be recycled for concretes etc. according to the guideline (IAEA 2004). On the other hand, the LLRW is treated by shallow-ground trench disposal etc. based on its activity level. Therefore, disposal of the LLRW results in immense amount of cost.

Precise estimation of the activities results in precise separation of waste. The activities are usually estimated with a neutron flux distribution obtained using a transport calculation as shown in Eq. (1).

$$A = \int f(E)\sigma(E)N\left(1 - e^{-\lambda t}\right)dE \qquad (1)$$

Here, A is an activity produced just after termination of the neutron irradiation, $f(E)$ is the neutron flux with an energy E, $\sigma(E)$ is the neutron capture cross section, N is the number of target atom, λ is decay constant of product, t is the irradiation time. The N depends on precision and fluctuation in analysis. The $f(E)$ is affected by a model of neutron transport in a simulation. One the other hand, the elemental composition in a concrete is varied due to fluctuation of distribution of aggregate and fluctuation of distribution of elemental composition in aggregate. When large amount of concrete samples are subjected to analysis, the compositions should show values close to the averages of the samples. In contrast, when small amount of samples are subjected to analysis, the compositions would be fluctuated. The local variation of elemental composition would give the fluctuation for a boundary depth of radioactive waste between LLRW and CLW. Therefore, we will evaluate the depth safely taking the fluctuation into consideration.

We have addressed to reduce the amount of the LLRW discharged from decommissioning of nuclear power plants by precise separation. We have investigated an analytical procedure for elemental compositions in concretes for computer simulation of radioactivity distribution as well as validation of simulating radioactivities produced around nuclear reactor have. In the present work, elemental compositions in concrete samples were analyzed to evaluate local fluctuation of the composition. We show boundary depth for the separation of the LLRW and CLW affected by elemental composition in the concrete.

2. EXPERIMENTAL

Mixture of ingredients which consist of 336 kg/m^3 of cement, 178 kg/m^3 of water, 838 kg/m^3 of fine aggregate (< 5 mm for a diameter), 937 kg/m^3 of course aggregate (5 – 20 mm for a diameter), and 4.2 kg/m^3 of additive, were solidified in 5 molds with a diameter of 10 cm and height of 20 cm. The concrete samples are named as A, B, C, D, and E.

The concrete samples were sliced in an approximately 1-cm-thick. 4 – 5 samples were taken at regular intervals from each concrete. A weight of each sample was approximately 200 g. A total of 22 of the sliced samples were analyzed for evaluation of fluctuation of the elemental composition. A photograph of the samples is shown in Fig. 1. The sliced concretes were pulverized using a stamp mill and then the samples were homogenized well. In addition, 1 g was taken from each pulverized sample and then mixed for determination of the average concentrations.

Known amounts of the samples were heated in HNO$_3$+HClO$_4$. The residues fused with KOH were suspended in HCl. The Ca, Co, Fe, Cs, and Eu dissolved in the extractant of HNO$_3$+HClO$_4$ solution and the HCl solution were measured with an ICP-AES or ICP-MS after suspended materials were removed by filtration. Concentrations of Ca, Co, Fe, Cs, and Eu were determined from 2 measurement data. On the other hand, known amounts of the samples were fused with KOH again. The residues were suspended in HCl. After the solutions were dried up, the residues were again suspended in HCl. The residue after heated at 850 °C was weighed to determine the Si concentration. The Si dissolved in the HCl

solution was determined with an ICP-AES. The Si concentration was determined from the weight of residue and concentration of the solution.

Fig. 1 Photograph of the sliced concrete samples.

3. RESULTS AND DISCUSSION

3.1. FLUCTUATION OF ELEMENTAL COMPOSITION

Concentrations of Si, Ca, Fe, Co, Cs, and Eu in the samples sliced from concrete cores of A – E are shown in Fig. 2. The averages from the mixed samples were also given with a solid line in the figure. Some data have different concentration by dozens percent from the average concentration. For example, Co in C-1, Eu in A-1 and C-1 show larger concentration, Cs in C-2 shows smaller concentration. Specific feature among different concrete cores and position in the concrete is not observed.

Arithmetic means of the analyzed 22 samples were 24.4% for Si, 14.4% for Ca, 1.77% for Fe, 9.4 ppm for Co, 1.9 ppm for Cs, and 0.49 ppm for Eu. The concentrations in the sample, which 1 g of each sample was mixed for the average, were determined to be 24.4% for Si, 14.3% for Ca, 1.76% for Fe, 9.4 ppm for Co, 1.9 ppm for Cs, and 0.50 ppm for Eu. The samples used in the present work contain 14.7% of cement, 36.5% of fine aggregate, 40.9% of course aggregate and 7.8% of water as described in Sec. 2. Therefore, main component of the concretes depend on elemental composition of the aggregates.

Sometimes, the fluctuation of the elemental composition is observed when the same samples are analyzed by different institutes. For example, nominal values of elemental compositions in the Japanese geochemical standard references were determined by data analyzed by dozens of institutes by means of different technique (Imai 1995). The 3σ of standard deviations of JG-3, one of the geochemical standard references, which cover 99% of data, are 15% of the arithmetic mean for Si, 9.6% for Ca, 48.9% for Fe, 28.4% for Co, 53.1% for Cs, 30.4% for Eu. We should note these fluctuations by the different analytical process. In contrast, the arithmetic means of the composition correspond well with the mixed sample in the present work. The fluctuation of the composition in the present work should be derived from the fluctuation in the samples rather than the fluctuation of precision by the analytical procedure. Therefore, the fluctuation by the analytical procedure is neglected in this work.

Histograms of each concentration are shown in Fig. 3. All data in Si are shown within ±8.2% of average. The ranges are ±14.3% of the average for Ca, ±17.5% for Fe, ±32.9% for Co, ±20.0% for Cs, and ±17.1% for Eu. Si showed the smallest fluctuation in the concentration. The other elements showed approximately 20 – 30% of fluctuation to the average. When we analyze the elemental composition from approximately 200 g of sample, we should take into account at least 30% of the fluctuation.
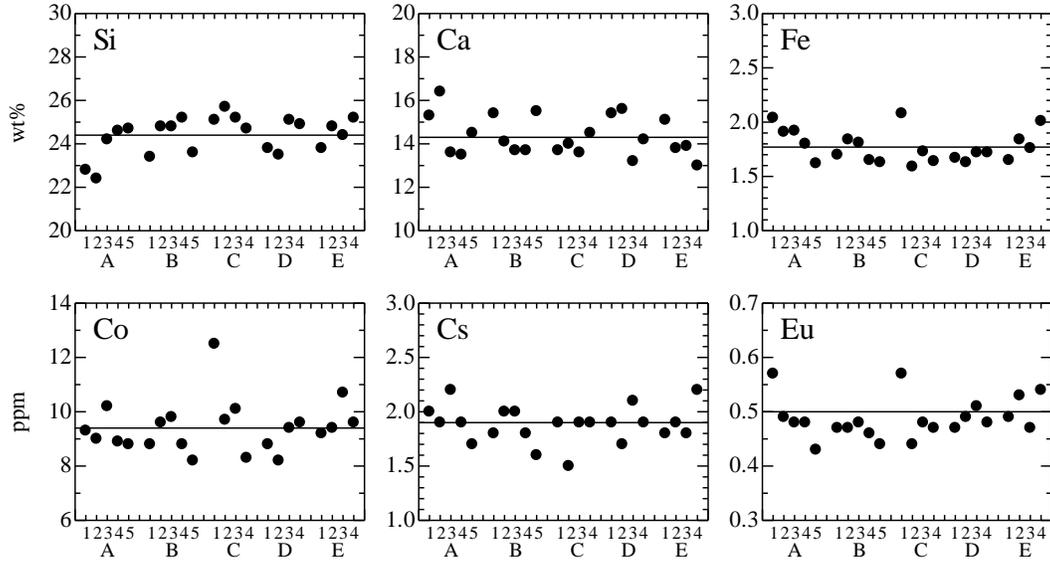
Fig. 2 Concentrations of Si, Ca, Fe, Co, Cs, and Eu in the sliced samples.
Solid lines denote the averages determined from the mixed sample.
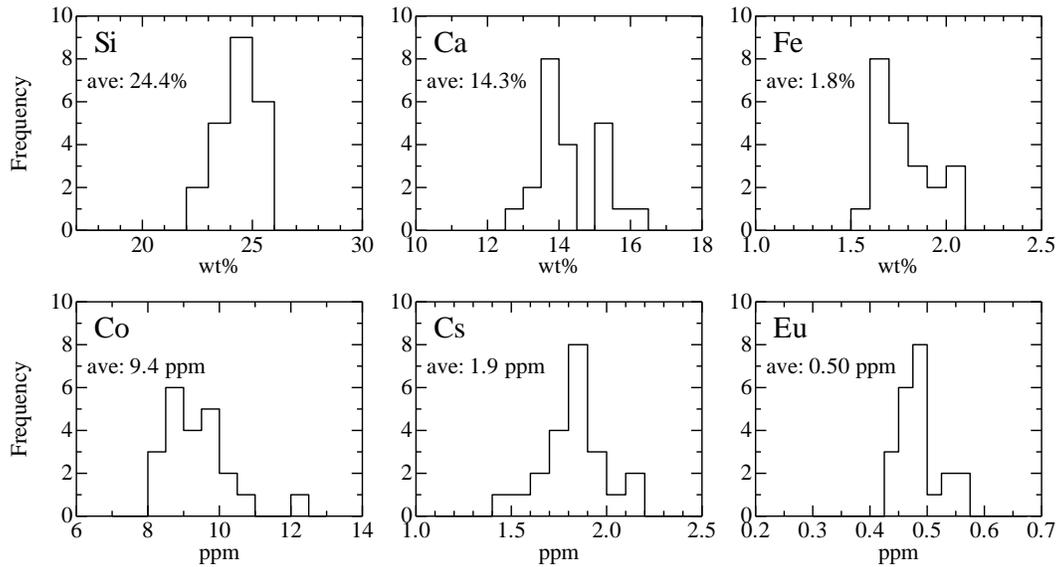

Fig. 3 Histograms of Si, Ca, Fe, Co, Cs, and Eu concentrations.

3.2. BOUNDARY DEPTH BETWEEN LLRW AND CLW

We examined a simulation of activities produced in the concrete which consist of the same elemental composition as NBS03 standard (McConn 2011) for main component and average values analyzed in the present work for Co, Cs, and Eu to evaluate the boundary depth between LLRW and CLW affected by the fluctuation of elemental composition. A density of the concrete was assumed to be 2.1 g cm^{-3}. The

elemental composition of the concrete is given in Table 1. We assumed that the concrete is exposed by the fission neutron with a flux of 1×10^8 cm^{-2} sec^{-1} for 40 years (Malkawi 2013). The flux approximately corresponds to the location outside biological shield (Remec 2016). The depth profiles of ^{41}Ca, ^{60}Co, ^{134}Cs, $^{152, 154}$Eu at 10 years after termination of activation were estimated using a monte-carlo simulation code of PHITS ver. 3.02 (Sato 2018, Iwamoto 2017) as shown in Fig. 4(a).

Table 1 Elemental composition used for the simulation

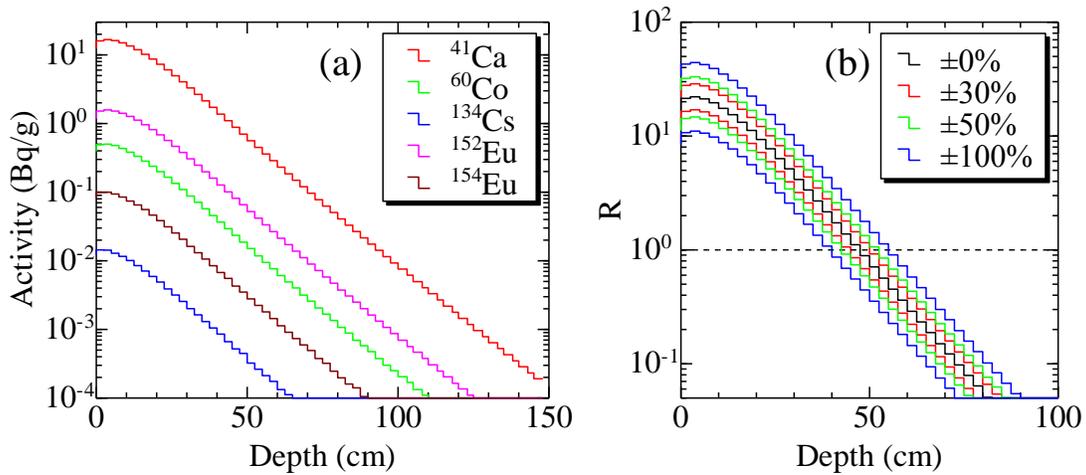| Element	Concentration
H	0.907 wt%
C	5.35 wt%
O	50.6 wt%
Si	15.5 wt%
Ca	26.4 wt%
Fe	1.18 wt%
Co	9.4 ppm
Cs	1.9 ppm
Eu	0.50 ppm

Fig. 4 Depth profiles activated by fission neutrons with a flux of 1×10^8 cm^{-2} sec^{-1} at 10 years after irradiation (a), summation of ratios to the clearance levels considering the fluctuation of elemental composition (b).

Each nuclide showed typical profile of the nuclide produced by a neutron capture reaction (Kinoshita 2009). The buildup of the activities is observed at a depth of 0 – 10 cm, and then, the activities decrease exponentially with a depth. Individual concentrations of the clearance levels are 100 Bq/g for ^{41}Ca and 0.1 Bq/g for ^{60}Co, ^{134}Cs, and $^{152, 154}$Eu according to the guideline (IAEA 2004). A summation of the ratios to the clearance level, R, which is calculated with Eq. (2), are shown in Fig. 4(b).

$$R = \sum \frac{A_n}{CL_n} \qquad (2)$$

Here, A_n and CL_n denote the activity concentration and is the concentration of the clearance level of nuclide, n, respectively. The radioactive wastes which the R is smaller than 1.0, can be treated as the CLW. In this case, the concretes located at a depth deeper than 50 cm can be treated as the CLW and the ones at shallower than 50 cm must be treated as the LLRW

The summation of the ratios to clearance levels, which 30%, 50%, and 100% uncertainty are assumed, are given in Fig. 4(b). Assuming +30%, +50%, and +100% fluctuations, the boundary depths between CLW and LLRW correspond to 52 cm, 53 cm, and 57 cm, respectively. The fluctuations with −30%, −50%, and −100% uncertainty were excluded because of the conservative evaluation. As described in Sec. 3.1, the elemental composition was fluctuated by 20 – 30% from the average concentrations when approximately 200 g of the concrete samples were analyzed. Assuming 30% fluctuation based on this work, the boundary depth is a few centimeters deeper for the safe evaluation.

CONCLUSION

We evaluated the fluctuation of the concentrations of Si, Ca, Fe, Co, Cs, and Eu in the concretes by analyzing 22 samples with approximately 200 g each taken from the concrete cores. The Si concentration had the smallest fluctuation in these elements. The concentrations of the other elements were fluctuated by 20 – 30%. Considering 30% fluctuation for the elemental composition, the boundary depth between CLW and LLRW was deeper by a few centimeters according to the simulation of activation using PHITS code (Sato 2018, Iwamoto 2017). More fluctuation should be considered when the elemental composition is analyzed from samples with smaller amount than 200 g. In addition, we should note that not only the fluctuation of elemental composition but also precision of simulation and elemental composition for the eventual safe evaluation.

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APPENDIX

Sample weight and concentration of each sample analyzed in the present work.

sample name	weight	Si	Ca	Fe	Co	Cs	Eu
	g	wt%	wt%	wt%	ppm	ppm	ppm
A-1	194.1	22.8	15.3	2.04	9.3	2.0	0.57
A-2	207.2	22.4	16.4	1.91	9.0	1.9	0.49
A-3	185.1	24.2	13.6	1.92	10.2	2.2	0.48
A-4	185.4	24.6	13.5	1.80	8.9	1.9	0.48
A-5	181.5	24.7	14.5	1.62	8.8	1.7	0.43
B-1	207.3	23.4	15.4	1.70	8.8	1.8	0.47
B-2	209.0	24.8	14.1	1.84	9.6	2.0	0.47
B-3	206.1	24.8	13.7	1.81	9.8	2.0	0.48
B-4	189.2	25.2	13.7	1.65	8.8	1.8	0.46
B-5	129.3	23.6	15.5	1.63	8.2	1.6	0.44
C-1	200.6	25.1	13.7	2.08	12.5	1.9	0.57
C-2	194.8	25.7	14.0	1.59	9.7	1.5	0.44
C-3	193.1	25.2	13.6	1.73	10.1	1.9	0.48
C-4	217.1	24.7	14.5	1.64	8.3	1.9	0.47
D-1	209.8	23.8	15.4	1.67	8.8	1.9	0.47
D-2	217.4	23.5	15.6	1.63	8.2	1.7	0.49
D-3	182.0	25.1	13.2	1.72	9.4	2.1	0.51
D-4	193.9	24.9	14.2	1.72	9.6	1.9	0.48
E-1	202.9	23.8	15.1	1.65	9.2	1.8	0.49
E-2	203.9	24.8	13.8	1.84	9.4	1.9	0.53
E-3	192.0	24.4	13.9	1.76	10.7	1.8	0.47
E-4	220.3	25.2	13.0	2.01	9.6	2.2	0.54
arithmetic mean	–	24.4	14.4	1.77	9.4	1.9	0.49
mixed sample	–	24.4	14.3	1.76	9.4	1.9	0.50