Evaluation of radioactive concentration produced in electric equipments and materials on the decommissioning of nuclear power plants

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A various cables used in nuclear power plants are treated as wastes in decommissioning. If part of them is treated as non-radioactive wastes, the amount of radioactive waste can be reduced. Furthermore, valuable resources such as copper can be recycled, and that contributes to the efficient use of resources. In this study, we have evaluated the produced nuclides in representative PWR cables by neutron irradiation calculated with the activation cross section data. As the results of evaluation, we have confirmed the possibility of the Ni-63 concentration exceeding the criterion for clearance near reactor vessel. To improve the accuracy of inventory evaluation, the uncertainty evaluation of activation cross section will be important.

1. Introduction

Aging nuclear power plants will increase in the coming decades, and most of them are expected to be shut down. Therefore decommissioning of nuclear power plant is one of the most important issue. Decommissioning of 1 GW class PWR (Pressurized Water Reactor) is estimated to generate hundreds kilotons of waste. If the part of them could be treated as non-radioactive wastes, the amount of radioactive waste could be reduced. Furthermore, valuable resources such as copper which used in electric equipments can be recycled, and that contributes to the efficient use of resources.

Radioactivity concentration of core component and main structure such as concrete or steel has been evaluated in several leading plant[1, 2], but sufficient investigation has not been conducted on electric components. Most of the cables laid outside the reactor pressure vessel are considered as non-radioactive waste due to relatively low neutron flux and small neutron cross section of copper.

In this study, we have evaluated the radioactivity inventory of copper material used in electric equipments.

2. Method

The evaluation method consists of the following three steps: (1) Chemical composition analysis of target material, (2) Neutron flux and spectrum evaluation at target position, (3) Activity inventory calculation.

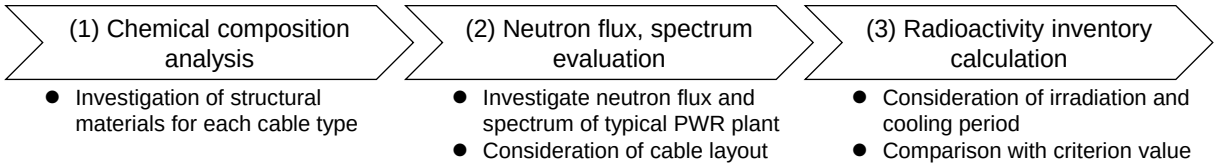

Fig. 1 Steps of evaluation

(1) Chemical composition analysis of target material

Cables used in PWR plants are roughly classified into five types: (A) High voltage cable, (B) Low voltage cable, (C) Control cable, (D) Instrumentation cable, and (E) special instrumentation cable. Figure 2 shows the cross sectional view of typical cable and table 1 shows the structural material of each layer. Then we have performed the chemical composition analysis using SEM/EDX (Scanning Electron Microscope /

Energy Dispersive X-ray Spectroscope) and ICP–OES (Inductivity coupled plasma optical emission spectrometer) for principal component. In addition, we have performed activation analysis using UTR-KINKI to investigate trace components below the detection limit of ICP–OES analysis.

Table 1 Structural material of each layer

	Layer	Cable type A: High voltage	B: Low voltage	C: Control	D: Instrument	E: Special
1	Conductor	Copper	TPCS	TPCS	TPCS	TPCS
2	Insulator	CLPE	FREP / FRV	FREP / FRV	FREP / FRV	FRV / CLPE
3	Strand	Jute	Jute	Jute	Jute	Jute
4	Shield	Copper tape	TPCT	TPCT	TPCT	TPCT
5	Tape	Fabric	Fabric	Fabric	Fabric	Fabric / PS
6	Sheath	FRV / FRPE	FRV / FRPE	FRV / FRPE	FRV / FRPE	FRV / FRPE

TPCS: Tin plated copper strand
CLPE: Cross-linked polyethylene
FRV: Flame retardance vinyl
TPCT: Tin plated copper tape
FREP: Flame retardance EP rubber
FRPE: Flame retardance polyethylene
PS: Polystyrene

(2) Neutron flux and spectrum evaluation at target position

Since most of cables are located outside the reactor pressure vessel in PWR plant, it is considered that thermal neutrons are dominant at the target position. Therefore, in this study we have focused on thermal neutrons. However, the thermal neutron flux at target position depends on the reactor type, then we used 10^{11} n/cm^2/s at the surface of reactor pressure vessel as the typical value of PWR plant[2, 3].

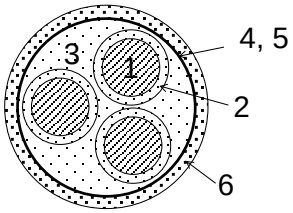

Fig. 2 Cross sectional view of typical cable

(3) Radioactivity inventory calculation

In this study, radioactivity inventory calculation was performed using PHITS 3.17 and DCHAIN-SP[4]. Considering that most of the reactors in Japan will be shutting down in 40 years and start the decommissioning process, we have set the irradiation period to 40 years. The neutron flux was constant during irradiation period without any suspension period due to the maintenance or accident. After irradiation period, the radioactivity concentration of each nuclide was compared with clearance level[5] as criterion.

3. Results

3.1 Chemical composition analysis of target material

(1) SEM/EDX measurement

We have performed SEM/EDX measurement to investigate the main component of each layer of cable. Each measurement samples that ware selected to cover the structural materials was polished the cut surface after fill the resin. SEM/EDX measurement was performed using Hitachi High Technologies Inc. SU-8000 with the acceleration voltage was 15 kV. As a results of measurement, metal elements except copper ware not detected from the conductor layer. On the other hand, Na, Mg, Al, Si, Cl, Ca, Zn, Sb and Pb were detected from other layer.

(2) ICP–OES measurement

We have performed ICP–OES measurement to investigate the trace component. Considering the SEM/EDX results, we have selected element to be measured for the following reasons:

- Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn: Elements have the potential to produce ^{63}Ni, ^{60}Co and ^{54}Mn by neutron reaction
- Mg, Al, Si, Ca, Sn, As, Bi, Pb: Elements have the potential to be contained as a trace element in conductor

For ICP–OES, each measurement sample was dissolved by adding acid and heating after disassemble and separated into conductor and other layer. ICP–OES measurement was performed using SII Nano Technology Inc. SPS-3100. Table 2 shows the result of measurement. Ca and Sn was detected as trace element. The detection limit was approximately several tens of ppm.

Table 2 Detected elements in conductor

Element	Composition (wt%)	Detection limit
Cu	99.95	2.1 ppm
Ca	1.192×10^{-3}	8.3 ppm
Sn	0.051	21 ppm

(3) Activation analysis

To investigate trace elements less than the detection limit of ICP–OES measurement, we have performed activation analysis using UTR–KINKI. Each measurement sample that was disassemble and separated into each layer filled in U–8 vessel has irradiated in the central stringer of the reactor. After irradiation, we have performed the radionuclides analysis produced from trace element by gamma spectroscopy using HPGe detector. As a result of analysis, another significant trace elements ware not detected. Therefore, we have decided the standard chemical composition of cable conductor as shown in table 2

3.2 Radioactivity inventory calculation

At the beginning, we have calculated average neutron flux inside the conductor region using PHITS 3.17 with parallel neutron beam to cable side. Figure 1 shows the two dimensional neutron flux.

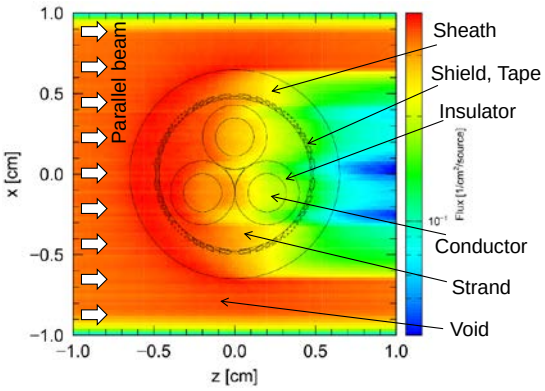

Fig. 3 Example of PHITS calculation (Two dimensional neutron flux)

Then we have calculated the radioactivity concentration in the conductor using DCHAIN-SP with JENDL/AD-2017[6] activation cross section. The irradiation period was 40 years, and the concentration was calculated from 20 years after the irradiation stopped. Figure 4 shows the calculation results. As a result of calculation, ^{41}Ca, ^{63}Ni, ^{65}Zn, $^{119,\,121,\,121m}$Sn, ^{125}Sb and ^{125m}Te was confirmed to be generated as a long half-life nuclide. Compared to the clearance level, radioactive concentration of ^{63}Ni and ^{65}Zn are greater than the clearance criterion at 40 years. Note that $^{119,\,121,\,121m}$Sn, ^{125}Sb, ^{125m}Te are excluded in this study due to the domestic regulation. Table 3 shows the calculated radioactivity concentration of ^{41}Ca, ^{63}Ni and ^{65}Zn at 40, 50 and 60 years.

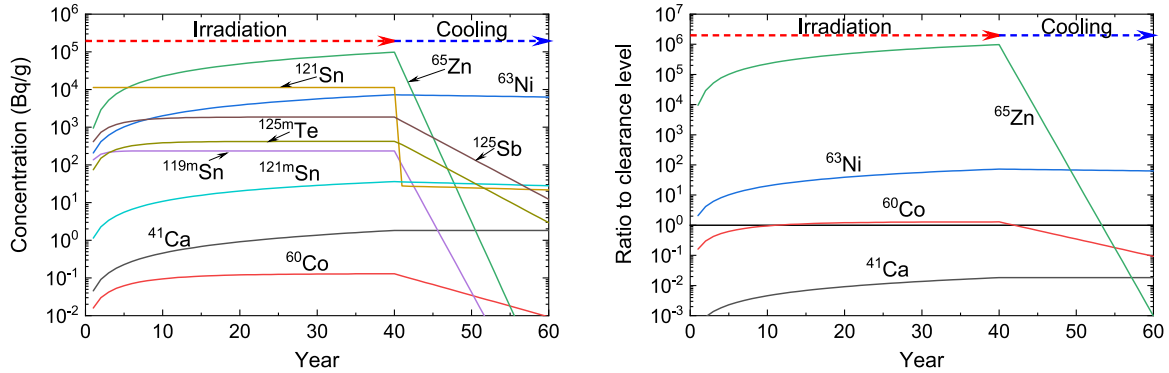

Fig. 4 Calculated radioactivity concentration (left) and ratio to clearance level (right)

Table 3 Calculated radioactivity concentration

Nuclide	$T_{1/2}$	Clearance level (Bq/g)	Radioactivity concentration (Bq/g)		
			40 y	50 y	60 y
^{41}Ca	1.03×10^{5} y	100	1.85	1.85	1.85
^{63}Ni	101.2 y	100	2.71×10^{3}	2.53×10^{3}	2.36×10^{3}
^{65}Zn	244.06 d	0.1	1.02×10^{5}	3.16	9.81×10^{-5}

4. Discussion

As a result of calculation, ^{65}Zn is dominant immediately after irradiation is stopped, however ^{63}Ni becomes dominant in a few years due to the short half-life of ^{65}Zn. Moreover, 10 years after irradiation stops, contribution of ^{63}Ni to clearance level becomes more than 90%. In this case, evaluation of ^{63}Ni radioactivity concentration must be required for clearance. However, direct measurement of ^{63}Ni is difficult due to its low energy β ray emission (maximum 0.0659 MeV, average 0.0174 MeV[7]) and no γ ray emission.

Therefore, it is necessary to consider evaluation using calculations including uncertainty evaluation. For this purpose, uncertainty evaluation of activation cross section is important. Figure 7 shows the comparison of cross section data between nuclear data libraries. The cross section of ^{40}Ca(n, γ)^{41}Ca reaction in each library is almost same. On the other hand, the cross section of ^{63}Cu(n, p)^{63}Ni reaction has large difference of about 10 times between libraries, especially in the thermal neutron region where no experimental data has been obtained. For this reason, it is considered that the uncertainty of the nuclear data contributes significantly to the uncertainty of the ^{63}Ni radioactivity inventory evaluation.

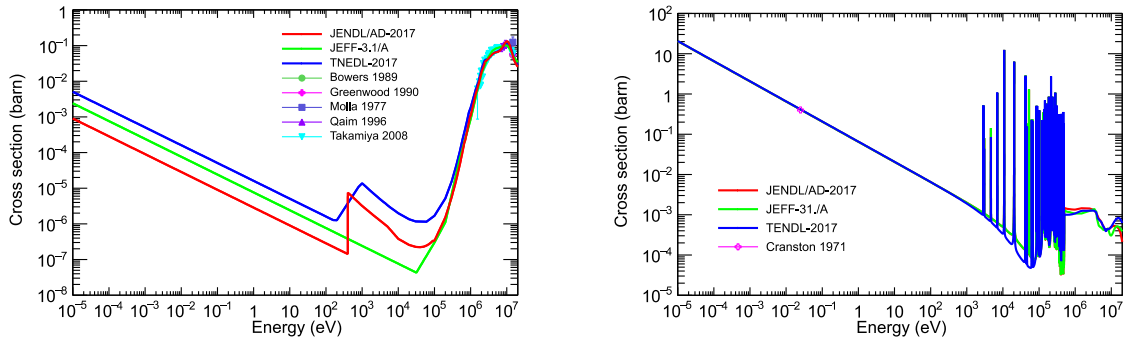

Fig. 5 Comparison between activation nuclear data libraries and experimental data of ^{63}Cu(n, p)^{63}Ni reaction (left) and ^{40}Ca(n, γ)^{41}Ca reaction (right)

5. Conclusion

In this study, we have evaluated the radioactivity inventory of copper material used in electric equipments by the basis of (1) Chemical composition analysis of target material, (2) Neutron flux and spectrum evaluation at target position and (3) Activity inventory calculation. As a result of evaluation, we have confirmed the radioactive concentration of ^{63}Ni and ^{65}Zn in the cable near the reactor vessel may exceed the clearance criterion and contribution of ^{63}Ni to clearance level becomes more than 90% after 10 years from irradiation stops. Since the direct measurement of ^{63}Ni is difficult, it is necessary to improve uncertainty evaluation of inventory calculation. For that purpose, the uncertainty evaluation of activation cross section is important.

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