Fast neutron fluence estimation by measurement of ^{93m}Nb and ^{93}Mo

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Abstract. The possibility for estimation of fast neutron fluence from ^{93m}Nb produced from Nb contained in structural component materials of nuclear power plants was investigated as an evaluation tool for extending plant operating lifetime. First, to establish a measurement method for ^{93m}Nb in structural component materials, chemical separations and measurements of ^{93m}Nb were carried out using specimens of fast neutron-irradiated XM-19 and X-750 alloy; these are used as structural component materials and contain Nb as a component. The ^{93m}Nb activities obtained from the measurements of both XM-19 and X-750 alloy agreed well with the calculated values of ^{93m}Nb, which were simply calculated from the evaluated fast neutron fluence. On the other hand, in the case of type 316L stainless steel, which contains Mo, Mo derived ^{93m}Nb presents a problem, so an approach was used that estimates the amount of Mo derived ^{93m}Nb based on ^{93}Mo measurements with the third specimen, neutron-irradiated type 316L stainless steel. The difference between the measured and calculated ^{93m}Nb values was about two-fold, indicating that the estimation method for ^{93m}Nb produced from Mo needs to be improved.

1 Introduction

One of the approaches for improving the estimation accuracy of fast neutron fluence experienced by nuclear power plant structural component materials is to estimate the fast neutron fluence based on the measurement of the activity of the activated nuclides in the materials. This means that, the results of measurements made after chemical separations of nuclides produced by the reaction between elements contained in the structural materials and fast neutrons are reflected in the estimation of the fast neutron fluence. ^{93m}Nb produced by the ^{93}Nb(n, n')^{93m}Nb reaction between Nb and fast neutrons has a relatively long half-life of 16.13 years and is used to estimate long-term fast neutron fluence. Types 304 and 316L stainless steels (SSs) are mainly used for structural components such as core shrouds and upper grid plates in BWRs, and it has been reported that type 304 SS contains several ppm to several tens of ppm of Nb [1], which may allow estimation of fast neutron fluence by the ^{93}Nb(n, n')^{93m}Nb reaction. In fact, estimations of fast neutron fluence from the ^{93}Nb(n,

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n')^{93m}Nb reaction have been made using Nb present in structural components on the order of several tens of mass ppm [2-4]. On the other hand, in the case of a material containing Mo such as type 316L SS, ^{93m}Nb is also produced from Mo in that structural material [3, 4], so the amount of ^{93m}Nb derived from Mo must be estimated. ^{93m}Nb from Mo is produced by the electron capture decay of ^{93}Mo that was produced from the thermal neutron capture reaction of ^{92}Mo, which is one of the stable isotopes of Mo. Therefore, in the case of a material containing Mo, ^{93m}Nb produced from both Mo and Nb is included, and if the activity of ^{93m}Nb produced from Mo is not negligible, a correction is necessary to subtract the activity of ^{93m}Nb produced from Mo from the ^{93m}Nb measured activity. (Of course, if the activity of ^{93m}Nb produced from Mo is overwhelmingly greater than the activity of ^{93m}Nb produced from Nb, the fast neutron flux from ^{93}Nb(n, n')^{93m}Nb cannot be estimated.) As a method for accurately estimating the produced amount of ^{93m}Nb from Mo, an estimation from the measured value of ^{93}Mo is effective, as ^{93}Mo is an intermediate product in the production of ^{93m}Nb from Mo.

In this study, the ^{93m}Nb measurement method was established using irradiated specimens of XM-19 and X-750 alloy containing Nb as a constituent element whose amount is within the material specifications such as set by the ASTM. The validity of the measurement method was verified by comparing the measured value with the calculated value of ^{93m}Nb estimated from the evaluated fast neutron fluence. The validated ^{93m}Nb measurement method was used to measure ^{93m}Nb for the third specimen, an irradiated type 316L SS specimen. To estimate the amount of ^{93m}Nb produced from Mo, ^{93}Mo was also measured for the above three specimens and the amount of ^{93m}Nb produced from Mo was estimated. The activity of ^{93m}Nb and ^{93}Mo of each specimen were measured by X-ray measurements with a Ge semiconductor detector after dissolving each specimen and separating ^{93m}Nb and ^{93}Mo from each other by an ion exchange separation method. The chemical separation between ^{93m}Nb and ^{93}Mo is necessary, because both are measured by X-ray measurements and both are measured at the same X-ray energy of 16.6 keV.

2 Experimental

2.1 Specimens

Irradiated XM-19, X-750 alloy and type 316L SS, which are used as structural component materials, were used as specimens and there was one test piece for each. XM-19 and X-750 alloy were used as specimens containing Nb as a component to establish the ^{93m}Nb measurement method. Table 1 summarizes their compositions listed on the respective mill test reports. In the absence of Nb or Mo concentration data on the mill test reports, the concentration was determined by measuring the solution of each specimen by inductively coupled plasma mass spectrometry (ICP-MS). The concentration measurements were performed after confirming sufficient accuracy in advance.

All specimens were irradiated in the Japan Materials Testing Reactor (JMTR) under the conditions shown in Table 2. The fast neutron fluence integrated by the specimens is equivalent to the cumulative irradiation in the core shrouds of Japanese BWRs during a period of 60 years.

Table 1. Composition of specimens.

Material	Composition (wt%)											
	C	Si	Mn	P	S	Ni	Cr	Mo	Nb	Ti	Al	Fe
XM-19	0.03	0.44	5.23	0.007	0.002	12.14	21.23	2.20	0.21	0.004	0.01	Bal.
X-750 alloy	0.047	0.13	0.11	0.007	0.003	72.18	15.42	—	0.99	2.68	0.60	7.6
316L SS	0.008	0.51	0.95	0.018	0.006	12.58	16.49	2.16	—	—	—	Bal.

Table 2. Irradiation conditions of specimens.

Reactor	JMTR (Thermal Power: 50MW)
Period	Jan. 1, 1999-May 14, 2001
Cycle	12 cycles
Time	299 days (2.58×10^{7} s)
Max. neutron fluence (Fast)	2.66×10^{21} n/cm^2 (E > 1 MeV)
Neutron fluence (Thermal)	4.13×10^{21} n/cm^2 (E < 0.683 eV) at max. neutron fluence (Fast)
Neutron flux (Fast)	1.03×10^{14} n/cm^2 s (E > 1 MeV)
Neutron flux (Thermal)	1.60×10^{14} n/cm^2 s (E < 0.683 eV) at max. neutron fluence (Fast)

2.2 Measurements

2.2.1 Separation of Mo and Nb

The separation of Nb and Mo was performed by applying an ion exchange separation method using a hydrofluoric acid system, with reference to the ^{93m}Nb separation method of Serén and Kekki [4] and the report of Fujimoto and Shimura [5] about element adsorption behaviors on an anion exchange resin in hydrofluoric acid and hydrochloric acid systems.

Approximately 20 mg of each specimen was dissolved in a mixture of nitric acid (HNO_3), hydrochloric acid (HCl), and hydrofluoric acid (HF) while heating on a hot plate. Then, the acidic solutions of dissolved specimens were evaporated to dryness by further hot plate heating. The residues were dissolved in 2 mol/L HF, and passed through an anion exchange column filled with DOWEX 1X-8 200-400 mesh. In the next step, 2 mol/L HF was passed through the column. This step flushed out the main structural material elements such as Fe, and the major radionuclides such as ^{60}Co, from the column. Next, Mo was eluted from the column using a mixture of 8 mol/L HF and 4 mol/L HCl. Finally, Nb was eluted from the column using 1 mol/L HCl.

2.2.2 Measurement of ^{93}Mo

The eluant of 8 mol/L HF and 4 mol/L HCl containing Mo was evaporated to dryness by heating on a hot plate and the residue was dissolved in 1 mol/L HNO_3. Then, 1 mg of lanthanum was added to the solution, and lanthanum hydroxide precipitate was formed by adding ammonia water. In this case, Mo remained in the liquid phase, while ^{60}Co, which could not be removed by ion exchange separation, was included on the precipitate side. After a vacuum filtration, 100 μg of Mo was added to the collected filtrate, which was then adjusted to a slightly acidic condition as confirmed by pH paper testing by adding HCl. Then bromine water and an ethanolic solution of α-benzoin oxime were added to get Mo precipitate. The precipitate was collected on a membrane filter (47 mm diameter; 0.45 μm pore size). The activity of ^{93}Mo in the precipitate was determined with a high-purity germanium low energy photon spectrometer (LEPS) by measuring the 16.6 keV radiation emitted by ^{93}Mo. To

estimate the recovery, the amount of Mo in the precipitate was determined by X-ray fluorescence (XRF). The LEPS and XRF apparatus were calibrated using standard samples.

2.2.3 Measurement of ^{93m}Nb

To remove HF, the 1 mol/L HCl solution containing Nb was evaporated to dryness by heating on a hot plate and the residue was dissolved with 1 mol/L HCl. After 500 μg of lanthanum was added to the solution, ammonia water was added to precipitate lanthanum hydroxide, and Nb was coprecipitated. The precipitate was collected using a membrane filter (47 mm diameter; 0.45 μm pore size). The activity of ^{93m}Nb in the precipitate was determined with the LEPS by measuring the 16.6 keV radiation emitted by ^{93m}Nb. To estimate the recovery of Nb, the activity of ^{94}Nb in the precipitate was determined with a germanium semiconductor detector. The LEPS and germanium semiconductor detectors were calibrated using standard samples.

2.3 Calculations

2.3.1 Activity of ^{93m}Nb from ^{93}Nb

The activity of ^{93m}Nb produced from ^{93}Nb ($A_{\mathrm{Nb93m(Nb)}}$) is calculated as follows:

$$A_{\mathrm{Nb93m(Nb)}} = N_{\mathrm{Nb93}}\,\sigma_{\mathrm{Nb93}}\,\varphi_{\mathrm{f}}\left(1 - e^{-\lambda_{\mathrm{Nb93m}}\,t}\right) e^{-\lambda_{\mathrm{Nb93m}}\,t_{\mathrm{m}}} \tag{1}$$

where, N_{Nb93} is the number of atoms of ^{93}Nb in the specimen, σ_{Nb93} is the cross section of the reaction ^{93}Nb(n, n')^{93m}Nb, φ_{f} is the fast neutron flux, λ_{Nb93m} is the decay constant of ^{93m}Nb, t is the irradiation time, and t_{m} is the time from the end of irradiation to the measurement.

In the calculations of this study, the cross section of ^{93}Nb(n, n')^{93m}Nb and the fast neutron flux were assumed to be constant. For the fast neutron flux values, the values shown in Table 2 were used. For the cross section values, the values corresponding to 2 MeV, the average energy of the fission spectrum, were used. The activity of ^{93m}Nb calculated from equation (1) was compared with the measured activity of ^{93m}Nb for each specimen to confirm the validity of the measured values.

2.3.2 Activity of ^{93m}Nb from ^{93}Mo

^{92}Mo produces ^{93}Mo by thermal neutron capture, and ^{93}Mo disintegrates to ^{93m}Nb. In this study, it was assumed that all the ^{92}Mo reacted with neutrons to become ^{93}Mo, and all the ^{93}Mo formed decayed to ^{93m}Nb. The activity of ^{93m}Nb produced from ^{93}Mo ($A_{\mathrm{Nb93m(Mo)}}$) is calculated as follows:

$$A_{\mathrm{Nb93m(Mo)}} = \lambda_{\mathrm{Nb93m}}\,N'_{\mathrm{Nb93m}} \tag{2}$$

$$N'_{\mathrm{Nb93m}} = \frac{\lambda_{\mathrm{Mo93}}}{\lambda_{\mathrm{Nb93m}} - \lambda_{\mathrm{Mo93}}}\,N_{\mathrm{Mo93}}\left(\mathrm{e}^{-\lambda_{\mathrm{Mo93}}\,t_{\mathrm{m}}} - \mathrm{e}^{-\lambda_{\mathrm{Nb93m}}\,t_{\mathrm{m}}}\right) + N_{\mathrm{Nb93m}}\,\mathrm{e}^{-\lambda_{\mathrm{Nb93m}}\,t_{\mathrm{m}}} \tag{3}$$

where, N'_{Nb93m} is the number of atoms of ^{93m}Nb produced from ^{92}Mo during the period between irradiation and measurement, λ_{Mo93} is the decay constant of ^{93}Mo, and N_{Mo93} and N_{Nb93m} are the numbers of ^{93}Mo and ^{93m}Nb atoms at the end of irradiation. N_{Mo93} and N_{Nb93m} are calculated as follows:

$$N_{\mathrm{Mo93}} = N_{\mathrm{Mo92}}\,\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}\left(\frac{\mathrm{e}^{-\,\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}\,t}}{\lambda_{\mathrm{Mo93}} - \sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}} + \frac{\mathrm{e}^{-\,\lambda_{\mathrm{Mo93}}\,t}}{\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}} - \lambda_{\mathrm{Mo93}}}\right) \tag{4}$$

$$N_{\mathrm{Nb93m}} = N_{\mathrm{Mo92}}\,\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}\,\lambda_{\mathrm{Mo93}}\left[\frac{\mathrm{e}^{-\,\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}\,t}}{(\lambda_{\mathrm{Mo93}} - \sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}})(\lambda_{\mathrm{Nb93m}} - \sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}})} + \frac{\mathrm{e}^{-\,\lambda_{\mathrm{Mo93}}\,t}}{(\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}} - \lambda_{\mathrm{Mo93}})(\lambda_{\mathrm{Nb93m}} - \lambda_{\mathrm{Mo93}})} + \frac{\mathrm{e}^{-\,\lambda_{\mathrm{Nb93m}}\,t}}{(\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}} - \lambda_{\mathrm{Nb93m}})(\lambda_{\mathrm{Mo93}} - \lambda_{\mathrm{Nb93m}})}\right] \tag{5}$$

where, N_{Mo92} is the initial number of atoms of ^{92}Mo in the specimen, σ_{Mo92} is the cross section of ^{92}Mo(n, γ)^{93}Mo, and φ_{th} is the thermal neutron flux. Here, φ_{th} is the thermal neutron flux that contributes to the reaction of ^{92}Mo(n, γ)^{93}Mo, and it was estimated from the measured activity of ^{93}Mo along with σ_{Mo92}. N_{Mo93} in equation (4) can be estimated from equation (6) based on the measured activity of ^{93}Mo (A_{Mo93}).

$$N_{\mathrm{Mo93}} = \frac{A_{\mathrm{Mo93}}}{\lambda_{\mathrm{Mo93}}\,\mathrm{e}^{-\,\lambda_{\mathrm{Mo93}}\,t_{\mathrm{m}}}} \tag{6}$$

When N_{Mo93} is obtained, $\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}$ can also be obtained from equation (4). Furthermore, once $\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}$ is obtained, N_{Nb93m} can also be obtained from equation (5).

Therefore, the procedure to obtain the activity of ^{93m}Nb produced from Mo based on the measured activity of ^{93}Mo is the following.

First, the number of atoms of ^{93}Mo at the end of irradiation, N_{Mo93}, is obtained from the measured activity of ^{93}Mo by equation (6). Next, after substituting the obtained N_{Mo93} into equation (4), the product of the cross section and thermal neutron flux, $\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}$, is obtained. Then, by substituting $\sigma_{\mathrm{Mo92}}\,\varphi_{\mathrm{th}}$ into equation (5), the number of atoms of ^{93m}Nb produced from ^{92}Mo at the end of irradiation, N_{Nb93m}, is obtained. By substituting N_{Mo93} and N_{Nb93m} into equation (3), the number of atoms of ^{93m}Nb produced from ^{92}Mo during the period between irradiation and measurement, N'_{Nb93m}, is obtained. Finally, N'_{Nb93m} is converted to the activity of ^{93m}Nb, $A_{\mathrm{Nb93m(Mo)}}$, by equation (2).

In this study, the activity of ^{93m}Nb produced from Mo in each specimen was evaluated using the measured activity of ^{93}Mo in each specimen and the above procedure.

3 Results and discussion

Table 3 lists the concentrations of Nb and Mo and the activities of ^{93m}Nb and ^{93}Mo for each specimen. The Nb/Mo ratio shown in Table 3 indicates the concentration ratio of Nb and Mo in each specimen. The Nb concentration of type 316L SS and the Mo concentration of X-750 alloy are the values obtained by measuring the solution of each specimen by ICP-MS. The Nb concentration of type 316L SS is 0.00054 % (= 5.4 ppm), which is similar to the reported analytical value of type 304 SS [1].

Table 3. Results of analysis.

Material	Mass (g)	Nb concentration in specimen (wt%)	Mo concentration in specimen (wt%)	Nb/Mo ratio	^{93m}Nb activity (Bq)	^{93}Mo activity (Bq)
XM-19	0.0192	0.21	2.2	9.5×10^{-2}	8.9×10^{4}	1.2×10^{2}
X-750 alloy	0.0196	0.99	0.27	3.7	4.7×10^{5}	1.3×10^{1}
316L SS	0.0200	0.00054	2.16	2.5×10^{-4}	6.2×10^{2}	1.6×10^{2}

Table 4 allows a comparison of the measured activity of ^{93m}Nb (M) and the calculated activity of ^{93m}Nb produced from Nb (C). The ratios of the measured and calculated values of XM-19 and X-750 alloy are 1.1 and 1.2, which are in relatively good agreement, and the measured values of ^{93m}Nb obtained by the measurement method in this study are reasonable.

On the other hand, in type 316L SS, the ratio of measured to calculated values is 2.8, and the agreement is poor. Focusing on the Nb/Mo ratio in each specimen, the ratio of type 316L SS is lower than the ratios of the other two. Baers and Hasanen [3] reported that the Nb/Mo ratio must be greater than a certain value (Nb/Mo ratio > 0.01 in their study) for the ^{93m}Nb produced from Nb to be dominant. This means that when the Nb/Mo ratio is low, the proportion of ^{93m}Nb produced from Mo in the activity of ^{93m}Nb in the specimen becomes large, and ^{93m}Nb produced from Mo cannot be neglected. In type 316L SS, the Nb/Mo ratio is low and the measured activity is higher than the calculated activity. This difference between measured and calculated activities suggests that the actual activity (i.e., the measured activity) was affected by ^{93m}Nb produced from Mo, resulting in a discrepancy between the measured and calculated values.

Table 5 shows the results of estimating the activity of ^{93m}Nb produced from ^{92}Mo based on the measured activity of ^{93}Mo shown in Table 3. The calculated activity of ^{93m}Nb produced from Mo in XM-19 and X-750 alloy is sufficiently small compared to the measured activity of ^{93m}Nb as shown in Table 4. On the other hand, in type 316L SS, the calculated activity of ^{93m}Nb produced from Mo is only about one order of magnitude smaller than the measured value, which is a non-negligible amount.

Table 6 is a comparison of the measured activity of ^{93m}Nb after subtracting the activity of ^{93m}Nb produced from ^{92}Mo (M') and the calculated activity of ^{93m}Nb produced from Nb (C). Even after subtracting the activity of ^{93m}Nb produced from Mo, the ratio of measured to calculated values in type 316L SS is 2.4, which is a slight improvement, but there is still a discrepancy compared to the XM-19 and X-750 alloy values. The reason for the discrepancy between the calculated and measured values may be due to uncertainty in the measured value of ^{93}Mo, uncertainty in the calculation model for the amount of ^{93m}Nb produced from ^{92}Mo, or other factors. The calculation of ^{93m}Nb produced from ^{92}Mo should be done in detail using an analysis code such as ORIGEN2[6].

Table 4. Comparison of the measured activity of ^{93m}Nb and the calculated activity of ^{93m}Nb produced from Nb.

Material	M Measured activity of ^{93m}Nb (Bq)	C Calculated activity of ^{93m}Nb produced from Nb (Bq)	M/C
XM-19	8.9×10^{4}	8.4×10^{4}	1.1
X-750 alloy	4.7×10^{5}	4.0×10^{5}	1.2
316L SS	6.2×10^{2}	2.2×10^{2}	2.8

Table 5. Calculated activity of ^{93m}Nb produced from ^{92}Mo.

Material	Calculated activity of ^{93m}Nb produced from ^{92}Mo (Bq)
XM-19	6.5×10^{1}
X-750 alloy	7.1
316L SS	8.7×10^{1}

Table 6. Comparison of the measured activity of ^{93m}Nb subtracting ^{93m}Nb produced from ^{92}Mo and the calculated activity of ^{93m}Nb produced from Nb.

Material	M' Measured activity of ^{93m}Nb subtracting ^{93m}Nb produced from ^{92}Mo (Bq)	C Calculated activity of ^{93m}Nb produced from Nb (Bq)	M'/C
XM-19	8.9×10^{4}	8.4×10^{4}	1.1
X-750 alloy	4.7×10^{5}	4.0×10^{5}	1.2
316L SS	5.3×10^{2}	2.2×10^{2}	2.4

4 Conclusions

In this study, the measurement method of ^{93m}Nb was confirmed using three irradiation specimens of XM-19, X-750 alloy and type 316L SS. In addition, as an estimation method of Mo derived ^{93m}Nb when the influence of ^{93m}Nb produced from Mo cannot be ignored, an estimation method of ^{93m}Nb produced from Mo based on the measured ^{93}Mo was examined.

The measured activity of ^{93m}Nb for XM-19 and X-750 alloy agreed well with the calculated activity assuming that ^{93}Nb is the source, and it was confirmed that the method used in this study can obtain reasonable ^{93m}Nb measurements.

On the other hand, in type 316 L SS with low Nb concentration relative to Mo concentration, compared to XM-19 and X-750 alloy, the measured activity of ^{93m}Nb did not agree well with the calculated activity assuming that ^{93}Nb is the source, and the effect of ^{93m}Nb from Mo was confirmed. Therefore, ^{93m}Nb from Mo was evaluated based on ^{93}Mo measurements and subtracted from the ^{93m}Nb measurements. However, the difference between the measured and calculated values of ^{93m}Nb was still about two-fold, indicating that the estimation method for ^{93m}Nb produced from Mo needs to be improved. It is planned to investigate the amount of ^{93m}Nb produced from Mo in detail in the future.

In addition, type 304 SS, which was not treated in this study, does not contain Mo as a constituent element, and there is a possibility that ^{93m}Nb produced from Mo can be neglected, and it is planned to study the feasibility of fast neutron fluence estimation from Nb contained in type 304 SS also.

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