Anion exchange separation for solutions containing irradiated steel specimen used to estimate neutron fluence

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Abstract

Conventionally, the amount of radioactivity of the ^{54}Mn activation product generated in iron dosimeters is measured to evaluate the neutron fluence of surveillance test specimens for a reactor pressure vessel. Since almost all Japanese nuclear power plants have remained offline for more than 5 years following the Great East Japan Earthquake, the amount of ^{54}Mn in the iron dosimeters has been steadily decreasing. Therefore, in dosimetry measurement by γ-spectrometry, it is difficult to detect the low-level activity of ^{54}Mn due to the spectral interference caused by the overwhelming presence of another activation production ^{60}Co also generated in the iron dosimeters. In this study, the anion exchange separation of ^{60}Co in a solution containing a dissolved irradiated steel specimen was conducted to evaluate its applicability to the dosimetry measurement procedure by γ-spectrometry.

A neutron irradiated steel specimen containing ^{54}Mn, ^{60}Co and the stable isotope ^{55}Mn was dissolved in HCl and the obtained solution was passed through an anion exchange resin column (Dowex 1x8, 100-200 mesh) using 9 mol dm^{-3} HCl. The eluate from the resin was collected, and the amount of ^{60}Co was determined by γ-spectrometry. The amount of stable Mn in the eluate was determined by inductively coupled plasma-mass spectrometry to estimate the recovery ratio of the separation procedure for Mn. The anion exchange separation removed ^{60}Co from the dissolved solution of irradiated steel specimen to a level of less than 10^{-4} of the initial amount and the recovery ratio of about 99 % was obtained for Mn.

1. Introduction

The amount of radioactivity of ^{54}Mn activation product generated in iron dosimeters is measured to evaluate the irradiation fluence of surveillance test specimens in a reactor pressure vessel. Fig.1 shows the transition of estimated radioactivity of ^{54}Mn in an iron dosimeter. Conventional surveillance tests were conducted within a year after reactor shutdowns, and at that time, the amount of radioactivity of ^{54}Mn in the iron dosimeters was sufficiently high enough to measure. However, almost all nuclear power plants in Japan have remained offline for more than 5 years following the Great East Japan Earthquake, and surveillance tests have not been conducted since. The amount of radioactivity of ^{54}Mn in the iron dosimeters has been steadily decreasing. The amount of radioactivity of ^{54}Mn will decrease to less than its detection limit within several years and neutron fluence evaluation using the iron dosimeters will not be able to be done finally. The amount of ^{54}Mn is measured by γ-spectrometry, and its detection limit is affected by ^{60}Co that is also generated in the stainless steel specimens. Fig.2 shows γ-spectrum of a specimen including ^{60}Co. The background value for ^{54}Mn in γ-spectrometry was elevated by Compton scattering caused by ^{60}Co.Therefore, in order to measure a lower activity of ^{54}Mn, an effective procedure is removing ^{60}Co from the solutions prepared by dissolving the iron dosimeters.

An anion exchange reaction using HCl is effective in removing Co. Co is strongly adsorbed on the anion exchange resin in high concentration HCl (7 to 12 mol dm^{-3}), whereas Mn is only slightly adsorbed or not adsorbed at all at the same HCl concentration [1].

The purpose of this study was to confirm the applicability of an anion exchange separation method to remove ^{60}Co with high recovery ratio for ^{54}Mn from solutions which are prepared from irradiated steel specimens to measure radioactivity of ^{54}Mn by γ-spectrometry.

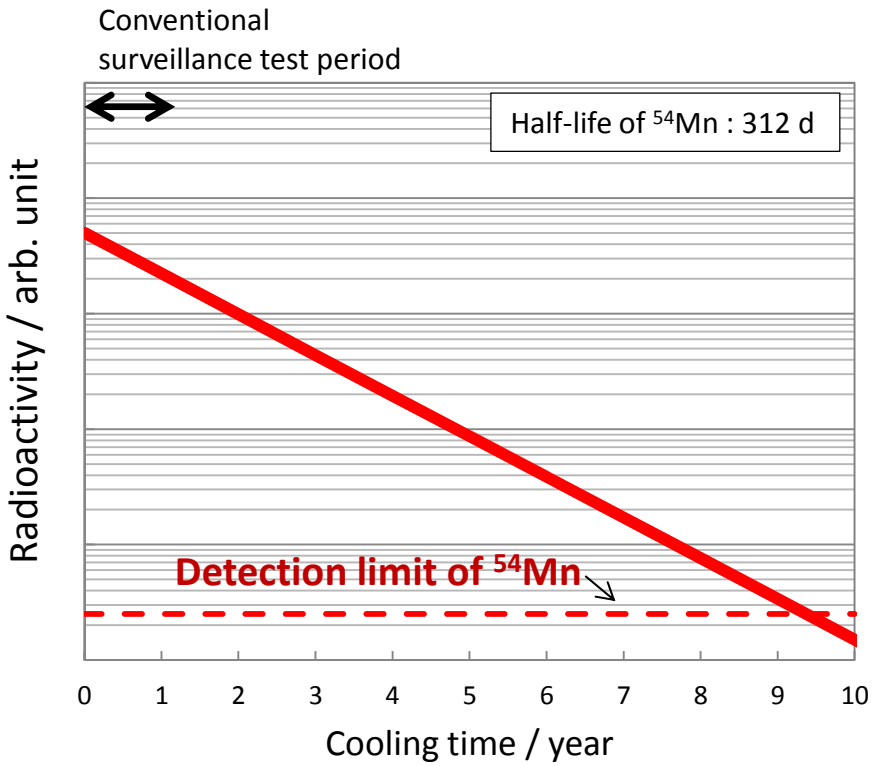

Fig. 1 Transition of the estimated radioactivity of ^{54}Mn in an iron dosimeter. The Detection limit is in the case of measuring ^{54}Mn by γ spectrometer (ORTEC, GEM10P4).

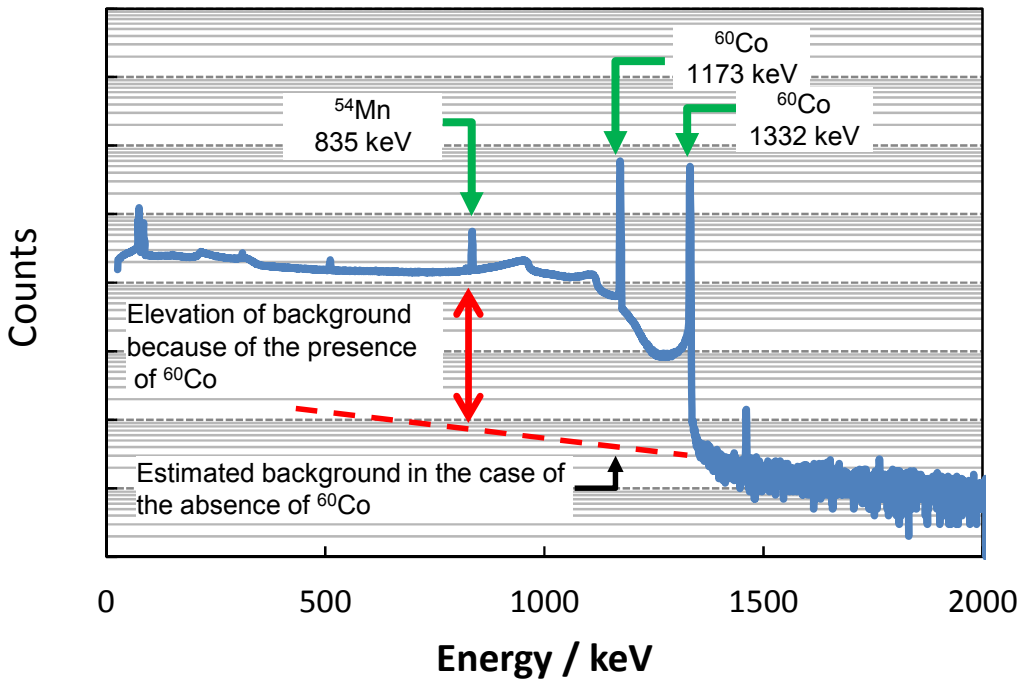

Fig. 2 Schematic γ-spectrum of a specimen containing ^{60}Co

2. Experimental

First, the eluate volume which is suitable to recovery Mn in an anion exchange resin was examined using stable isotopes of Mn and Co. The eluate was HCl, and its concentration was determined from reference [1]. The 9 mol dm^{-3} HCl containing 1 mg amounts of the stable isotopes of Mn and Co was loaded onto an anion exchange column (diameter: 10 mm, height: 60 mm) containing about 5 cm^3 of Dowex 1x8 100-200 mesh resin. Then 5 cm^3 of 9 mol dm^{-3} HCl was passed through the anion exchange resin in a batch, and 50 cm3 of eluate was collected. The amounts of Mn in the eluate were measured with an inductively coupled plasma-mass spectrometer (Seiko Instruments Inc., SPQ9400).The recovery ratio of Mn was estimated as the amount of Mn in the eluate divided by the initial amount of stable isotope of Mn (= about 1 mg). From this result, the optimum eluate volume for recovering almost all the Mn from the column was determined.

Next, neutron irradiated steel that included ^{60}Co and ^{54}Mn was used to examine applicability of the separation procedure. The irradiated steel was type 316L stainless steel irradiated in a test reactor up to 7.3×10^{25} n m^{-2} (E >1 MeV). The amount of irradiated steel specimen was a few hundred micrograms and its radioactivity was shown in Table 1. Fig. 3 shows the separation scheme. The specimen was dissolved with 9 mol dm^{-3} HCl, and then 1 mg amounts of the stable isotopes of Mn and Co were added as carriers. The acid solution containing the dissolved specimen and carriers was transferred to the anion exchange column (diameter: 10 mm, height: 60 mm) containing about 5 cm^3 of Dowex 1x8 100-200 mesh resin. Mn was eluted from the resin with 9 mol dm^{-3} HCl. The recovery ratio of ^{54}Mn was estimated as the amount of stable isotope of Mn in the eluate divided by the initial amount of stable isotope of Mn (= about 1 mg). The amount of radioactivity of ^{60}Co was measured with a calibrated γ-spectrometer (ORTEC, GEM10P4). The amount of stable isotope of Mn (^{55}Mn) was measured with the inductively coupled plasma-mass spectrometer.

Table 1 Radioactivity of the irradiated steel

Nuclide	Radioactivity / Bq
Mn-54	1.2×10^3
Co-60	1.0×10^6

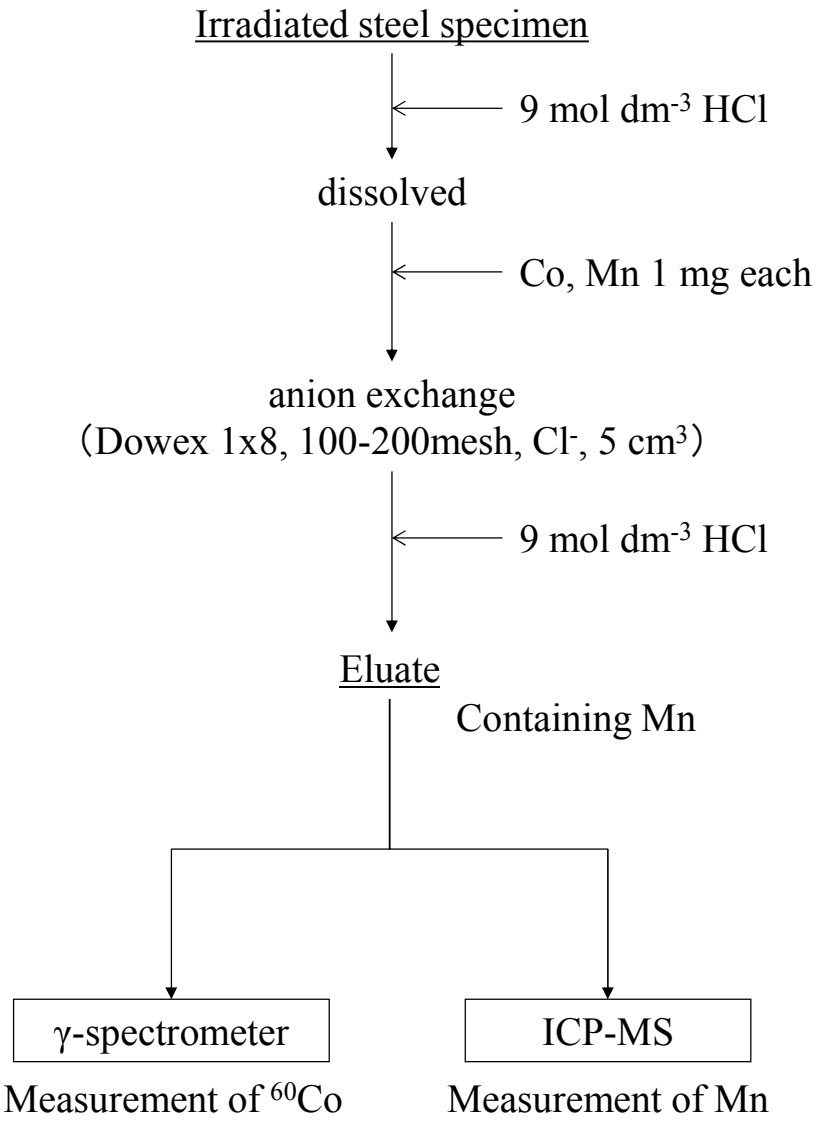

Fig. 3 Separation scheme

3. Results

Fig. 4 shows elution pattern of Mn in the anion exchange obtained in the experiment using stable isotopes. It was found that almost all the Mn could be recovered using 35 cm^3 HCl in the anion exchange. In the experiment using the irradiated steel specimen, the Mn recovery ratio was 98.9% as shown in Table 2.

Fig. 5 shows γ-spectra of the irradiated steel specimen before and after the anion exchange separation. In spectrum of the specimen before the anion exchange separation, Compton scattering caused by ^{60}Co overlapped ^{54}Mn gamma peaks. On the other hand, ^{54}Mn gamma peaks in the spectrum of the specimen after the anion exchange separation was clearly appeared. Table 3 shows the amount of radioactivity of ^{60}Co in the specimen before and after the anion exchange separation. The amount of

radioactivity of ^{60}Co after the anion exchange separation was reduced to less than 10^{-4} of the initial amount of radioactivity, indicating that ^{60}Co was effectively removed.

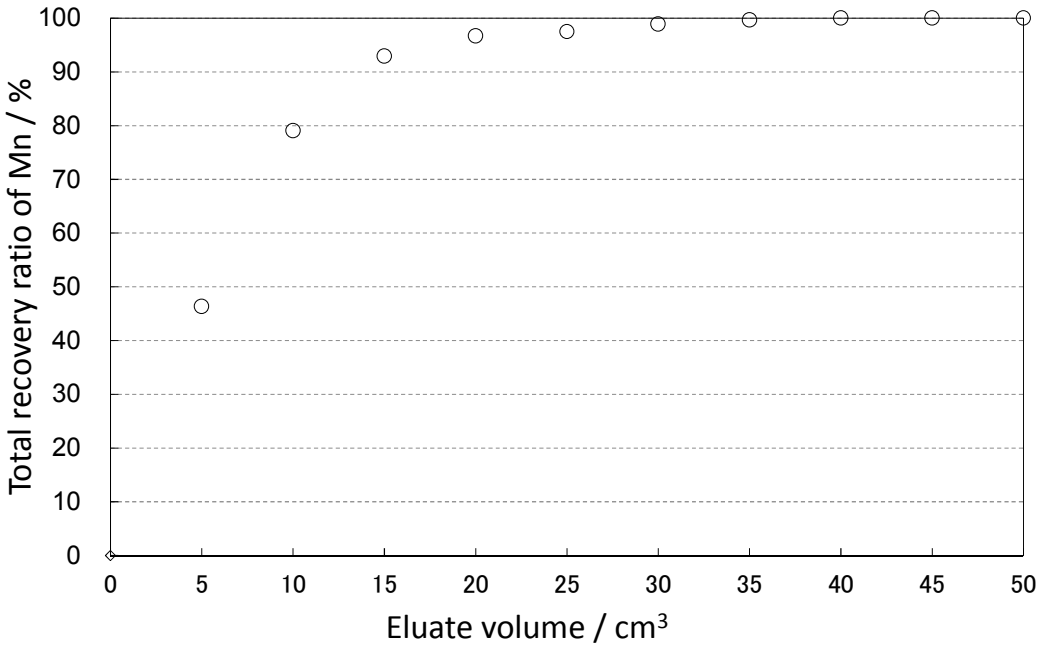

Fig. 4 Elution Pattern of 1mg Mn from the Column. Column was Dowex 1x8 100-200 mesh (5 cm^3) and eluate was 9 mol dm^{-3} HCl.

Table 2 Recovery ratio of Mn using the irradiated specimen

Mn in solution /mg	Mn in eluate /mg	Recovery ratio /%
0.998	0.987	98.9

Eluate was 9 mol dm^{-3} HCl, and the amount was 35ml.

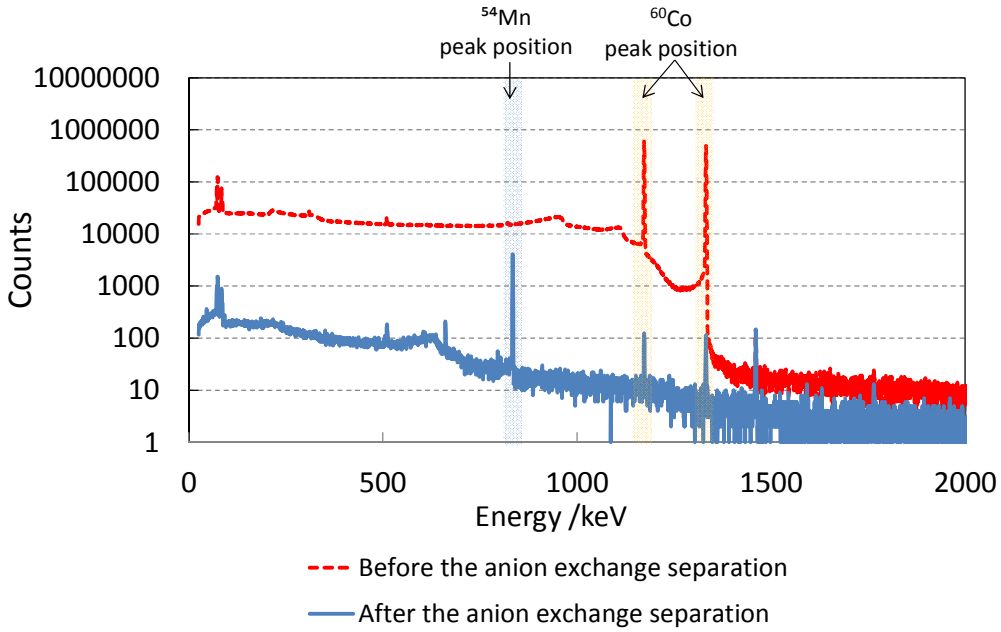

Fig. 5 γ-spectra of the specimen

Table 3 Amount of radioactivity of ^{60}Co in the specimen

Nuclide	^{60}Co
Before the anion exchange separation / Bq	1.0×10^{6}
After the anion exchange separation / Bq	6.3×10^{1}

4. Conclusion

Anion exchange separation for solutions containing irradiated steel specimen used to estimate neutron fluence was studied, and our anion exchange separation method was able to remove ^{60}Co with high recovery ratio for ^{54}Mn in the solution of an irradiated steel specimen. ^{60}Co was able to be reduced to less than 10^{-4} of the initial amount and 98.9 % of the ^{54}Mn recovery ratio was obtained. From this result, measurable period of ^{54}Mn in the iron dosimeters can be expected to be extended by several years with this method.

Reference

[1] E. Akatsu, Data of Ion Exchange, JAERI-M 7168, 1977.