ESTIMATION AND MEASUREMENT OF Fe-55 AND Ni-63 IN HARDWARE ACTIVATED AT HIGH ENERGY ACCELERATOR FACILITIES

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1,Introduction

At high energy accelerator facilities, accelerator components are exposed to primary and secondary particles with energy spectra from thermal to high energy,and as a result,they become radioactive through various types of nuclear reactions including spallation reactions[1, 2]. From a viewpoint of accelerator radiation protection, evaluation and measurement of radioactivities in these hardware is one of the most important things. Most radioisotopes produced are γ -emitters, and their radioactivities can be readily determined with a conventional radiation detector. While ^{55}Fe(EC decay,half-life:2.7y) and ^{63}Ni(β^- decay,half-life:100.1y) ,which are formed in accelerator components composed of iron and steel, emit no γ -ray, and from their nuclear properties, chemical separation of these radioisotopes from the irradiated samples is unavoidable prior to the measurement. These long-lived radioactivities accumulate in proportion to the machine operation time and become dominant in the residual activities several years after beam-off. These isotopes are conceivable to be produced abundantly because of the large cross-sections leading to their formation. Therefore,measurement and evaluation of these radioactivities are very important for safe handling of these accelerator components, and no report however is available on the measurement of ^{55}Fe and ^{63}Ni formed in the bulk hardware.

This paper is concerned with the development of measurement methods and the evaluation of ^{55}Fe and ^{63}Ni activities formed in iron and steel commonly used in accelerator hardware. The chemical separation of these radioisotopes from the irradiated materials was studied extensively. The ^{55}Fe and ^{63}Ni radioactivities were also estimated on the basis of the neutron fluxes measured by using a set of Au foils and Cu threshold detectors and compared with those experimentally obtained by a liquid scintillation counting method.

2,Experimental

In order to measure the integrated hadron fluxes(mostly neutrons),an activation method was adopted: a set of Au foils(about 1g) and Au foils wrapped with 1mm-thick Cd and copper discs(about 6g and/or 3.4g) were placed at various positions in the beam-line tunnel(EP2) of the National Laboratory for High Energy Physics(KEK) 12-GeV proton synchrotron. Iron and steel discs were placed at the same positions. Table 1 shows the cross-sections and effective threshold energies used for the calculation of integrated hadron fluxes in a copper threshold detector. The beam line tunnel is about 40m long, 6m wide and 4m high,being enclosed by heavy concrete blocks. The average intensity of primary protons was $(0.5\text{-}1.3)\times10^{12}$ protons per second, which was continuously monitored with SEC(secondary emission chamber). After the irradiation of a certain term,the radioactivities formed in the activation detectors and samples were measured with a high resolution Ge-detector connected to 4k PHA. To avoid the intereferences of short-lived radioisotopes,their radioactivities were measured about two weeks after the end of irradiation.

The ^{55}Fe and ^{63}Ni were measured with a liquid scintillation counter(LSC)(Packard Tri-Card 200CA/LL). Fig.1 shows the chemical separation procedures of iron and nickel from the irradiated discs and the preparation for the counting samples for LSC. 16mL of EX-H emulsfier was added to the iron or nickel solution thus separated in a glass counting vial of 20 mL and shaken vigorously. The milky gel-samples for LSC thus

prepared were allowed to stand for about one day and then measured several times for 30 min. A window of 0-70 keV was set to cover the entire energy region of β^--rays from ^{63}Ni(E_{max}=65.9keV), whereas for the measurement of ^{55}Fe radioactivity, three window widths of 0-7 keV,7-1200 keV and 0-1200 keV were set to measure the X-rays of 5.8 keV(K_α) and 6.4 keV(K_β) and Auger electrons of 5.2-5.8 keV. The extent of quenching was determined by the external standard method using a ^{137}Cs source.

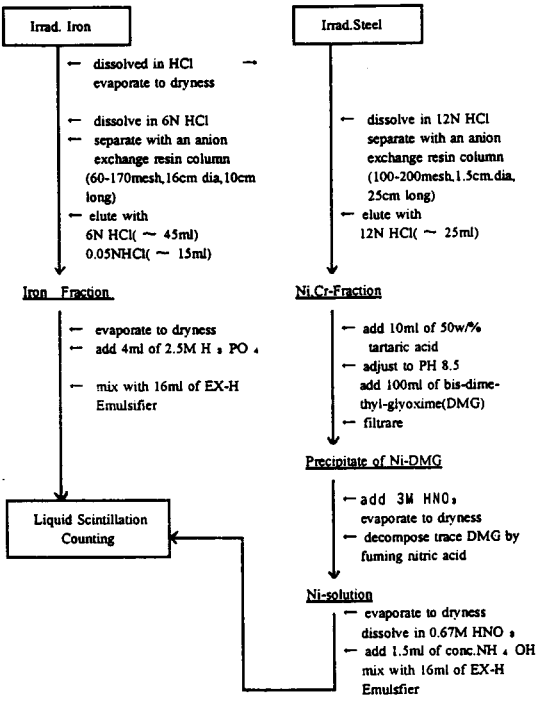

Fig.1 Procedures for chemical separation of ^{55}Fe and ^{63}Ni from irradiated iron and steel samples and preparation of counting samples for LSC

3, Results and Discussion

The chemical yields for the whole procedures shown in Fig.1 were about 91% for ^{55}Fe and 94% for ^{63}Ni. Both γ-ray and liquid scintillation spectrometries showed no contamination in the samples thus prepared. The counting sample preparations for LSC were simple and applicable to the ^{55}Fe and ^{63}Ni measurement with high sensitivity. For iron and steel samples. the detection limits were 0.82 Bq/g(Fe) and 0.035 Bq/g(SUS,Ni content of 15%) by assuming the counting time of 100 min. and 95% confident level(2 σ).

Thermal neutron fluxes were $(2.0\text{-}5.2)\times10^5 cm^{-2}s^{-1}$, and almost the same at any places in the EP2 tunnel. ^{63}Ni is primarily produced in steel discs through thermal neutron capture reactions of ^{62}Ni with natural abundance of 3.66%; its cross-section is 14.6 b. The steel samples used were all SUS discs with nickel content of 15%. As Table 2 shows, the ^{63}Ni radioactivities calculated from the thermal neutron fluxes were relatively good agreement with those obtained experimentally. On the other hand,the ^{55}Fe mainly formed through the thermal neutron capture of ^{54}Fe and (n,2n) reaction of ^{56}Fe, which have large cross-sections of 2.25 b and about 600 mb for the neutrons of thermal and 18 MeV, respectively. In order to calculate the ^{55}Fe activity, the cross-sections for the (n,2n) reaction were taken from ENDF/HE-VI library. Fig.2 shows the relation between the radioactivities calculated and exprimentally obtained at six different positions. The calculated values for the two positions downstream from beam-loss points and close to the beam line were considerably overestimated, indicating that the (p,pn) reaction of ^{56}Fe is

Table 1. Cross sections and effective threshold energies for calculating the integrated hadron fluxes using a Cu threshold detector

Nuclide	Threshold * energy(MeV)	Cross-section ** (mb)	Half-life
^{60}Co	13	7.8	5.27y
^{58}Co	23	18	70.9d
^{56}Co	35	3.7	77.1d
^{54}Mn	50	13	312d
^{48}V	72	5.5	16.0d
^{46}Sc	85	4.7	83.8d

*Routti(1974) [3] , **Asano et al.(1983) [4] ,

considered to contribute somewhat the ^{55}Fe formation at the positions close to the beam line. This disagreement can be explained in terms of coexisting high energy protons in the measured fluxes and the difference of the excitation functions of the (n,2n) and (p,pn) reactions. ^{55}Fe and ^{63}Ni survive for long time after beam-off and emit no γ-ray. Therefore no radiation level does not necessarily mean that all radioactivity had decayed out. From a viewpoint of radiation safety, it is very important to evaluate their radioactivities prior to an actual handling of radioactive accelerator components. The present study indicated that a rough estimation of ^{55}Fe and ^{63}Ni activities is possible by measuring the neutron fluxes by a set of Au foils and Cu threshold detectors.

Table 2. ^{63}Ni radioactivity in SUS samples

	n_{th} ($cm^{-2} s^{-1}$)	^{63}Ni(Cal.) Bq/g(SUS)	^{63}Ni(Exp.) Bq/g(SUS)
Sample-1	2.2×10^{5}	2.0 ± 0.6	3.7
Sample-2	2.0×10^{5}	2.2 ± 0.7	3.7
Sample-3	3.0×10^{5}	2.7 ± 0.7	2.1
Sample-4	5.2×10^{5}	3.2 ± 1.1	1.4
Sample-5	4.0×10^{5}	2.0 ± 0.6	2.7

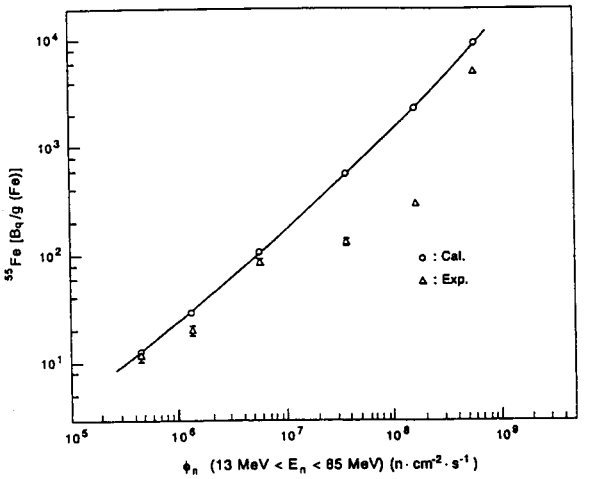

Fig.2 Relation between ^{55} Fe activity [Bq/g(Fe)] and neutron fluxes (Φ n;13MeV < En < 85MeV,n/cm^2 /s)

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5,References

1, M.Numajiri,T.Miura,K.Kondo,T.Suzuki,Y.Oki,M.Takasaki,K.H.Tanaka,M.Ieiri, Appl.Radiat.Isot.,42,577-581(1991).

2,Y.Oki,M.Numajiri,T.Suzuki,T.Miura,K.Kondo, Appl.Radiat.Isot.,43,1355-1362(1992).

3,J.T.Routti, Phys.Scripta 10,107-110(1974).

4,T.Asano,Y.Asano,Y.Iguchi,H.Kudo,S.Mori,M.Noguchi,Y.Takada,H.Hirabayashi,H.Ikeda,K.Katoh,K.Kondo, M.Takasaki,T.Tominaka,A.Yamamoto, Phys.Rev. C28,1718-1724(1983).