Nickel-63 Radioactivity in Steel and Copper Activated at High Energy accelerator Facilities

Masaharu Numajiri, Yuichi Oki, Takenori Suzuki,
Taichi Miura, Masabumi Taira, Yukio Kanda and Kenjiro Kondo

Radiation Safety Control Center, National Laboratory
for High Energy Physics,
Oho 1-1, Tsukuba-shi, Ibaraki-ken, Japan 305,

Abstract

^{63}Ni(β^- decay, half-life; 100.1 y) is abundantly produced in hardware composed of SUS and copper at high energy accelerators. The measurement method of its activity by a liquid scintillation counter was developed. Its rough activity could be estimated on the basis of thermal and fast neutron fluxes measured by an activation method using gold and copper activation monitors. After a few tens of years from beam-off, no radiation level on these accelerator hardware does not necessarily mean that all activity had decayed out, there is a possibility that an appreciable amount of ^{63}Ni still presents in them.

I. INTRODUCTION

At high energy accelerator facilities, accelerator components (e.g., vacuum pipe, flange, valve, bellows, collimator, beam-stop, etc.) are exposed to high energy primary and/or secondary particles and become radioactive. Most of these radioactive nuclides are γ-ray emitters, mainly contributing to an external dose. Whereas there are several isotopes, which are produced in a large quantity and decay via electron capture or emission of β ray with extremely low energy. ^{55}Fe(EC decay, $t_{1/2}$=2.7y) and ^{63}Ni(β^- decay, $t_{1/2}$=100.1y) are representive isotopes among them. From their decay properties, it is practically impossible to estimate their activities by direct measurements.

For ^{55}Fe, the measurement method of its radioactivity has been studied extensively for iron and copper activated at high energy accelerator facilities, and it has been also pointed out that its activity was quantitatively evaluated from hadron fluxes experimentally measured[1]. There are few papers on the measurement methods of ^{63}Ni activity in sludge of reactor cooling-water[2] and environment samples[3], however no report concerning evaluation and measurement methods of ^{63}Ni radioactivity produced in accelerator components is available.

^{63}Ni is produced mainly by the ^{62}Ni(n,r) and ^{63}Cu(n,p) reactions; The thermal-neutron capture reaction has a cross-section of 14.2 b, and the maximum cross-section for the latter reaction is 133 mb at 10 MeV. Flanges, pipes for cooling water, beam- ducts, and so on, are usually made of stainless steel, which usually contains nickel of 10 to 20 % in weight. Natural abundances of ^{62}Ni and ^{63}Cu are 3.66% and 69.1%. Since ^{63}Ni is the longest-lived isotope with a half-life of 100.1 years, its radioactivity is accumulated in proportion to machine operation time, and it becomes one of principal isotopes after several years' operation.

This paper is concerned with the development of measurement methods and evaluation of ^{63}Ni activity formed in copper and SUS samples, which are commonly used in accelerator hardware at high energy accelerator facilities. The chemical separation of ^{63}Ni from the above materials was extensively studied, and its activity was measured by a liquid scintillation counting(LSC) method with a high efficiency. ^{63}Ni activity was evaluated on the basis of cross-section data and thermal and high energy neutron fluxes obtained using an activation detector. The calculated activities were compared with those obtained experimentally.

II. EXPERIMENTAL

A. Measurement of thermal and high energy neutron fluxes

In order to measure the hadron fluxes (mostly neutrons), an activation method was adopted; a set of Au foil and Au foil wrapped with Cd of 1mm thick and copper discs were placed at various positions in the beam-line tunnel(EP2) of the National Laboratory for High Energy Physics(KEK) 12 GeV proton synchrotron. The monitors were irradiated for about two weeks, and their radioactivities were measured a few days later from the end of irradiation with a high resolution Ge detector connected to 4k PHA.

B. Preparation of ^{63}Ni standard counting samples for LSC

The ^{63}Ni standard samples for liquid scintillation counting (LSC) were prepared by using the ^{63}Ni standard solution of 0.1N HCl(^{63}Ni activity; 1.676kBq/ml, overall uncertainty of activity:3%). 1.5 ml of conc.NH_4OH was added to the nickel solution of 0.67M HNO_3, and Ni

ions were changed to purple nickel amine complex. Then 16 ml of EX-H emulsifier as a scintillation cocktail was added to the solution. The gel-samples thus prepared were measured 5 times for 30 min by a scintillation counter. The quenching effect was examined from the relations of counting rate vs Ni concentration.

C. Chemical separation of ^{63}Ni from SUS and copper

SUS and copper were dissolved by an acid solution. Ni was first separated by an ion exchange method,and the pure Ni solution was obtained through the formation of Ni-bis-dimethylglyoxime complex. The chemical yield for nickel was over 94%. The successive procedure for the counting samples for LSC is the same as described above. The counting efficiency for the samples thus prepared was determined from the nickel concentration in the sample by using the quench curve prepared in advance.

III. RESULTS AND DISCUSSION

A. Estimation of ^{63}Ni activity

Thermal neutron fluxes were measured at several positions of sample 1-3 and 5 in EP2. As Table 1 and 2 show, the fluxes were (2.0-4.8)x10^5cm^{-2}s^{-1} for an average intensity of primary protons of (0.4-1.1)x10^{12}, and almost same at any places in the EP2 beam-line tunnel within a factor of 3. A value for the average thermal neutron flux from a source in an enclosure(n_{th}) can be estimated by the following equation[4]:

$$n_{th} = 1.25xQ/S$$

where Q is the source strength(s^{-1}) and S the surface area of the beam-line tunnel.

Q corresponds to a beam spill of 1.1x$10^{12}s^{-1}$ in the tunnel. The surface area of the tunnel is about 770 m^2. This leads to an average thermal neutron flux 1.8x10^5 cm$^{-2}s^{-1}$. This value was approximately same within a factor of 3 as those obtained experimentally.

^{63}Ni is primarily produced through thermal-neutron capture reactions. The samples used for the chemical separation are all SUS bolts, and the sampling positions, irradiation conditions, and their nickel contents were indicated at the footnote of Table 1and 2. The ^{63}Ni radioactivities calculated from n_{th} values showed relatively good agreement with those obtained experimentally.

Table 2 shows the integrated hadron(mostly neutron) fluxes obtained experimentally at the positions for sample-4 and 5. ^{63}Ni from copper is produced mainly through the high energy neutron reaction of $^{63}Cu(n,p)$ in the energy region of 5 to 20 MeV. This reaction has the highest cross-sections of 133 mb at 10MeV. A copper threshold detector does not cover the energy region below 13 MeV as shown in Table 2.

In order to make a rough estimation of the integrated neutron flux in the energy region less than 13 MeV, the neutron spectrum($\Phi_n(E)$) at the energy region above an evaporation peak was assumed to be simply expressed in a smooth curve of $\Phi(E)=aE^b$. a and b were calculated to be 1.8x10^5cm$^{-2}s^{-1}$ and-1.7 for sample-4, and 1.4 x10^6cm$^{-2}s^{-1}$ and -1.7 for sample-5. The excitation function for the above (n,p) reaction up to 20MeV is tabulated in a Ref.5, however no data is available for the energy region above 20 MeV. For the rough calculation of ^{63}Ni radioactivity, it was assumed that the cross-sections at above 20MeV decreases linearly with a neutron energy to 30 mb at 100MeV. The contribution from this energy region is however considered to be less than 15 % of the total activity. Table 2 shows the ^{63}Ni radioactivities thus calculated, together with those obtained experimentally. From these results, it was indicated that a rough estimation of ^{63}Ni radioactivity, produced in SUS and copper at high energy accelerator facilities, is possible by using gold and copper activation detectors.

Many radioisotopes other than ^{63}Ni are also produced in the samples, through various nuclear reactions. The saturation activities of principal radioisotopes($t_{1/2}$>70 days) and their time variations after the end of irradiation were calculated, and Fig.1 shows their time variation for SUS sample. ^{55}Fe is the most abundant isotope in the cooling time less than 10^4days,but ^{63}Ni is only one radioisotope about 25 years later. On the other hand,the most abundant isotope in Cu is ^{63}Ni in the entire cooling time,and its radioactivity is about 100 times that for SUS. From the figure, ^{63}Ni radioactivity may be estimated roughly on the basis of the radioactivity of γ-emitters at a certain cooling-time, if the irradiation conditions are known.

B. ^{63}Ni activity measurement by LSC

Fig.2 shows the dependency of counting efficiency on the nickel concentration in LSC. The maximum value of FOM was obtained at about 200 mg of nickel.

The background was 17.1 cpm for the samples containing 150 mg of Ni. The detection limit of ^{63}Ni was 0.035 Bq/g(SUS,Ni content;15%) by assuming the counting for 100 min and 95 % confidence level. This sensitivity was good enough for the purpose of measurements of ^{63}Ni for a radiation safety

IV. REFERENCES

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Table 1 ^{63}Ni radioactivity in SUS samples

*1)	n_{th} ($cm^{-2}s^{-1}$)	^{63}Ni(Cal.) Bq/g(SUS)	^{63}Ni(Exp.) Bq/g(SUS)
Sample-1	2.0×10^{5}	2.7±0.98	3.7
Sample-2	5.2×10^{5}	3.6±1.1	1.4
Sample-3	4.0×10^{5}	2.0±0.59	2.7

*1, Sampling positions and irradiation conditions; All samples are SUS bolts used in the EP2 tunnel. Ni content; 15% for Sample-1, 9.3% for Sample-2, and 7.1% for Sample-3. An average primary-beam intensity was 1.1×10^{12} protons/s, and the samples were exposed to secondaries for about 2±0.6years.
Sample-1; a bolt attached to the EP2-ES 40cm apart from the beam axis. Sample-2; a bolt on the floor 70cm beneath the beam axis and about 5m downstream from the beam splitter. Sample-3; a bolt on the wall about 2.5m apart from the beam-line.

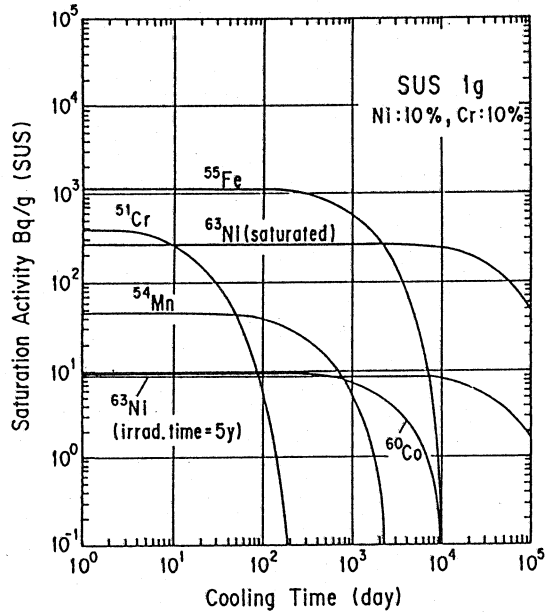

Fig.1 Saturation activities and time variations of the nuclides [Bq/g(SUS)] produced in SUS activated at the position for sample-5 in the high-energy proton beam-line tunnel(EP2).

Table 2 Integrated hadron fluxes and ^{63}Ni radioactivity produced in Cu

A) Integrated hadron fluxes.

		Integrated neutron fluxes($n/cm^2/s$)					
	n_{th}	>13MeV	>23MeV	>35MeV	>50MeV	>72MeV	>85MeV
Sample-4	---	3.3E4	1.8E4	9.7E3	5.5E3	1.2E3	6.3E2
Sample-5	4.8E5	2.3E5	1.3E5	6.5E4	2.5E4	1.1E4	5.0E3

E5 stands for 10^5.

B) ^{63}Ni radioactivity.

*1)	^{63}Ni(Cal.) Bq/g(Cu)	^{63}Ni(Exp.) Bq/g(Cu)
Sample-4(A)	0.74±0.20	0.69
Sample-4(B)		0.73
Sample-5	6.6±0.9	3.4

Sampling positions and irradiation conditions;
*1) Sample-4; a piece of a copper cooling pipe attached to the Q-magnet in the EP2-B line about 70m downstream from the beam splitter and 50cm apart from the beam axis. The beam line was operated at an average beam intensity of 1.2×10^{11}p/s, and the sample had been exposed to secondaries for 1.5±0.4years.
Sample-5; a piece of a copper cable attached to the Q-magnet in the EP2-C line about 2m upstream from K3 target(Pt, 1cm dia., 5cm long) and 55cm apart from the beam axis. An average beam intensity of this line was 0.4×10^{12}p/s and the sample was exposed to secondaries for 3±0.9years.

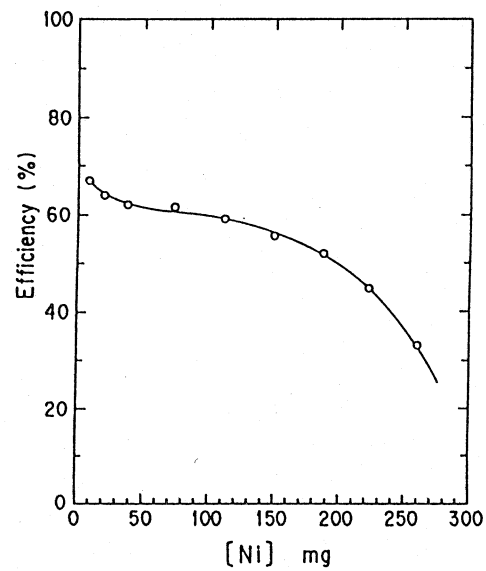

Fig.2 Dependence of counting efficiency on nickel concentrations in a liquid scintillation counting.