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Measurement of inclusive photonuclear (γ,n) reaction cross section for ^{129}I

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Abstract. Long-life non-γ emitting radioactive wastes such as ^{129}I and ^{14}C are produced from nuclear waste facilities. The photonuclear reaction is supposed to be usable for measuring the quantity of such nuclides without destruction of waste drum. Photonuclear products of ^{128}I and ^{13}C emit gamma rays and their radiation can be easily detected by a Ge detector. In this study, the inclusive photonuclear reaction cross section of ^{129}I(γ,n) ^{128}I was measured by the use of bremsstrahlung photons from 30-MeV electron LINAC. The amount of produced ^{128}I was measured with an HPGe gamma-ray spectrometer. Experiments were analyzed for both (γ,n) and accompanying (n,γ) reactions. Flux calculations were performed by EGS and MCNP codes. The measured average activation cross section of ^{129}I(γ,n) ^{128}I is 0.096 ± 0.005 barn on the basis of ^{197}Au(γ,n) reaction value. The measured value agrees with the evaluated (IAEA photonuclear data library) cross section within a deviation of 17%. The cross section is usable for non-destructive measurement of the amount of ^{129}I in radioactive waste.

1 Introduction

Huge amount of radioactive waste is produced from nuclear facilities. One of major problems in the nuclear engineering today is in the management and disposal of high-level radioactive waste. In many countries around the world much research efforts are being made in the possibility of partitioning and transmuting of radioactive waste, which reduces its toxicity by a factor of 100 [1–3]. Recently a number of roadmaps for nuclear waste transmutation have been proposed [4,5]. Frequently discussed methods involve transmutation by bombardment with neutrons from a nuclear reactor. Other approaches were proposed to this problem, for example laser-driven high-brightness gamma generation for photo-transmutation [6]. A UK report on the transmutation of nuclear waste [7] emphasized the importance of launching new ideas in radioactive waste management. Technique has recently been proposed for waste management by a particle accelerator [8,9].

Radioactive waste contains transuranic or fissile nuclides. They should be classified according to their radioactivity levels, so that they would be properly disposed. The waste generally includes a large amount of long-lived non-gamma emitting "difficult to measure" nuclides such as ^{129}I and ^{14}C. Measurement of the quantity of such radioactive nuclide has been carried out by a conventional method like the scaling factor method (SF) [9].

However, before applying the SF method, the correlation needs to be known between "easy-to-measure" nuclides that are readily measured non-destructively and "difficult-to-measure" ones such as pure β-radioactive nuclide. Since the correlation experiment is accompanied by the chemical analysis on "difficult-to-measure" nuclides, it takes a long time and much cost. We propose a new technique [9]: β-emitting long-lived radio nuclides in a waste drum are first converted into short-lived γ-emitting ones by high energy photons from an electron accelerator. After that, their radioactivity is measured non-destructively from the outside of the waste drum with a Ge spectrometer.

This work introduces the above mentioned new method and presents the measurement of the inclusive photonuclear reaction cross section of ^{129}I(γ,n) by using bremsstrahlung photons emitted from a target irradiated by 30-MeV electron LINAC. The experimental data are analyzed by the calculation with Monte Carlo codes EGS and MCNP.

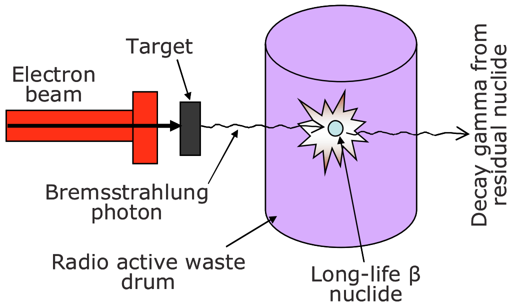

Fig. 1. Concept of non-destructive detection method using electron accelerator.

2 Non-destructive detection method

The principal of the "non-destructive detection" method is illustrated in figure 1. This method consists of an electron accelerator, a heavy-metal target like tantalum, a drum containing radioactive waste and a Ge gamma-ray detector.

An electron beam strikes on the heavy-metal target and generates high-energy bremsstrahlung photons. Produced photons react with the waste and convert long-lived β-nuclides in the waste drum into short-lived γ-emitter ones. The electron

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Table 1. Example of photonuclear reactions applicable to non-destructive detection.

			Giant region		Residual nuclide			
Nuclide	Half life (Year)	Reaction	Peak (MeV)	Estimated cross section σ(mb)	Nuclide	Half life	Decay mode	Major decay γ energy (keV)
C-14	5.73 E3	(γ,p)	25	10	B-13	17.4 ms	β^-	3680
Cl-36	3.01 E5	(γ,2n)	25	8	Cl-34 m	32.3 m	β^+	2128
Ni-59	7.50 E5	(γ,p)	20	30	Co-58	70.9 d	EC	811
Se-79	6.50 E4	(γ,p)	20	25	As-78	1.5 h	β^-	614 1309
Tc-99	2.13 E5	(γ,3n)	22	20	Tc-96 m	51.5 m	EC	778
I-129	1.57 E7	(γ,n)	15	300	I-128	25 m	β^-	443 969
Zr-93	1.53 E6	(γ,p)	22	20	Y-92	3.54 h	β^-	934 1405

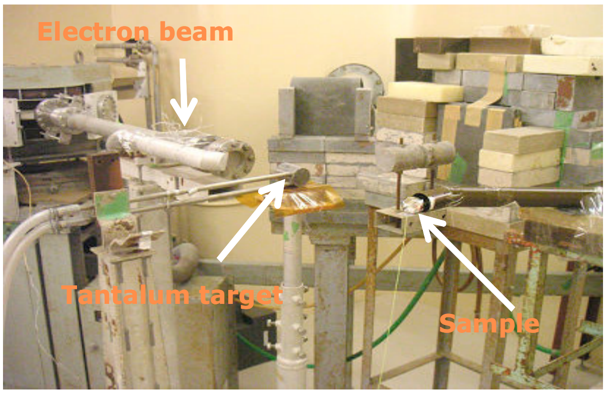

Fig. 2. Irradiation experiment layout.

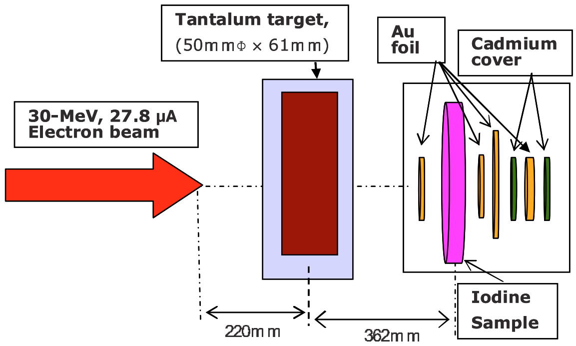

Fig. 3. Sample geometrical position in the experiment.

energies are determined so that the bremsstrahlung photon spectrum covers the giant dipole resonance region.

Typical long-lived nuclides to be concerned are listed [9] in table 1. Among these nuclides, ^{129}I and ^{14}C are of large amount in the nuclear waste. The photonuclear cross sections for these radioactive nuclei have not measured so far.

3 Experimental

The irradiation experiment was carried out for a radioactive iodine sample (radioactivity of 2.65 MBq.) by using a 30-MeV electron linear accelerator at kyoto University Research Reactor Institute. Figure 2 shows the experiment layout. Bremsstrahlung photons were generated from a thick tantalum target (50 mmϕ × 61mm) in front of the electron beam accelerator. Figure 3 shows the sample geometrical position in the experiment. The radioactive iodine sample was placed 33 cm apart from the target along the beam axis. The iodine sample (27.8 mmϕ × 2.8 mm) contains radioactive ^{129}I at 68% and stable ^{127}I at 32%. In addition to the sample gold foils were irradiated by the photons to measure photon and neutron fluxes at the sample position. The gold foils were placed in front and back of the iodine sample to obtain the photon flux in the sample position. One of the gold foils was covered by cadmium sheets to know the slow-neutron flux information.

Reactions in the iodine sample are expected to be

i) ^{129}I(γ,n)^{128}I
ii) ^{129}I(n,γ)^{130}I
iii) ^{127}I(n,γ)^{128}I.

The activation experiment of gold foils gives the photon and neutron fluxes through the reaction of

i) ^{197}Au(γ,n)^{196}Au
ii) ^{197}Au(n,γ)^{198}Au.

The cross sections on gold were taken from IAEA photonuclear data library [10].

4 Flux calculation

The measured photon flux data obtained from the gold irradiation experiment were checked by the calculated values by EGS code. Photon fluxes at the iodine sample position were determined by averaging fluxes on gold foils in front and back of the iodine. To know the contribution of ^{127}I(n,γ)^{128}I reaction to the yield of total ^{128}I, fast neutron flux was calculated by means of the Monte Carlo code MCNP. In addition, slow-neutron flux in the thermal and epithermal regions was derived from the experimental data of gold foils with and without cadmium.

The slow neutron flux was assumed to have a Maxwellian distribution with an amplitude and a temperature parameters. The two parameters were determined by the gold activation results with and without cadmium. The neutron transport through the cadmium sheet was taken into account.

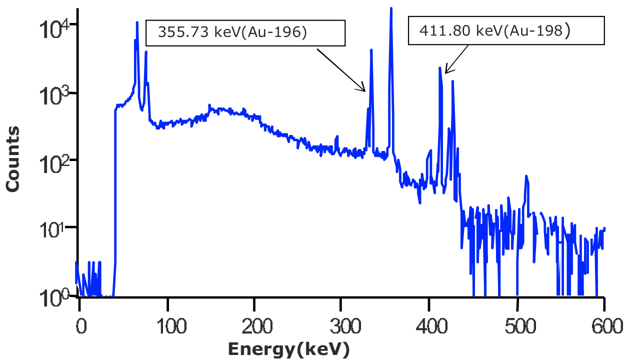

Fig. 4. Gamma spectrum of irradiated gold foil.

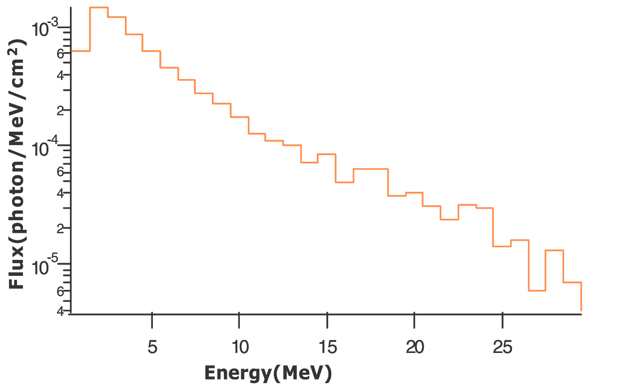

Fig. 5. Photon flux from 30-MeV incident electron using EGS5 code (number of incident electron = 1 million/batch.)

Table 2. Yields obtained from ^{197}Au foil irradiation.

Gold foil	Yields (Bq.) ^{196}Au	^{198}Au
Front Au	2.38×10^5	1.39×10^4
Back Au	2.31×10^5	1.19×10^4
Big Au (back)	1.99×10^6	1.32×10^5
Au + Cd	2.12×10^5	5.23×10^3

The temperature of slow neutron was given as 2.69×10^{-2} keV. The contribution of ^{127}I(n,γ)^{128}I was obtained with the slow neutron flux.

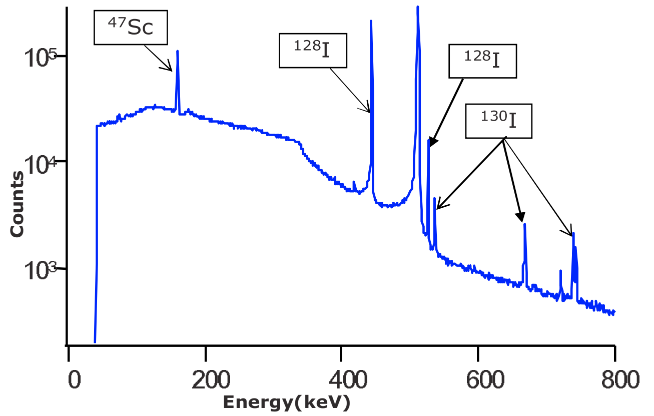

Fig. 6. Gamma spectrum of irradiated iodine (included ^{127}I) sample.

Table 3. Yields obtained from iodine irradiation.

Nuclide	Yields (Bq.) Measured total	^{129}I(γ,n)^{128}I	^{127}I(n,γ)^{128}I
^{128}I	5.08×10^6 (±5.5%)	4.99×10^6	8.87×10^4

Table 4. Activation cross section of ^{129}I averaged over bremsstrahlung photon spectrum.

Average reaction cross section Measured (barn)	Evaluated (IAEA Data library) (barn)	Ratio (Measured/Evaluated)
0.096 (±0.005)	0.116	0.83 ± 0.04

5 Results and discussion

5.1 Results from gold irradiation experiment

Figure 5 shows the example of γ spectrum from irradiated gold. The peak at 355.73 keV originates from the decaying isotope ^{196}Au through ^{197}Au(γ,n)^{196}Au reaction and that at 411.80 keV from ^{198}Au through ^{197}Au (n,γ)^{198}Au. The yields obtained from the irradiation are listed in table 2. The photon flux was calculated by EGS code. Figure 4 shows the example of photon spectrum. The peak energies of giant dipole resonance are 15 and 13.5 MeV for iodine and gold, respectively. For ^{197}Au(γ,n)^{196}Au reaction, the intensity of calculated average flux deviated by 25% from the experimental one. The calculated value was factored to reproduce the experimental gold foil data.

5.2 Results from iodine irradiation experiment

Figure 6 shows the γ spectrum of irradiated iodine sample. The peaks at 442.9 and 526 keV originate from the decaying isotope ^{128}I through ^{129}I(γ,n)^{128}I reaction and those at 536, 668.5 and 739.5 keV from ^{130}I through ^{129}I(n,γ)^{130}I. Since the iodine sample includes ^{127}I at 32%, ^{127}I (n,γ) reaction may occur simalteniously togater with ^{129}I(γ,n) reaction.

Therefore the contribution of ^{127}I(n,γ) reaction to production of ^{128}I was estimated on the basis of the preceding neutron flux and given in table 3. The yields obtained from the irradiation of ^{129}I are listed in table 3. The contribution of (n,γ) reaction is as small as 1.8% due to minimized use of cooling water for the target.

The average cross sections of ^{129}I(γ,n)^{128}I reaction are shown in table 4. From the experiment, the cross section is 0.096 ± 0.005 barn, whereas from IAEA evaluated photonuclear data library [10] gives an average cross section of 0.116 barn. The cross section ratio of (measured/evaluated) is 83%. The evaluated value shows an agreement with the experimental one with a deviation of 17%. The result is applicable to non-destructive measurement of ^{129}I in a radioactive waste by bremsstrahlung photon irradiation.

6 Conclusions

Non-destructive detection of long-lived *difficult-to-measure* nuclides requires the photonuclear cross sections. The ^{129}I(γ,n)^{128}I reaction cross section has not been checked so far. We measured the integrated cross section of ^{129}I(γ,n) reaction by using bremsstrahlung photons.

The experimental flux was checked by the Monte Carlo codes of EGS and MCNP. The measurement needed information about the influence of (n,γ) reaction caused mainly by slow neutrons. We estimated the ^{127}I(n,γ) reaction rate in production of ^{128}I, and the result of photo neutron contribution was found to be an admissible level. The measured average ^{129}I(γ,n)^{128}I cross section gave a deviation of 17% from the evaluated value (IAEA evaluated photonuclear data library).

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