NUCLEAR MEASUREMENT ACTIVITIES OF PNC FOR TRANSMUTATION OF FISSION PRODUCTS

Hideo Harada, Shoji Nakamura, Yoshiaki Shigetome and Toshio Katoh

Power Reactor and Nuclear Fuel Development Corporation, Tokai works, Tokai-mura, Ibaraki-ken, 319-11 JAPAN

Abstract. This paper reviews the PNC's activity on nuclear data measurements for the study of the transmutation of fission products. Thermal neutron capture cross sections and resonance integrals of ^{137}Cs, ^{90}Sr, ^{99}Tc, and ^{129}I were measured for the study of the transmutation method using thermal neutrons. High resolution measurements of photonuclear reaction cross section are also in progress for the study of the transmutation method using monocromatic photons.

INTRODUCTION

To investigate the system transmuting fission products(FP), accurate nuclear reaction cross sections are required for FP. What kind of nuclear reactions should be used for FP transmutation depends on how large is the cross section utilized. For the study of the transmutation methods using a high flux fission reactor, a high intensity proton accelerator, and also a high flux fusion reactor, thermal neutron cross sections(σ_0) and resonance integrals (I_0) are especially important. However, these data are old and sometimes reported values differ each other. Therefore, we have measured these cross sections first for ^{137}Cs, ^{90}Sr, ^{99}Tc, and ^{129}I. As a results, we have found out that the σ_0 and I_0 of ^{90}Sr and ^{137}Cs are too small to be transmuted by using thermal neutrons.

As an alternative method transmuting ^{90}Sr and ^{137}Cs, we started the study of a monochromatic photon method; a high energy photon can transmute a nucleus via photonuclear reaction. If there is a sharp and intense resonance peak in the photonuclear reaction cross section, the monochromatic photon of the same energy with the resonance can effectively transmute a nucleus. However, very little is known about the precise photon energy dependence of the photonuclear reaction cross section. To obtain the intrinsic resonance peak width and intensity, we proposed a new experimental method.

The results of the experiments and the simulations are briefly described below.

THERMAL NEUTRON CAPTURE CROSS SECTIONS AND RESONANCE INTEGRALS

Accurate data of neutron cross sections are required for the research of the transmutation method of nuclear waste. Data reported, however, show discrepancies between them. Therefore, we designed experiments to obtain more accurate data of neutron cross sections of important long-lived fission products, ^{137}Cs[1,2], ^{90}Sr[3], ^{99}Tc[4] and ^{129}I[5]. Standardized solution of each radioactive nuclide was irradiated at a thermal neutron reactor of Japan Atomic Energy Research Institute(JAERI) or Rikkyo University. Samples were irradiated with or without a Cd shield. The number of nuclei in the target to be irradiated was determined by an isotope ratio method(IRM) or an efficiency tracing technique(ETT) which meets the nuclear property of each nuclide. The amount of reaction products, ^{138}Cs, ^{91}Sr, ^{100}Tc and ^{130}I, by neutron capture were obtained by measuring gamma-ray intensities from these nuclei. The reaction rates obtained were analyzed, and thermal neutron cross sections and resonance integrals were deduced. The results were compared with the data reported by others[6-13]. Some data of cross sections were revised from the data by others and some data of resonance integrals were newly obtained[1-5]. Measurements of neutron capture cross section of ^{135}Cs are in progress.

The targets of ^{137}Cs and ^{90}Sr were irradiated at the swimming pool type reactor JRR-4 of JAERI with or without a Cd shield by using a pneumatic tube system. The reactor has a 1/E neutron spectrum in the resonance neutron region. The irradiation position was characterized as having a thermal neutron spectrum of 4×10^{13} n/(cm^2 • s) and an epithermal index in the Westcott's convention[14] of 0.03. After the irradiation, a chemical procedure for purification of the irradiated target was carried out[2,3]. The impurity activities such as ^{24}Na and ^{38}Cl were removed from the target. The targets of ^{99}Tc and ^{129}I were irradiated in the rotary specimen rack(RSR) of the TRIGA MARK II reactor of Rikkyo University. The

thermal neutron flux of this irradiation position was 5 x 10^{11} n/(cm^2 · s) and the epithermal index 0.03. The chemical procedure was not applied to the targets of ^{99}Tc and ^{129}I since half-lives of these are short.

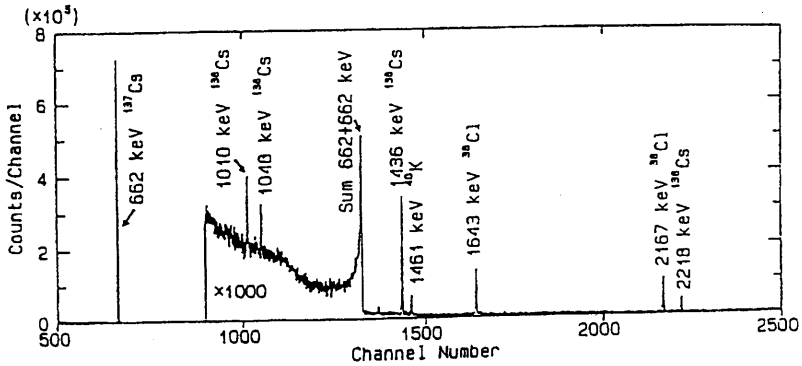

Fig.1 Gamma-ray spectrum obtained from neutron-irradiated and chemically purified ^{137}Cs sample in 10-min measurement

Table I. Results of the neutron capture cross section and resonance integrals

Nuclides	present results(b)	previous data(b)	Authors
^{137}Cs	$\sigma_0 = 0.25 \pm 0.02$[2] $I_0 = 0.36 \pm 0.07$[2]	$\sigma_0 = 0.110 \pm 0.033$	Stupegia[6]
^{90}Sr	$\sigma_0 = 0.0153 \begin{matrix} +0.0013 \\ -0.0042 \end{matrix}$[3] $I_0 \leqq 0.16$[3]	$\sigma_0 =$ 0.0140 ± 0.0024 0.8 ± 0.5	McVey et al.[7] Zeisel[8]
^{99}Tc	$\sigma_0 = 22.9 \pm 1.3$[2] $I_0 = 398 \pm 38$[2]	$\sigma_0 = 20 \pm 2$ $I'_0 = 186 \pm 16$	Lucas[9] Lucas[9]
^{129}I	$\sigma_0^{2+} = 17.4 \pm 1.7$[2] $I_0^{2+} = 17.8 \pm 2.0$[2] $\sigma_0^{5+} = 12.6 \pm 2.2$[2] $I_0^{5+} = 15.5 \pm 2.9$[2] σ_0 (total)= 30.0 ± 1.4 I_0(total)= 33.2 ± 1.5	$\sigma_0 = 26.7 \pm 2.0$ 31 ± 4 $I_0 = 36.0 \pm 4.0$	Roy[10] Block[12] Roy[10]

The radioactivity of the irradiated targets were measured by using a high purity Ge-detector system of 90% relative efficiency combined with a fast data acquisition system. For example, weak gamma-rays from ^{138}Cs were measured together with strong gamma-

rays from ^{137}Cs simultaneously(see Fig. 1). Details of the experiments and the analysis were published in ref. [1-5]. Results obtained are summarized in Table I.

The results show that the cross section of ^{137}Cs obtained is about twice of the previous one[6] and an experimental data of the resonance integral was newly obtained. Our results for ^{137}Cs are more reliable than that by Stupegia[6], since our method(IRM) could remove various sources of uncertainty as mentioned in ref. [1-3]. The cross section obtained for ^{90}Sr is in agreement with the value by McVey et al.[7] but not with that by Zeisel[8]. The resonance integral of ^{90}Sr was also obtained. The cross section of ^{99}Tc is almost equal to the previous data[9], and an experimental resonance integral is about twice of the value reported previously. The cross sections of ^{129}I were obtained for formation of the ground state and the isomeric state of ^{130}I separately. Previous data[10-13] show only a cross section of formation of the ground states. The resonance integrals were also obtained for formation of two states ^{130}I separately.

FINE STRUCTURE OF PHOTONUCLEAR REACTION CROSS SECTION

Fine structure of photonuclear reaction cross section in the giant resonance(GR) region can provide important information about the excitation mechanism of the GR and also for the study of the nuclear transmutation process using monochromatic photons[15]. In particular, the intrinsic width of fine peak in the GR is important because it determines the peak value of the cross section. However, very little is known about the width at present. The energy resolution for the cross section measurement using tagged photons[16,17] or monochromatic photons[18] was typically 100-500keV. Therefore, peak widths observed for some nuclei were almost determined by the experimental resolution. To obtain the intrinsic peak width, we proposed an experimental method that measures transmitted photons using a high resolution and high energy photon spectrometer(HHS)[19].

Figure 2 shows the conceptual setup of the experiment. The incident white photons should cover the energy range of interest. The photons are obtained by electron induced bremsstrahlung or more effectively by laser Compton scattering[20,21]. The transmitted photons from a thick target is measured by the HHS composed of two large germanium(Ge) detectors surrounded by a $Bi_4Ge_3O_{16}$(BGO) anti-coincidence spectrometer. The energy resolution of the experiment in the GR region should be improved to 10-20keV because the Ge detector has an energy resolution of about 0.1%[22] for high energy photons. The design of the HHS is shown in Fig.3. To obtain the large photopeak efficiency for high energy photons, two large N-type Ge detectors (relative efficiency of each crystal was 90% at 1.33MeV) were arranged like twins along a beam axis. To improve their peak/background (=total-peak) ratio, the twins were surrounded by thick BGO crystals that were used as an anti-coincidence spectrometer. Figure 4 shows the response function of the HHS for a collimated 15MeV photon beam of 2cmϕ. This was the result simulated by a Monte Carlo electron-gamma-shower code EGS4[23]. The response functions of the single Ge detector and the twin Ge detectors are also shown in the figure. The photopeak counts in the three spectra were normalized to be the same number, and the spectra were broaden with the energy resolution($\Delta E\gamma/E\gamma)10^{-3}$ of the Ge detector.

To simulate the photonuclear cross section measurement with the HHS, the flux distribution of transmitted photons from a thick water target was calculated as an example. The size of the target was 5cm in diameter and 60cm in length. The density of the water

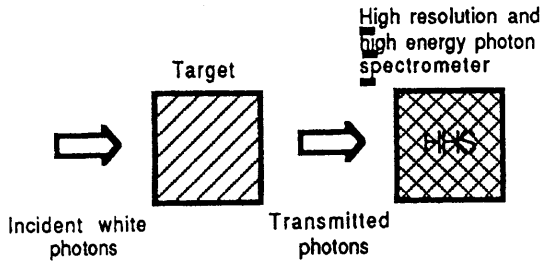

Fig.2 Schematic setup of high resolution measurement of photonuclear cross section using HHS

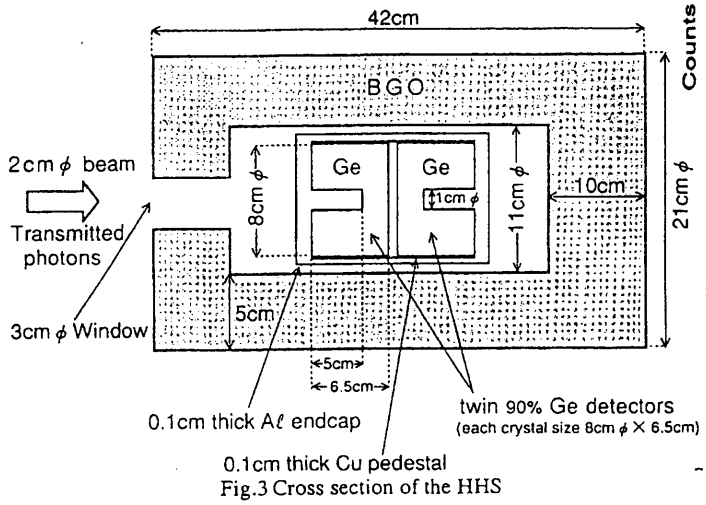

Fig.3 Cross section of the HHS

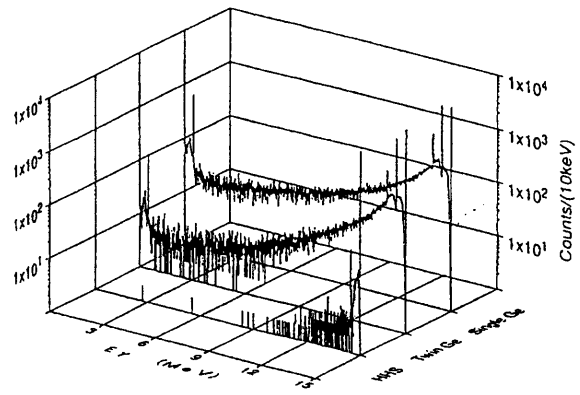

Fig.4 Response functions of three type spectrometer for 15 MeV photon

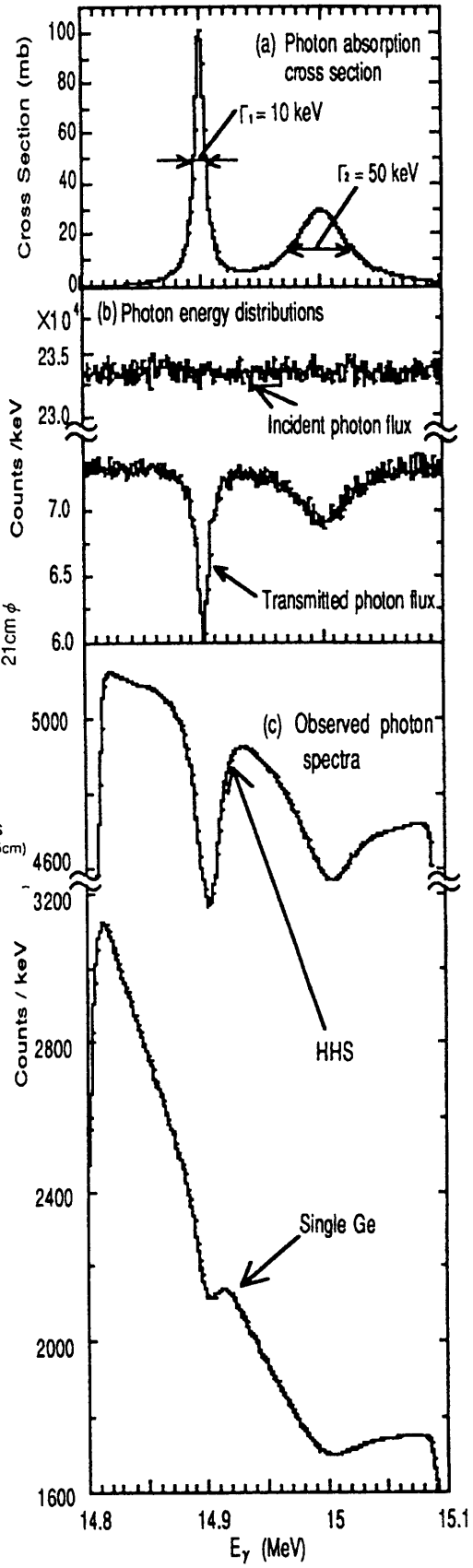

Fig.5 (a) Assumed photonuclear absorption cross section of ^{16}O,
(b) Incident white photon flux (upper) and transmitted photon flux (lower), and
(c) Transmission photon spectrum observed by
a single germanium detector(lower) and the HHS (upper)

target was assumed to be 1.0g/cm^3. The simulation code EGS4 was modified[24] to include the photonuclear cross section of ^{16}O as a part of photon-material interaction. Two narrow photon absorption cross section peaks of ^{16}O were artificially included in the code to simulate narrow dips in the transmission spectrum. The artificial cross section of the Breit-Wigner shape is shown in Fig.5(a). The width and height of the peak at 14.9MeV are 10keV and 100mb, and those at 15.0MeV were 50keV and 30mb, respectively. An incident photon flux was assumed to be white with an energy range of 14.8 to 15.1MeV. Figure 5(b) shows the incident white photon flux and the transmitted photon flux. Two dips can be clearly seen in the transmitted photon flux.

Figure 5(c) shows the transmission spectra observed by the single Ge detector(lower spectrum) and the HHS (upper spectrum). These were obtained by folding the transmitted photon flux(Fig.5(b)) using the response function of each detector. The dips are not clear in the spectrum observed by the single Ge detector because of its low peak/background ratio. On the other hand, the dips are clearly shown in the spectrum observed by the HHS. The FWHM of the dip observed at 14.9MeV is about 20keV. Therefore, widths of fine peaks in the GR region can be measured with the energy resolution of 10^{-3} by observing transmitted photons using the HHS, and correcting the detector energy resolution . Details of the simulation were described in ref. [19].

Fine structure of the photonuclear reaction in the GR energy region was shown to be observable using the specially designed high resolution and high energy photon spectrometer, HHS with an energy resolution of 10-20keV. The measurements using the HHS are in progress to supply data for nuclear transmutation studies using monochromatic photons.

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References

[1] HARADA, H., WATANABE, H., SEKINE, T., HATSUKAWA, Y., KOBAYASHI, K., KATOH, T.: *J.Nucl. Sci. Technol.*, **27** (1990) pp.577.
[2] SEKINE, T., HATSUKAWA, Y., KOBAYASHI, K., HARADA, H., WATANABE, H., KATOH, T.:*J.Nucl. Sci. Technol.*, **30** (1993) pp.1099.
[3] HARADA, H., SEKINE, T., HATSUKAWA, Y., SHIGETA, N., KOBAYASHI, K., OHTSUKI, T., KATOH, T.: *J. Nucl. Sci. Technol.*, **31** (1994) pp.173.
[4] HARADA, H., NAKAMURA, S., KATOH, T., OGATA, Y.: *J. Nucl. Sci. Technol.*, **32** (1995) pp.395.
[5] NAKAMURA, S., HARADA, H., KATOH, T., OGATA, Y.: *J. Nucl. Sci. Technol.*, **33** (1996) pp.283-289.
[6] STUPEGIA, D.C.: *J. Nucl. Energy.*, **A12** (1960) pp.16.
[7] MCVEY, L.A.,BRODZINSKI, R.L.,TANNER, T.M.: *J. Radio. Chem.*, **76** (1983) pp.131.
[8] ZEISEL, G.: *Acta. Phys. Austr.*, **23** (1966) pp.223.
[9] LUCAS, M., HAGEMANN, R., NAUDET, R., RENSEN, C., CHEVALIER, C.: *IAEA Report*, **IAEA-TC-119/14** (1977) pp.407.

[10] ROY, J.C., WUSCHKE, D.: *Can. J. Chem.*, **36** (1958) pp.1424.
[11] EASTWOOD, T.A. et al.: *Proc. 2nd Int. Conf. On Peaceful Uses of Atomic Energy, 58 GENEVA*, **16** (1958) pp.54.
[12] BLOCK, R.C., SLAUGHTER, G.G., HARVEY, J.A.: *Nucl. Sci. Engin.*, **8** (1960) pp.112.
[13] PATTENDEN, N.J: *Nucl. Sci. Engin.*, **18** (1963) pp.371.
[14] WESTCOTT, C. et al.: *Proc. 2nd Int. Conf. On Peaceful Uses of Atomic Energy, 58 GENEVA*, **16** (1958) pp.70.
[15] SATOH. I: *Private communication*, (1993).
[16] SPRINGHAM, S.V., et al.: *Nucl. Phys.*, **A517** (1990) pp.93.
[17] ANNAND, J.R.M., et al.: *Phys. Rev. Lett.*, **71** (1993) pp.2703.
[18] BERMAN, B.L., FULTZ,S.C.: *Rev. Mod. Phys.*, **47** (1975) pp.713.
[19] HARADA, H. and SHIGETOME, Y.: *J. Nucl. Sci. Technol.*, **32** (1995) pp.1189-1191.
[20] KASAGI, J.: *Private communication*, (1994).
[21] OHGAKI, H., et al.: *Nucl. Instrum. Methods*, **A353** (1994) pp.384.
[22] CECIL, F.E., et al.: *ibid.*, **A234** (1985) pp.479.
[23] NELSON, W.R., et al: *Nucl. Instrum. Methods*, **A356** (1995) pp.362.
[24] KASE, T., et al.: *PNC Rep.* **TN8410 92-350**, pp.70-72 [in Japanese]; KASE, T.,: Dr. Thesis, Tohoku University, (1995), [in Japanese].