DAEHAN HWAHAK HWOEJEE
(Journal of the Korean Chemical Society)
Vol. 17, No. 1, 1973
Printed in Republic of Korea

$^{93}Nb(n,\alpha)^{90}Y$, $^{90}Zr(n,p)^{90}Y$, $^{93}Nb(n,\alpha)^{90m}Y$ 및 $^{90}Zr(n,p)^{90m}Y$ 반응의 평균 핵분열 중성자 반응 단면적

李 澈·任 永 昌·鄭 求 珣*·朴 惠 一**
원자력연구소 화학연구실
(1973, 1, 4 접수)

Average Fission Neutron Cross Section for $^{93}Nb(n,\alpha)^{90}Y$, $^{90}Zr(n,p)^{90}Y$, $^{93}Nb(n,\alpha)^{90m}Y$ and $^{90}Zr(n,p)^{90m}Y$ Reactions

Chul Lee, Yung Chang Yim, *Koo Soon Chung and ****Hae-Ill Bak**
Chemistry Division, Atomic Energy Research Institue, Seoul, Korea
(Received Jan. 4, 1973)

요 약. $^{93}Nb(n,\alpha)^{90}Y$, $^{90}Zr(n,p)^{90}Y$, $^{93}Nb(n,\alpha)^{90m}Y$ 및 $^{90}Zr(n,p)^{90m}Y$ 반응의 평균 핵분열 반응 단면적을 결정하였다. α-히드록시 이소부티르산을 용출제로한 양이온 교환 수지통을 사용하여 생성된 핵종을 정량적으로 분리하였다. ^{90m}Y 및 ^{90}Y의 절대측정은 γ-선 분광법 및 보정된 2π계측기로 각각 계측하였다. 본방법에 의하여 $^{93}Nb(n,\alpha)^{90}Y$, $^{90}Zr(n,p)^{90}Y$, $^{93}Nb(n,\alpha)^{90m}Y$ 및 $^{90}Zr(n,p)^{90m}Y$ 반응의 단면적을 측정한 바 각각 0.14±0.01mb, 0.83±0.02 mb, 0.018±0.002 mb 및 0.033±0.002 mb이었다. ^{90}Y 대신 ^{90m}Y를 사용하는 편이 니오브를 정량하기 위한 보다 좋은 분석방법이 될 수 있음을 알았으며 그 가능성에 대하여 검토하였다.

Abstract. The average fission neutron cross sections were determined for the following reactions, $^{93}Nb(n,\alpha)^{90}Y$, $^{90}Zr(n,p)^{90}Y$, $^{93}Nb(n,\alpha)^{90m}Y$ and $^{90}Zr(n,p)^{90m}Y$. The cation exchange column was used for the quantitative separation of the product nuclides using α-hydroxyisobutyric acid as the eluent. The absolute activites of ^{90m}Y and ^{90}Y were determined by the gamma ray spectrometry and a calibrated 2π gas flow counter, respectively. The cross sections of $^{93}Nb(n,\alpha)^{90}Y$, $^{90}Zr(n,p)^{90}Y$, $^{93}Nb(n,\alpha)^{90m}Y$ and $^{90}Zr(n,p)^{90m}Y$ reactions were found to be 0.14±0.01 mb, 0.83±0.02 mb, 0.018±0.002 mb and 0.033±0.002 mb, respectively. The possible use of ^{90m}Y instead of ^{90}Y was discussed as a better means for the determination of niobium.

1. Introduction

One of the most practical and interesting uses of the research reactor is as a source of neutrons for the activation analysis. The bulk of the published literatures on the subject pertains to the thermal neutron flux. There is the enough evidence to show that the reactor neutron spectrum extends to well over 20 Mev.[1] When one realizes that many of the fast neutron reactions have thresholds in the region of 1—10 Mev. and that the total unmoderated fission flux in a swimming pool reactor, like TRIGA MARK II, is comparable to the thermal flux, it becomes evident that some of the fast neutron reactions

* Present address; Department of Chemistry, Sogang University, P.O. Box 1142, Seoul, Korea
** Present address; Department of Nuclear Engineering, Seoul National University, Seoul, Korea

can be useful for activation analysis especially when the use of the short lived isotope formed by (n,γ) reaction requires special irradiation and counting facilities. Several authors[2,3] have investigated the average reactor cross section for $^{93}Nb(n,\alpha)^{90}Y$ reaction and applied this reaction for the activation analysis of niobium.

In the present investigation, the average cross section were determined for $^{93}Nb(n,\alpha)^{90m}Y$ as well as $^{93}Nb(n,\alpha)^{90}Y$. An attempt was also made to explore the possibility of utilizing ^{90m}Y instead of ^{90}Y as a better means for the determination of niobium. The average cross section were determined for $^{90}Zr(n,p)^{90m}Y$ and $^{90}Zr(n,p)^{90}Y$ since these reactions could interfere with the niobium determination owing to the production of the same nuclides.

2. Principle of the determination

Taking into considerations the energy distribution of the neutrons used for the irradiation and the dependence of the cross sections on the neutron energy, the rate of the reaction is given[4] by

$$\int_{E_t}^{\infty} \sigma(E) \cdot \phi(E) \cdot dE$$

where E_t is the threshold energy of the reaction, $\sigma(E)$, the differential cross section and $\phi(E)$, the neutron flux of energy E in the spectrum. From the reaction rate, an average cross section, $\bar{\sigma}$, can be calculated from the following relation[4],

$$\bar{\sigma}=\frac{\int_{Et}^{\infty}\sigma(E)\cdot\phi(E)\cdot dE}{\int_{0}^{\infty}\phi(E)\cdot dE}=\frac{\int_{0}^{\infty}\sigma(E)\cdot\phi(E)\cdot dE}{\int_{0}^{\infty}\phi(E)\cdot dE}$$

provided the intergral flux is known. Since there is sufficient evidence to show that the distribution of reactor flux above 1 Mev. is similar to pure fission flux[5,6], a monitor like $^{58}Ni(n,p)^{58}Co$ with a known value of its average cross section in a fission flux can be used to determine the integral equivalent fission flux. In the present study nickel was used as a monitor for neutron flux and 93 mb were taken as the average cross section for $^{58}Ni(n,p)^{58}Co$ reaction[6].

3. Nuclear Data

The nuclear data relevant to the present investigation are shown in *Table* 1.

4. Experimental

4.1. Irradiation of the sample. 40 mg each of specpure niobium and zirconium, Alfa Inorganic, Ventron, were weighed and sealed in separate polyethylene vials of ca. 1 ml capacity. 50 mg of nickel metal sponge, 99.999 %, Ishizu Pharmaceutical Co., Japan, were weighed in a polyethylene tube of 3 mm —i. d. and sealed. The samples were wrapped

Table 1. Related Nuclear Data

Target	Abundance (%)	Reaction	Isotope formed	Half-life (hour)	Energy (Mev.) and intensity of radiation (%)
^{93}Nb	100	(n,α)	^{90}Y	64.0	β^-: 2.27 (99.7)
^{93}Nb	100	(n,α)	^{90m}Y	3.1	γ: 0.202 (97) 0.482 (91)
^{90}Zr	51.46	(n,p)	^{90}Y	64.0	β^-: 2.27 (99.7)
^{90}Zr	51.46	(n,p)	^{90m}Y	3.1	γ: 0.202 (97) 0.482 (91)
^{92}Zr	17.11	(n,p)	^{92}Y	3.5	γ: 0.448 (2.3) 0.560 (2.6)

in a 0.8 mm cadmium foil and irradiated in the pneumatic tube of TRIGA MARK II reactor for 1 hour. The thermal flux at the irradiation position was 3×10^{12} neutrons per cm^2 per sec. The ratio of the thermal versus fast flux was approximately five.

4. 2. Instrumental. The gamma emitting nuclides were assayed using a 400 channel pulse height analyzer with a $3''\times3''$ NaI(Tl) detector and associated photomultiplier tube, with 9.0 % resolution. The disintegration rates of the isotopes were calculated by total counting efficiency curve of *Fig.* 1. This curve was obtained using calibrated standard sources, ^{241}Am, ^{57}Co, ^{203}Hg, ^{22}Na, ^{137}Cs, ^{54}Mn, ^{60}Co and ^{88}Y. The β emitter, ^{90}Y, was assayed using 2π gas flow counter, Fujitsu Co., Japan, the counting efficiency of which was measured with a substitute standard of UX_1–UX_2, Tracer Lab. Model R-12A(C).

4. 3. Separation of Yttrium. After 30 minutes cooling, the irradiated niobium and zirconium metals were transferred into each platinum crucible of 50 ml capacity and dissolved by treating with 2 ml of conc. nitric acid and a few drops of conc. hydrofluoric acid. 100 λ of yttrium carrier solution (ca. 0.1 mg of yttrium) were added into each crucible and excess acids were evaporated by heating on a hot plate. The sample solutions were quantitatively transferred from the platinum crucibles into each 10 ml volumetric flask and diluted to the volume with distilled water. 1 ml of the solution was pipetted and loaded on a column of the Dowex 5×8 resin (14 cm × 6 mm, 200–400mesh, hydrogen form) which was previously equilibrated with 0.5 *M* α-hydroxy-isobutyric acid solution (pH=1.80).

The column was eluted with 6 ml of 0.5 *M* α-hydroxy-isobutyric acid (α-HIBA) at pH 1.80 in order to remove niobium or zirconm from the column. The remaining yttrium was then eluted from the column by eluting with 10 ml of 0.25 *M* α-HIBA at pH 3.8. The flow rate was adjusted to 0.3 ml per min. with an air compressor. The eluates in fraction of 24 drops, ca. 1 ml, were taken into a test tube by means of a fraction collector. The activity of each tube was measured for 1 min. with a well type scintillation counter and was plotted against the number of the test tube.

As reported by the present authors[7], both niobium and zirconium was quantitatively eluted through the column with 6 ml of 0.5 *M* α-HIBA at pH 1.80. This was confirmed by ^{97}Nb-^{97}Zr tracer which was prepared by irradiating zirconium element[7]. It was also confirmed by tracer work that the remaining yttrium was quantitatively eluted with 20 ml of 0.25 *M* α-HIBA at pH 3.80[7].

The yttrium portion of the eluate in the plotted curve was collected into a 50 ml beaker. 2 ml of La^{3+} carrier solution (10.4 mg of La_2O_3/ml) were added. The yttrium was coprecipitated with lanthanum at pH 4—5 by adding 2 ml of saturated oxalic acid. The precipitates were filtered through a demountable glass filter containing a 2.4 cm dia. disc. of Whatman No. 42 paper, which were then washed with water and finally with acetone. The two filter papers each containing oxalate precipitate, *i.e.*, one from Nb and another from Zr, were removed, and dried under an infra-red lamp. Using scotch tape each was mounted on a 1/32 inch thick aluminum plate.

5. Measurements

5. 1. Flux Monitor ^{58}Co. After 2 days' cooling the irradiated polyethylene tube containing nickel powder was attached on a aluminum plate and counted for the γ-ray spectrum of ^{58}Co. From this spectrum the radiochemical

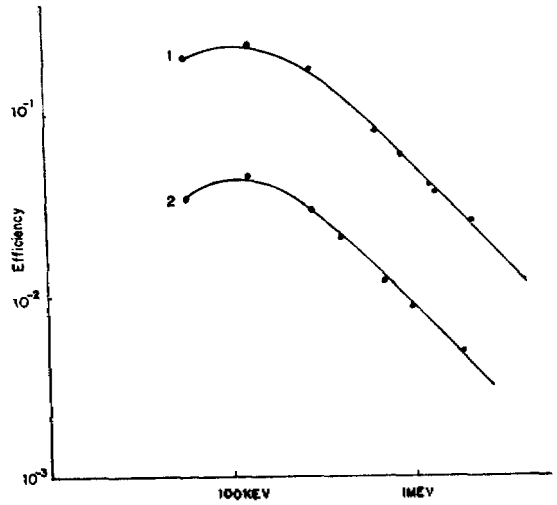

Fig. 1. Efficiency curve of 3″×3″ NaI (Tl)
curve 1: 12 cm distance from crystal.
curve 2: 34cm distance from crystal.

purity of ^{58}Co was confirmed. The disintegration rate of ^{58}Co was calculated with the counting efficiency curve of *Fig.* 1.

5.2. ^{90m}Y and ^{90}Y. The two aluminum plates containing yttrium oxalates were first measured for γ-ray spectrum of ^{90m}Y, and from this spectrum, the radiochemical purity of ^{90m}Y was confirmed. The decay curve was found to be a straight line showing a half life of 3.1±0.2 hours, which is in good agreement with the value of the literature[8]. The disintegration rate of ^{90m}Y was measured with the curve of *Fig.* 1.

After one day the same two yttrium samples were measured for activity of ^{90}Y. The decay curve obtained was a straight line showing a half life of 64±2 hours which is also in good agreement with the value of the literature[8]. The disintegration rate of ^{90}Y was calculated with the measured efficiency (23.1±0.5 %) of 2π gas flow counter. When counting was over the two precipitates were transferred into each platinum crucible, ignited to the oxides in the muffle furnace and weighed as the lanthanum oxides. The observed counting rates were corrected for the chemical yeild (90—95 %).

6. Results and Discussion

The values of the average cross section obtained in the present study were compared with others' as shown in *Table* 2.

The average cross section of $^{93}Nb(n,\alpha)^{90}Y$ and $^{90}Zr(n,p)^{90}Y$ are the total cross section of target nuclides because β rays of ^{90}Y were counted after 24 hours from the end of irradiation. In the present study, the value of 0.14 mb was obtained for the $^{93}Nb(n,\alpha)^{90}Y$ reaction, which is about 1.8 times higher than the value measured by Sanker Das and is about 8.5 times lower than the value predicted by Roy and Hawton[9] as shown on *Table* 2. The cross section, 0.83 mb, obtained in the present study for $^{90}Zr(n,p)^{90}Y$ is comparable with the predicted value, 0.9mb[9]. The nuclide of ^{92}Y produced by $^{92}Zr(n,p)^{92}Y$ (the estimated cross section, 0.16 mb), can interfere with ^{90m}Y because ^{92}Y is the γ-emitter of 0.45—0.56 Mev. photon. However, the γ-ray intensity of ^{92}Y in this energy range is 10 times lower than that of ^{90m}Y. The interference of ^{92}Y was therefore negligible in this work.

The detection limits, LD, of niobium and zirconium were calculated by Currie[10] using the equation $LD=2.71\times3.29\sqrt{\mu_B}$, where μ_B is the limiting mean of blank. The results are shown

Table 2. Average Fission Neutron Cross Section in mb

Reaction	Cross section of this work	Cross section of others
$^{93}Nb(n,\alpha)^{90}Y$	0.14±0.01	0.077[3] 0.039[2] 0.0585[12]
$^{93}Nb(n,\alpha)^{90m}Y$	0.018±0.002	0.0221[12]
$^{90}Zr(n,p)^{90}Y$	0.83±0.002	0.90*
$^{90}Zr(n,p)^{90m}Y$	0.033±0.002	

* estimated cross section[9]

Table 3. Detection Limits of Nb and Zr

Reaction	Cross section (mb)	Detection limit (μg)		
		β-ray	γ-ray	
			0.202Mev.	0.482Mev.
$^{93}Nb(n,\alpha)^{90}Y$	0.14	200		
$^{93}Nb(n,\alpha)^{90m}Y$	0.018		20	60
$^{90}Zr(n,p)^{90}Y$	0.83	80		
$^{90}Zr(n,p)^{90m}Y$	0.033		20	40

on *Table* 3 each of which was counted for 1 hour. From this *Table* one can estimate that the detection limit of niobium attainable by the present method using $^{93}Nb(n,\alpha)^{90m}Y$ reaction could be 10 times as sensitive as that by the $^{93}Nb(n,\alpha)^{90}Y$ reaction[3]. The results on the determination of niobium using the $^{93}Nb(n,\alpha)^{90m}Y$ reaction will be published[11].

Recently De Regge[12] et al. also reported the mean fission cross sections for the reaction of $^{93}Nb(n,\alpha)^{90}Y$ and $^{93}Nb(n,\alpha)^{90m}Y$ as shown in *Table* 2. The value of (22.1±0.3) micro barns for $^{93}Nb(n,\alpha)^{90m}Y$ reaction agrees rather well with our result. However, (58.5±2.2) micro barns for $^{93}Nb(n,\alpha)^{90}Y$ reaction is lower than our value by the factor of about three. The present authors are unable to offer adequate explanation for this difference.

At present the cross section 0.033±0.002 mb, obtained in the present investigation for $^{90}Zr(n,p)^{90m}Y$ reaction can not be compared with others' because of the lack of available data.

Reference

1. J. C. Roy and D. Wuschke, AECL-877 (1959).
2. E. Sealand and K. Samsahl, from R. S. Rochlin "Nucleonics Data Sheet No. 28", Nucleonics 17, No. 1, 54, 1959.
3. M. Sankar Das, presented at the Study Group Meeting of the I. A. E. A., in Dec. 1964, at the Tata Institute of Fundamental Research, Bombay, India.
4. D. J. Hughes, "Pile Neutron Research" Addison-Wesley Publishing Co., Cambridge, Mass., 1953.
5. C. E. Mellish, Nucleonics 19, No. 3, 114 (1961).
6. J. Moteff, Nucleonics 20, No. 12, 56 (1961).
7. C. Lee, Y. C. Yim and K. S. Chung, "Column Chromatographic Separation of Ti, Zr and Nb", *J. Korean Chem. Soc.* **17**, 15 (1973).
8. C. M. Lederer, J. M. Hollander and I. Perlmann, Tables of Isotopes, John Wiley and Sons, New York, 1968.
9. J. C. Roy and J. J. Hawton, AECL-1181, 1960.
10. L. A. Currie, *Anal. Chem.*, 40, 586 (1968).
11. C. Lee, Y. C. Yim and K. S. Chung, "Simultaneous Determination of Ti, Zr and Nb by Neutron Activation", will be sumitted to *J. Korean Chem. Soc.* (1973).
12. P. De Regge, R. Dams and J. Hoste, *Radiochimica Acta* **17**, 69 (1972).