

CROSS SECTIONS FOR THE $^{nat}\text{Zr}(p,xn)^{89,90}\text{Nb}$ REACTIONS INDUCED BY 27.7 MeV PROTONS

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Received 05 May 2015

Accepted for publication 11 September 2015

Abstract. *The cross-sections for the formation of ^{89}Nb and ^{90}Nb radionuclides in proton induced nuclear reactions on zirconium were measured by using the well known activation method. The natural zirconium (^{nat}Zr) target and copper (^{nat}Cu) monitor foils were irradiated by 27.7 MeV proton beam at the MC50 Cyclotron of the Korea Institute of Radiological and Medical Science (KIRAMS), Korea. The induced gamma activities of the reaction products were measured by a coaxial high purity germanium (HPGe) detector coupled to a PC-based multichannel analyzer. The obtained cross sections for each nuclide are compared with those existing in literature and with the theoretical cross sections calculated by the TALYS - 1.4 code.*

Keywords: proton induced reaction, cross-section, activation method, HPGe detector, TALYS code, natural zirconium.

I. INTRODUCTION

Measurements of charged particle induced nuclear reaction cross sections are of increasing importance for various fields of applications such as medical radioisotope production, spallation neutron sources or accelerator-driven system for transmutation of nuclear waste, space radiation effect, radiation damage of materials as well as radiation shielding design and activation analysis. Furthermore, integral cross sections are the most important data for testing and validating nuclear model codes.

In this study, we determined the cross sections for the $^{nat}\text{Zr}(p,xn)^{89,90}\text{Nb}$ reactions at proton energy of 27.7 MeV. The zirconium alloys have low neutron-capture cross-section and high mechanical and chemical resistance. Therefore in practical applications zirconium becomes an important reactor material like cladding of fuel elements. So far, the cross sections of the $^{nat}\text{Zr}(p,xn)^{89,90}\text{Nb}$ reactions were given by several authors, however there are some discrepancies among the reported data [1–5]. In addition, the ^{90}Nb is a positron emitting radioisotope that can be used for immune-PET scans [6, 7].

The aim of this work was to measure the cross section of the $^{nat}\text{Zr}(p, xn)^{89}\text{Nb}$ and $^{nat}\text{Zr}(p, xn)^{90}\text{Nb}$ reactions with 27.7 MeV protons. The experiments were carried out by using the well known activation method in combination with HPGe gamma ray spectroscopy. The obtained results together with the existing reference data may help in understanding the excitation function of the proton induced nuclear reactions.

II. EXPERIMENTAL PROCEDURE

In this experiment, the irradiation was performed with proton beam at the Cyclotron Application Laboratory, Institute of Radiological and Medical Science (KIRAMS), Seoul, Korea. In this investigation, the proton beam with energy of 27.7 MeV was used. The activation foil was a high purity natural zirconium with isotopic abundances as follows: ^{90}Zr (51.45%), ^{91}Zr (11.22%), ^{92}Zr (17.15%), ^{94}Zr (17.38%), ^{96}Zr (2.8%). The diameter of zirconium foil is $1/2$ inch and the thickness is 0.1 mm.

For irradiation, the Zr and Cu foils with the same diameter were placed at 25 cm from the window of the proton beam and irradiated simultaneously. These foils were irradiated for 3 min with 27.7 MeV protons at a beam current of 50 nA. The gamma ray activities of the activated foils were measured by using a high-purity HPGe detector (ORTEC) with energy resolution of 1.8 keV at the 1332.5 keV peak of ^{60}Co . The detector was coupled to a computer-based multichannel analyzer card system, which could determine the photo-peak area of the gamma spectrum. The counting efficiency of the detector was determined experimentally using a set of gamma standard sources. The photo-peak efficiency, ε , was calibrated with a set of standard sources such as ^{241}Am , ^{109}Cd , ^{137}Cs , ^{54}Mn , ^{60}Co , ^{152}Eu and ^{133}Ba . In order to optimize the dead time and the coincidence summing effect, we have chosen the appropriate distance between the sample and the detector for each measurement. Each sample was measured several times in order to follow the decay of the different isotopes. Gamma spectra of the irradiated Zr and Cu foils are shown in Fig. 1 and Fig. 2, respectively.

III. DATA ANALYSIS

The nuclear reactions to be considered here are $^{nat}\text{Zr}(p, xn)^{89}\text{Nb}$, $^{nat}\text{Zr}(p, xn)^{90}\text{Nb}$ and $^{63}\text{Cu}(p, 2n)^{62}\text{Zn}$, respectively. The nuclear reactions and their characteristics are given in Table 1 [8]. The measured gamma spectra were analyzed by using the Gamma Vision software, version 5.10 (EG&G ORTEC). Then, the radioactive isotopes were identified based on their characteristic gamma ray energies and half-lives. The activity of each reaction product was determined from the area under photo peaks of the gamma rays with high intensity and well separated.

The production cross sections of the $^{nat}\text{Zr}(p, xn)^{89}\text{Nb}$ and $^{nat}\text{Zr}(p, xn)^{90}\text{Nb}$ reactions were determined based on the measured activity of the reaction products. The activities were measured based on the following gamma rays: 920.70 keV (1.39%) and 1627.20 keV (3.4%) for ^{89}Nb , and 141.178 keV (66.8%) and 1129.224 keV (92.7%) for ^{90}Nb , respectively. After measurements and making necessary corrections, the reaction cross section, σ , was calculated as follows [9, 10]:

$$\sigma = \frac{N_{obs}\lambda}{\phi_p N_o \varepsilon I_\gamma F (1 - e^{-\lambda t_i}) e^{-\lambda t_d} (1 - e^{-\lambda t_c})} \quad (1)$$

where N_{obs} is the measured activity, N_o is the number of the target atoms, ϕ_p is the flux of the proton beam, I_γ is the gamma ray intensity, ε is the absolute photopeak efficiency value of the

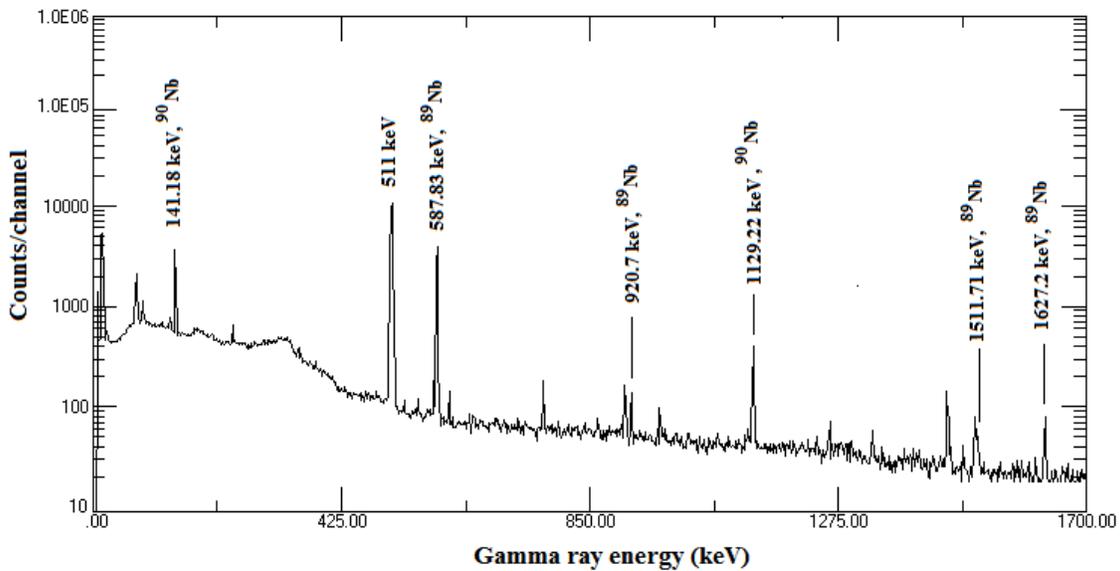


Fig. 1. The gamma spectrum of the ^{nat}Zr foil irradiated with 27.7 MeV proton beam ($t_i = 3$ min, $t_d = 26$ min and $t_c = 10$ min)

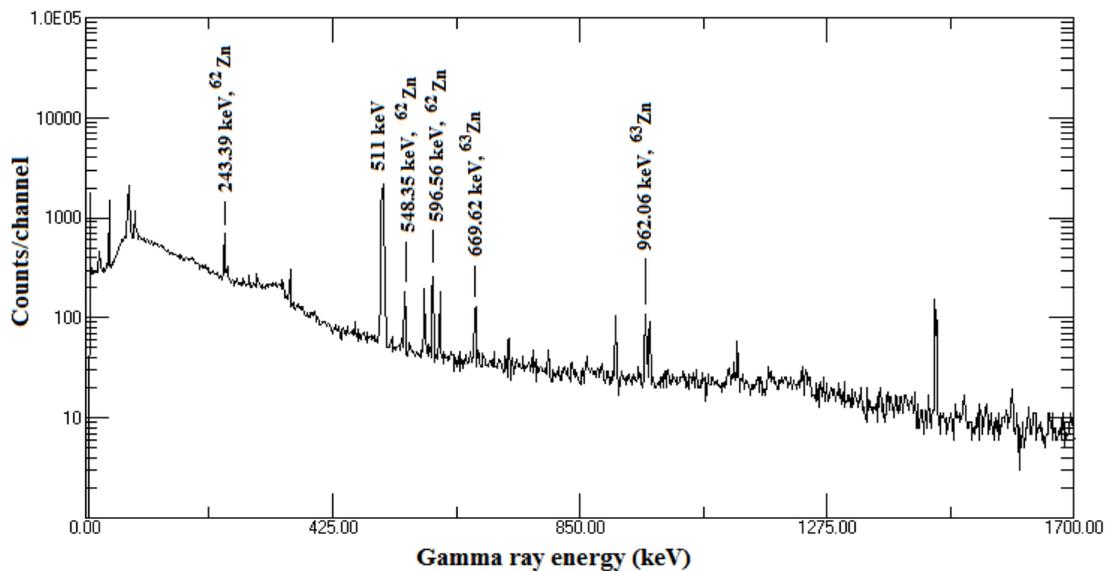


Fig. 2. The gamma spectrum of the ^{nat}Cu foil irradiated with 27.7 MeV proton beam ($t_i = 3$ min, $t_d = 110$ min and $t_c = 15$ min)

specific gamma-ray, F is the correction factor for counting losses due to the γ -ray attenuation [11] and the true coincidence summing effect [12], and λ is the decay constant of the product nucleus,

and t_i , t_d and t_c are irradiation, waiting and counting times, respectively. The incident proton flux at the irradiation position, ϕ_p , was determined based on the monitor reaction $^{63}\text{Cu}(p, 2n)^{62}\text{Zn}$ with known cross section [13] and decay data as listed in Table 1.

Table 1. Nuclear reactions investigated and decay data of the produced radionuclides [8]

Nuclei	Half-life, $T_{1/2}$	γ -energy, E_γ (keV)	γ -ray intensity, I_γ (%)	Contributing reactions	Threshold energy, E_{th} (MeV)
^{89}Nb	1.9 h	587.83	1.39	$^{90}\text{Zr}(p, 2n)^{89}\text{Nb}$	17.24
		920.70	1.39		24.44
		1511.71	1.87	$^{91}\text{Zr}(p, 3n)^{89}\text{Nb}$	
		1627.20	3.4		
^{90}Nb	14.6 h	132.716	4.13	$^{90}\text{Zr}(p, n)^{90}\text{Nb}$	6.97
		141.178	66.8	$^{91}\text{Zr}(p, 2n)^{90}\text{Nb}$	14.24
		371.307	1.80		22.97
		1129.224	92.7	$^{92}\text{Zr}(p, 3n)^{90}\text{Nb}$	
^{62}Zn	9.186 h	243.39	2.52	$^{63}\text{Cu}(p, 2n)^{62}\text{Zn}$	13.48
		507.60	14.8		
		548.35	15.3		
		596.56	26.0		

IV. THEORETICAL CALCULATION

With a view to validate the experimental data and test the nuclear theory models, the reaction cross-sections were calculated theoretically using the TALYS-1.4 code. TALYS is a software package for the simulation of nuclear reactions induced by almost possible incident particles in the 1 keV - 200 MeV energy range and for target nuclides of mass numbers between 12 and 339.

It is in general an implementation of many of the latest nuclear models for direct, compound, pre-equilibrium and fission reactions. TALYS can be used for calculations of total and partial cross sections, residual production cross sections, energy spectrum angular distributions, . . . [14].

In this work, the excitation functions for the $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ and $^{nat}\text{Zr}(p,xn)^{90}\text{Nb}$ reactions were theoretically calculated by the TALYS 1.4 code. For each activation product, the reaction cross sections on the individual target isotopes were calculated. According to the abundance of natural zirconium isotopes, a weighted summation was made to obtain the production cross-section.

V. RESULTS AND DISCUSSION

The experimental cross sections for the $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ and $^{nat}\text{Zr}(p,xn)^{90}\text{Nb}$ reactions together with the available reference data are given in Table 2. The total uncertainties were estimated to be 8%. The main sources of the uncertainty for the experimental results are due to the statistical error ($\sim 2\%$), detection efficiency ($\sim 2.5\%$), photo-peak area determination ($\sim 3\%$), coincidence summing effect ($\sim 1.5\%$), proton beam ($\sim 3\%$) and nuclear data used ($\sim 3.5\%$) and others ($\sim 4\%$), respectively.

Table 2. Experimental cross-sections of the $^{nat}\text{Zr}(p,xn)^{89,90}\text{Nb}$ nuclear reactions

Proton energy (MeV)	Cross-section of $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ (mb)		Proton energy (MeV)	Cross-section of $^{nat}\text{Zr}(p,xn)^{90}\text{Nb}$ (mb)	
	This work	Refs.		This work	Refs.
17.8		43 ± 26 [5]	24.58		142.5 ± 12.8 [2]
18.4		64 ± 31 [5]	26.6		108.8 ± 6.8 [3]
19.0		91 ± 40 [5]	27.7	140.9 ± 11.3	
27.7	203.4 ± 16.3		28.79		140.0 ± 11.2 [2]
			30.7		146.0 ± 11.7 [2]

The experimental and calculated results based on the TALYS-1.4 code are shown in figures 3 and 4. The present measured cross-section of the $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ reaction is in agreement with the tendency of the values measured by S. Busse *et al.* [1]. In the case of ^{90}Nb , the obtained result is also in agreement with the results of M. S. Uddin *et al.* [3] and M. U. Khandaker *et al.* [4]. As

can be seen in figures 3 and 4, almost experimental data close to the excitation functions calculated by the TALYS-1.4 code.

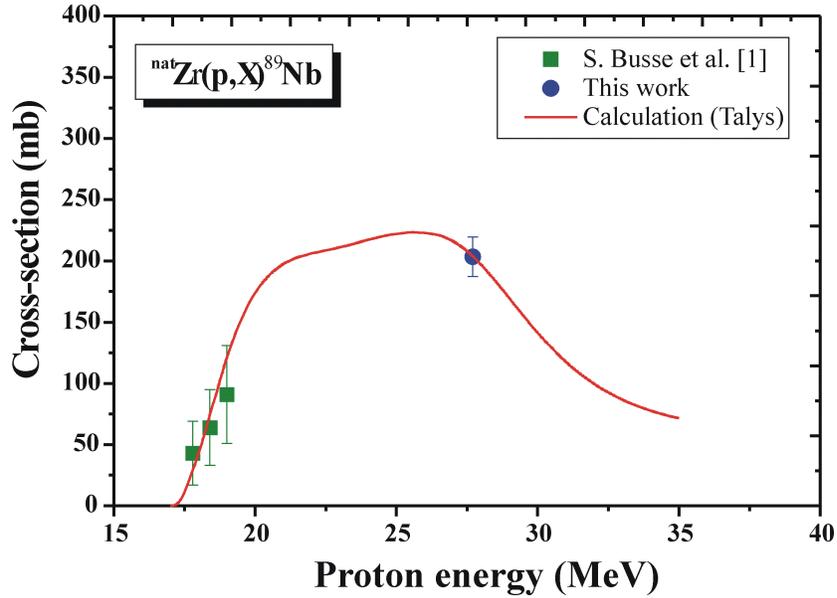


Fig. 3. Energy dependence of the cross section for the $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ reaction

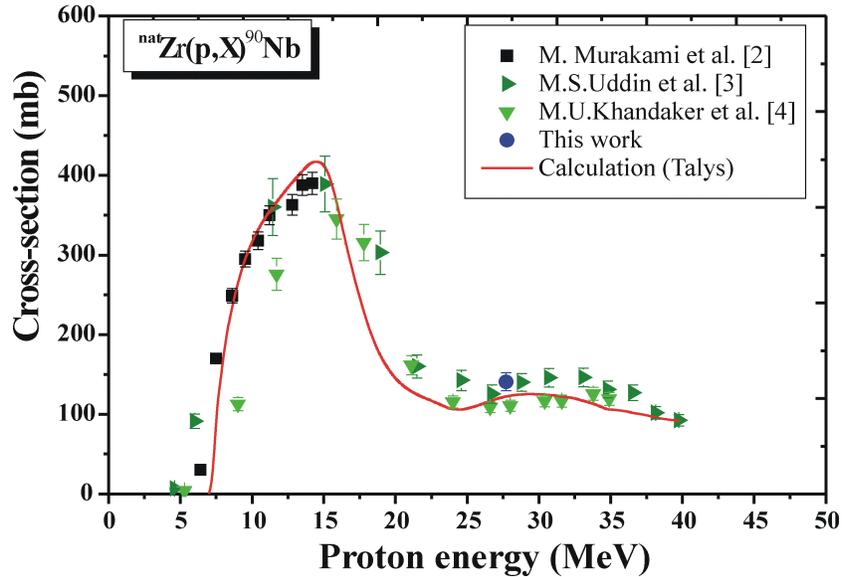


Fig. 4. Energy dependence of the cross section for the $^{nat}\text{Zr}(p,xn)^{90}\text{Nb}$ reaction

VI. CONCLUSION

We have measured the cross section of the $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ and $^{nat}\text{Zr}(p,xn)^{90}\text{Nb}$ reactions induced by the 27.7 MeV proton beam by using the well known activation method. We have also calculated the excitation function of the $^{nat}\text{Zr}(p,xn)^{89}\text{Nb}$ and $^{nat}\text{Zr}(p,xn)^{90}\text{Nb}$ reactions by using the TALYS-1.4 code. The existing literature data were collected and compared with our results. The present cross sections are in good agreement with both the experimental and calculated data in the literature.

ACKNOWLEDGMENTS

The authors would like to express their sincere thanks to the Cyclotron Application Laboratory, KIRAMS, and to the Pohang Accelerator Laboratory, POSTECH, Korea for their valuable support in carrying out this experiment.

REFERENCES

- [1] S. Busse, F. Rosch, S.M. Quim, *Radiochim. Acta* **90** (2001) 1.
- [2] M. Murakami, H. Haba, S. Goto, J. Kanaya, H. Kudo, *Appl. Radiat. Isot.* **90** (2014) 149.
- [3] M.S. Uddin, M.U. Khandaker, K.S. Kim, Y.S. Lee, G.N. Kim, *Nucl. Instrum. Meth. B* **266** (2008) 13.
- [4] M.U. Khandaker, K. Kim, M.W. Lee, K.S. Kim, G.N. Kim, Y.S. Cho, Y.O. Lee, *Appl. Radiat. Isot.* **67** (2009) 1341.
- [5] M. Al-Abyad, A.S. Abdel-Hamid, F. Tarkanyi, F. Ditroi, S. Takacs, U. Seddik, I.I. Bashter, *Appl. Radiat. Isot.* **70** (2012) 257.
- [6] F. Szelecsenyi, G.F. Steyn, Z. Kovacs, C. Vermeulen, K. Nagatsu, M.R. Zhang, K. Suzuki, *Nucl. Instrum. and Meth. B* **343** (2015) 173.
- [7] V. Radchenko, H. Hauser, M. Eisenhut, D. J. Vugts, G. A. M. S. van Dongen, and F. Roesch, *Radiochim. Acta* **100** (2012) 857.
- [8] Richard B. Firestone, "Table of Isotopes", 8th Edition, Wiley-Interscience, 1996.
- [9] Nguyen Van Do and Pham Duc Khue. *Comm. Phys.* **4** (2005) 193.
- [10] Sung-Chul Yang, Guinyun Kim, M. Zaman, Kwangsoo Kim, Tae-Yung Song, Young-Ouk Lee, Sung Gyun Shin, Young-Uk Key, Moo-Hyun Cho, Duc Khue Pham, Van Do Nguyen, Haladhara Naik, Tae-Ik Ro, *J. Radioanal. Nucl. Chem.* **302** (2014) 467.
- [11] Nguyen Van Do, Pham Duc Khue, Kim Tien Thanh, Nguyen Thi Hien, Guinyun Kim, Sungchul Yang, Young-Sik Cho, Tae-Yung Song, Young-Ouk Lee, Sung Gyun Shin, Moo-Hyun Cho, Man Woo Lee, *Nucl. Instr. Meth. B* **335** (2014) 1.
- [12] Nguyen Van Do, Pham Duc Khue, Kim Tien Thanh, Nguyen Thi Hien, G. N. Kim, S. Yang, K. S. Kim, S. G. Shin, M. H. Cho, M. W. Lee, *Nucl. Instr. Meth. B* **310** (2013) 10.
- [13] International Atomic Energy Agency (IAEA), "Charged Particle Reference Cross Section", <https://www-nds.iaea.org/medical/cup62zn0.html>.
- [14] A. Knoning, S. Hilaire, S. Goriely, "Talys 1.4 User Manual", NRG, 1st edition, 28/12/2011.