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Production of Residual Nuclides by Proton-Induced Reactions on Target W at an Energy of 72 MeV

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Investigations of cross-sections for residual nuclide production on the target element W by proton-induced reactions were performed by irradiating the target with 72 MeV protons using the cyclotron facilities at Paul-Scherrer Institute, Zurich, Switzerland. Residual nuclides were measured by gamma-spectrometry of HpGe detectors calibrated with standard gamma sources. The measured data contains 104 individual cross-sections for 20 identified nuclides in the proton energies between 52.5 - 68.9 MeV. These nuclear data is important in the study of spallation neutron source and in accelerator driven technologies such as waste transmutation and energy amplification. The present data are compared with the shape of the excitation functions of earlier only one measurement at higher energies and they are in good agreement to each other.

KEYWORDS: proton-induced reactions, target element, spallation neutron source, transmutation, energy amplification

I. Introduction

The spallation reaction between target materials and a high energy proton beam generates many neutrons. The productivity of neutrons is favoured by the use of target materials of high density and high neutron yield. In this regard, the target W (Tungsten) is a probable candidate to use it as a spallation neutron source. The cross-sections data for the proton-induced reactions on the target W in the energy range from 2600 MeV towards down to 68.3 MeV have previously been measured.[1)] But in that measurement, the cross-sections for many numbers of product nuclides especially below 100 MeV were not identified. Up to now, there is no experimental data below than the mentioned lowermost energy point (68.3) of the previous measurement for the same target.

In the present study, the investigation of residual nuclide production by 72 MeV protons on the target W was carried out by using the cyclotron at PSI. The present measured data are of importance for the calculation of theoretical model, for environmental sciences, medicine (nuclide production and radiation theraphy), accelerator driven transmutation and energy amplification.

II. Measurement of Cross-Sections

1. Formula for the Determination of Cross-section

When a target with a number of atoms N_t is irradiated with protons of flux ϕ ($cm^{-2}s^{-1}$) for a time t_{irr}, the activity of a product p at the end of irradiation A_{irr} is given by

$$A_{irr} = N_t \sigma \phi \left(1 - e^{-\lambda_p t_{irr}}\right) \tag{1}$$

where, σ is the cross-section and λ_p is the decay constant.

After the end of irradiation, as the time will be going on, the activity of the product will be decreasing and after a waiting time of t_w (time between end of irradiation and beginning of counting), the activity is given by

$$A_w = A_{irr} e^{-\lambda_p t_w} \tag{2}$$

The net counts N of a full energy peak of gamma-transition of the same nuclide for a counting time t_c is given by

$$N = \int_0^{t_c} A_{irr} e^{-\lambda_p t} \epsilon_\gamma I_\gamma dt \tag{3}$$

where, ϵ_γ is the peak efficiency of the detector with necessary corrections, I_γ is the absolute branching ratio for a particular γ-energy of the product nuclide. Integrating Equ. (3), the activity at the end of irradiation yields as

$$A_{irr} = \frac{\lambda_p N}{(1 - e^{-\lambda_p t_c})\epsilon_\gamma I_\gamma} e^{\lambda_p t_w} \tag{4}$$

Combining Equ. (1) and Equ. (4), we obtain

$$\sigma = \frac{\lambda_p N e^{\lambda_p t_w}}{N_t \phi (1 - e^{-\lambda_p t_{irr}})(1 - e^{-\lambda_p t_c})\epsilon_\gamma I_\gamma} \tag{5}$$

which is the required formula for the determination of cross-section.

2. Irradiation of Targets and Flux Monitoring

The irradiation of target stacks was performed for a period of 7.8 h with initial protons of energy 72 MeV at PSI, Switzerland. Each stack was composed of target W, Al and Cu foil for flux monitoring. The foils in stacks were arranged in such a way that Al and Cu-foils were placed in front and behind of target foils respectively. The target in a stack consisted of three thin foils of natural tungsten (0.13% ^{180}W, 26.3% ^{182}W, 14.3% ^{183}W, 30.67% ^{184}W and 28.6% ^{186}W)[2)] of high purity

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materials (99.99%) with diameters of 15 mm and a thickness of 0.050 mm. Like targets in stacks, sets of three thin Al-foils (thickness 0.036 - 0.125 mm) and Cu-foils (thickness 0.025 - 0.050 mm) were used to monitor the proton flux. The inner ones of three foils for both target and monitor were used to prevent the cross contaminations from other elements due to the destruction of foils by radiation damage or heating during irradiation. In this measurement, an arrangement of six stacks was used by a supporting device on which the center of the stacks was in the beam line.

At the beginning of the measurement, the beam was positioned on an empty stackholder and the beam current was measured by an uncalibrated Faraday Cup. During irradiation, the beam currents were continously recorded by the Faraday cup connected with a digital meter coupled to a PC. The contributions due to the fluctuation of proton flux and the secondary particles produced in the target stack was taken into account and found to be negligible. The flux for the corresponding target positions of different stacks was measured from the nuclide ^{22}Na via the reaction $^{27}\mathrm{Al}(p, 3p3n)^{22}\mathrm{Na}$. For this measurement, the cross-sections for the standard reaction at specific proton energies were taken from the measurement.[3] The uncertainty of proton flux was mainly determined by the uncertainties of efficiency of detectors and the mass of the monitor foil. No uncertainties due to the monitor cross-sections were included in this calculation and the reason has been explained elsewhere.[4] The correction for the activity due to the self absorption of γ -rays in the targets itself was taken into account very carefully.

The calculation of proton energies for all individual target foils was performed by a computer program based on the work as in literature,[5] giving the initial proton energy of 71.9±0.513 MeV and hence the calculated energy range in the first and the last target foils in stacks is 52.2 - 68.9 MeV. In the calculation of the uncertainty of proton energy, the uncertainty due to its initial energy just leaving the accelerator, its energy spread and straggling passing through the target stacks was taken into account attentively. The resultant uncertainty for this contribution is less than 1% .

After the end of irradiation, the samples were transported to Cologne within 12 h where the stacks were dismounted and the individual foils separated for further measurement. Subsequently, the γ -spectrometric measurements for all irradiated samples were performed by means of Ge(Li) and HpGe detectors calibrated with standard gamma sources with certified accuracies of their activities $\leq$ 2.0%. The repeated measurements during the reasonable time span allowed us to get a minimum spectra of four for each nuclide. To avoid problems of high dead times and pile-up effects, the distances between the sample and detector were varied to keep the dead time below 10%. The large distances were used only for the short-time measurements of nuclides those who have high activities after the irradiation. Background measurements were taken for about 3 days and its contribution was subtracted for the same counting time for the corresponding nuclides. After then these spectra were stored for further analysis.

3. Analysis of Spectra

The identification of product nuclides and the corresponding activities were carried out by using the computer code GAMMA-W.[6,7] The identification process starts with the spectra having the longest decay times.Considering the correction for the γ-rays in the targets first such lines are selected which can be unambiguously identified. Then, successively going back in time, the identified nuclides are searched in the preceeding spectra by calculating activities and looking whether these activities should be detectable in the spectrum under consideration. Applying this algorithm decreases the number of possible nuclides in a spectrum.

In the step of identification of nuclides, the half-lives from the chart of nuclides,[8] energies and branching ratios of γ-rays were taken from the compilation.[9] By knowing the proton flux and measuring the activity for the respective product nuclide, the cross-sections for specific proton energies at different target positions were determined using the program SPECTER[10] according to Equ. (5).

III. Result and Discussion

The measured cross-sections for the element of Tungsten contain 104 individual cross-sections for 20 identified nuclides in the proton energies between 52.2 - 68.9 MeV. All the identified nuclides with their half-lives are presented in **Table 1**. The cross-sections data for all reactions in either tabular form or graphics will contain a large volume in this paper. As such as, only a few excitation functions of them with the present experimental data and the previously measured one up to the energy of 105 MeV have been presented in **Figures 1, 2, 3** and **4**.

From these figures, it is observed clearly that the recent measured data at 68.9 MeV coincides with the previous one at the energy of 68.3 MeV and they are in excellent agreement with the structure to each other. Moreover, the uncertainty of cross-sections in the present measurement is rather smaller than the previous one. For only a few cases of remaining reac-

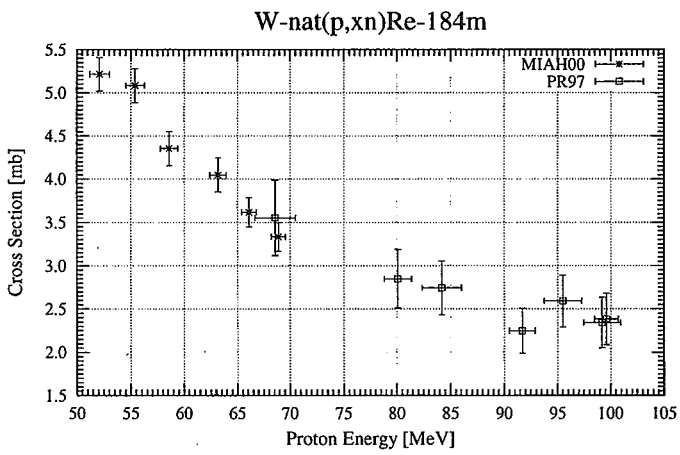

Fig. 1 Experimental excitation function for the production of ^{184m}Re together with the data of previously measured by Protoschill 97.[1]

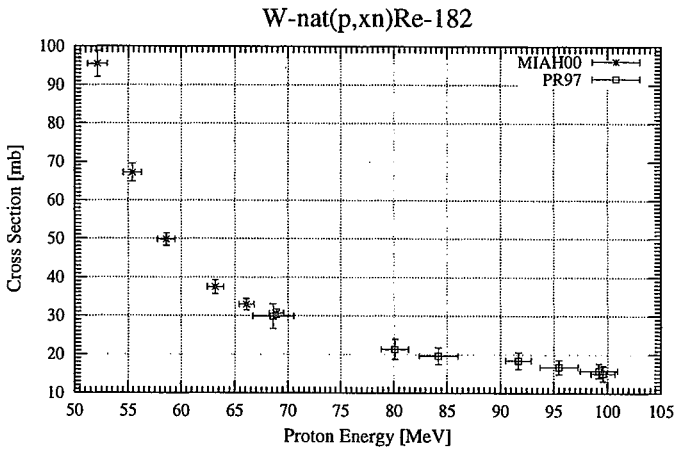

Fig. 2 Experimental excitation function for the production of ^{182}Re together with the data of Protoschill 97.[1)]

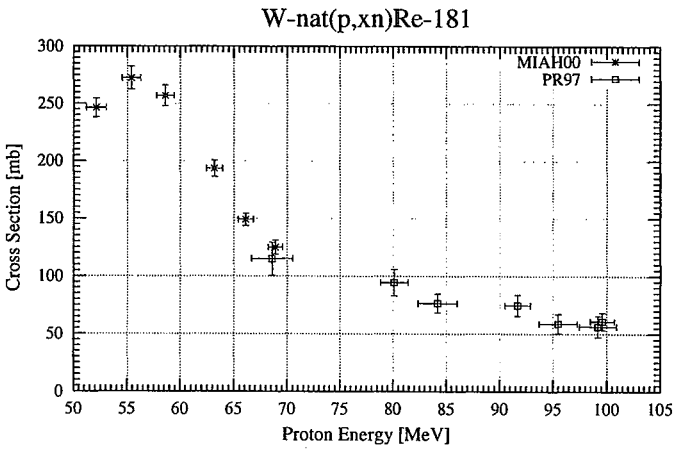

Fig. 3 Experimental excitation function for the production of ^{181}Re together with the data of Protoschill 97.[1)]

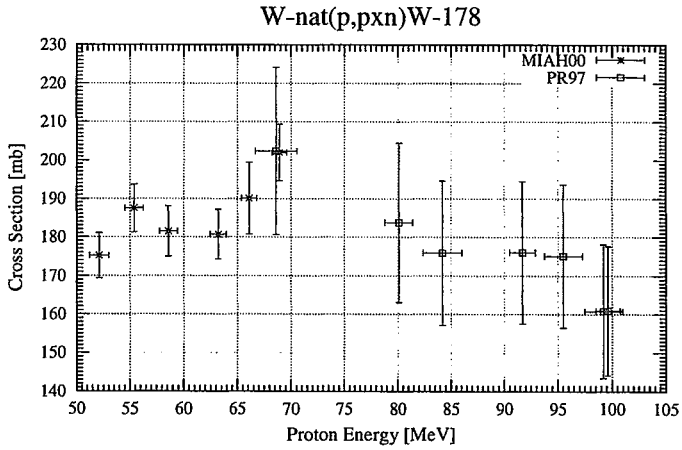

Fig. 4 Experimental excitation function for the production of ^{178}W together with the data of Protoschill 97.[1)]

Table 1 Product nuclides with their half-lives of Tungsten.

Product	Half-life (days)	Product	Half-life(days)
^{186}Re	13.78	^{181}Re	0.83
^{184g}Re	38.0	^{183}Ta	5.10
^{184m}Re	165	^{182}Ta	115
^{183}Re	70.0	^{177}Ta	2.36
^{182g}Re	2.67	^{176}Ta	0.34
^{182m}Re	0.53	^{178}W	22.0
^{181}Hf	42.4	^{179B}Hf	25.1
^{175}Hf	70.0	^{173}Hf	1.0
^{177g}Lu	6.71	^{173}Lu	500.38
^{88}Zr	83.4	^{88}Y	106.6

Table 2 Main sources of uncertainty with the maximum limit of errors in the present measured cross-sections.

Sources of uncertainty	Errors in %
Target mass	negligible
Flux fluctuation during irradiation time	negligible
Irradiation time and counting time	negligible
Half-lives	1.0
Intensity of gamma-rays	1.0
Self absorption of gamma-rays in target foils	< 1.0
Nuclide production by secondary particles	negligible
Contamination due to recoil losses	not considered
Absolute efficiency calibration	5.0
Proton fluence	5.5
Mass of monitor foil	< 1.0
Self absorption of gamma-rays in monitor foils	negligible
Dead time and pile-up losses in γ-spectrometry	negligible
Statistical uncertainty in net peak areas	2.0

tions of same kinds, the shape of the present excitation functions are slightly deviated with the previous one. The fact behind this reason was most probably due to the determination of cumulative cross-sections (i.e; the cross-sections for the production of nuclides via the nuclear reactions and via the decay of precursors) in the previous measurement. In our measured data, all the main sources of uncertainties and their resulting errors are presented in **Table 2**. Here to obtain the total uncertainty, all the uncertainties were added in quadrature according to the law of error propagation.

From the preliminary survey on the magnitude of production cross-sections it is seen that the large cross-sections are preferred for short-lived nuclides and on the contrary, small cross-sections are for long-lived ones. The two fission products ^{88}Zr and ^{88}Y as shown in **Table 1** exhibit an important feature in this study. The excitation functions for both of them increase monotonically up to the highest proton energies investigated, though their absolute cross-section values are very small (in micro-barn). Only by looking the shape of the individual excitation functions of products, it is not enough to distinguish the modes of reactions such as spallation, fission and reactions leading to the residual nuclides close to the target nuclei. To solve this problem, the theoretical model calculation using the computer codes system based on the experimental

data is suited for understanding the intranuclear cascade (INC) followed by an evaporation as well as preequilibrium process between the mentioned two.

IV. Conclusion

The nuclide production cross-sections on thin target element W were measured by off-line γ-spectrometry for 20 identified nuclides in the proton energy range of 52.2 - 68.9 MeV at different target foil positions. The obtained results were compared graphically with the only one measrement (Protoschill 97) in a small interfering energy region for the same kind of products and they are in excellent agreement to each other.Now it is necessary to see the excitation function below 50 MeV for the near target products.

The theoretical model calculations with ALICE-IPPE on the basis of experimental data base for the corresponding product nuclides is now in the final stage and its outcome will be published in a separate issue.Furthermore, the present data base will also provide a tool to improve the models of nuclear reactions in a wide energy region.

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