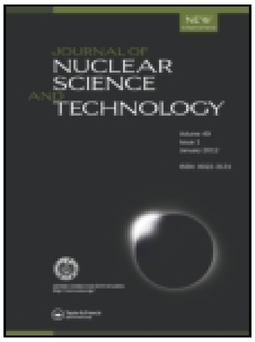

Journal of Nuclear Science and Technology

ISSN: 0022-3131 (Print) 1881-1248 (Online) Journal homepage: https://www.tandfonline.com/loi/tnst20

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To cite this article: M.S. Uddin, M. Hagiwara, N. Kawata, T. Itoga, N. Hirabayashi, M. Baba, F. Tarkanyi, F. Ditroi & J. Csikai (2004) Measurement of excitation functions of the proton-induced activation reactions on tantalum in the energy range 28-70 MeV, Journal of Nuclear Science and Technology, 41:sup4, 160-163, DOI: 10.1080/00223131.2004.10875669

To link to this article: https://doi.org/10.1080/00223131.2004.10875669

Published online: 27 Aug 2014.

Journal of NUCLEAR SCIENCE and TECHNOLOGY, Supplement 4, p. 160–163 (March 2004)

Measurement of excitation functions of the proton-induced activation reactions on tantalum in the energy range 28-70 MeV

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Excitation functions for the proton-induced activation reactions on Ta were measured as a function of proton energy in the range 28- 70 MeV using the k=110 MeV AVF Cyclotron of Tohoku University. In this work the production of ^{175,176,177,180}Ta, ^{173,175}Hf, ^{178}W and ^{179}Lu radionuclides were investigated. For ^{181}Ta(p,x)^{176}Ta and ^{181}Ta(p,x)^{179}Lu reactions no data existed and for other reactions only contradictory data existed in the literature. Therefore our measurements have given new data for all of these reactions. In most of the cases, the results of this work are consistent with the available literature values.
Keywords: Ta+ p reactions, excitation functions, 28-70 MeV, activation

I. Introduction

Activation cross-sections data are required for radiation safety and estimation of radioactive wastes. Activation cross-sections of the Ta+p reactions are of interest for evaluation of radioactivity around accelerators because tantalum is used in various parts of accelerators and spallation neutron sources, for production of medical radioisotopes (^{178}W/^{178}Ta, ^{172}Hf/^{172}Lu, ^{177}Lu, etc). However, the predictions of model calculations are unreliable. Therefore experimental data are of vital importance. Only very few laboratories perform systematic experimental investigations for dedicated applications and in many cases they are not detailed enough for the applications.

Despite of the importance of the Ta+p reaction, only a very few cross-section measurements were published in the literature[1-6]. Besides the published data are very contradictory to each other and to the results of the theoretical calculations. No recommended data were proposed on the basis of the presently available experimental data. In view of these considerations, the present experiment was undertaken to obtain new and reliable cross-section data for the activation in the reactions of Ta+p in the energy range of 28-70 MeV. The theoretical cross-section calculations compiled in MENDL[7] and by the PHITS[8] code which is a new version of NMTC/JAM code based on INC and GEM models were compared with the experimental data.

II. Experimental Techniques

The excitation functions were obtained by irradiating stacked Ta foils and Cu and Al monitor foils of natural isotopic compositions by a 70 MeV incident energy proton beam. Stacks were made of aluminum, tantalum, copper, titanium, iron, platinum, palladium and zinc. Each stack was consisted of six different types of the above metallic target foils. Two experiments were taken place in the same experimental conditions. The foil arrangement in the stack of the 1st irradiation was different from that of the 2nd irradiation. These stacks were irradiated by a collimated proton beam of 100 nA for 1 hour using the k=110 MeV AVF Cyclotron at Cyclotron and Radioisotope Center (CYRIC), Tohoku University, Sendai, Japan[9]. The stacked samples were brought instantly into the irradiation place by an automated transfer system newly designed at CYRIC. It was necessary to ensure that equal areas of the monitor and the target foils intercepted the beam. The irradiation geometry used guaranteed that practically the whole entering beam passed through every foil. Aluminum (98 μm) and copper (54 μm) foils were inserted at the front and the back of the tantalum foil (9 μm for the 1st irradiation and 210 μm for the 2nd irradiation) in the stack respectively to confirm the beam intensity and the energy, and also to check the recommended data.

III. Data Analysis

The activities of the produced radionuclides were measured nondestructively by a high resolution HPGe γ-ray detector by coupling with a 4096 multi-channel analyzer. To identify the produced radioisotopes and to obtain the independent and "cumulative cross-sections", the activity of the sample was measured repeatedly after irradiation. Activity measurements were done at two distances (5 and 19 cm i.e. short and long) from the detector surface and after sufficient cooling time for the decay of most of undesired short lived activities to identify and separate complex gamma lines. The efficiency versus energy curve of the detector was determined experimentally using standard gamma-ray sources with known strength, ^{152}Eu, ^{133}Ba, ^{241}Am, ^{60}Co and ^{137}Cs at both of 5 cm and 19 cm from the detector surface. We also calculated the detector efficiency using the EGS4 Monte Carlo Code[10]. The experimental efficiencies agreed with theoretically obtained values. The proton energy degradation along the stack was determined using the computer program TRIM[11] and the uncertainties

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within 4.2%. In the present experiment, thin foils were used and the effect of proton energy broadening is negligible. Estimation of a photo peak area of gamma-ray spectra was done by using computer program for automatic analysis[12]. We measured the proton flux from top monitor foil and considered constant through the foil stack. The proton beam intensity was determined via the standard monitor reactions $^{27}Al(p,x)^{22,24}Na$ and $^{nat}Cu(p,x)^{56}Co,^{62,65}Zn$. The cross-sections were deduced for the production of $^{175,176,177,180}Ta$, $^{173,175}Hf$, ^{178}W and ^{179}Lu residual nucleus from 28 to 70 MeV bombarding energies using the well known activation formulae. The decay data for monitors and tantalum were taken from EXFOR[6] and ENSDF[13] respectively. We had to make coincidence–summing correction for some of the detected nuclides. The data were corrected for the coincidence-summing effect caused by the coincidence detection of two or more gamma-rays by using the SUMECC code[14]. We observed good agreement between the measured results at short and long distances from detector surface after coincidence-summing correction. The following errors were considered to derive total uncertainty on cross-section value: statistical error (0.3-10%), error deriving from the monitor flux (5%) and error in efficiency calibration depending on gamma ray energy (0.5-3%). The complete excitation functions of the monitor reactions are shown in **Figs.1** and **2**. From the above figures, it has been observed that the measured data for monitor reactions are in good agreement with the recommendation and literature data[15]. For $^{nat}Cu(p,x)^{58}Co$ reaction, presently no recommended data for cross section exist and the data of this study will play important role for evaluation. The good agreement of the data of the monitor reactions with the recommended values confirms the reliability of the determination of proton energy, beam intensity and cross-section.

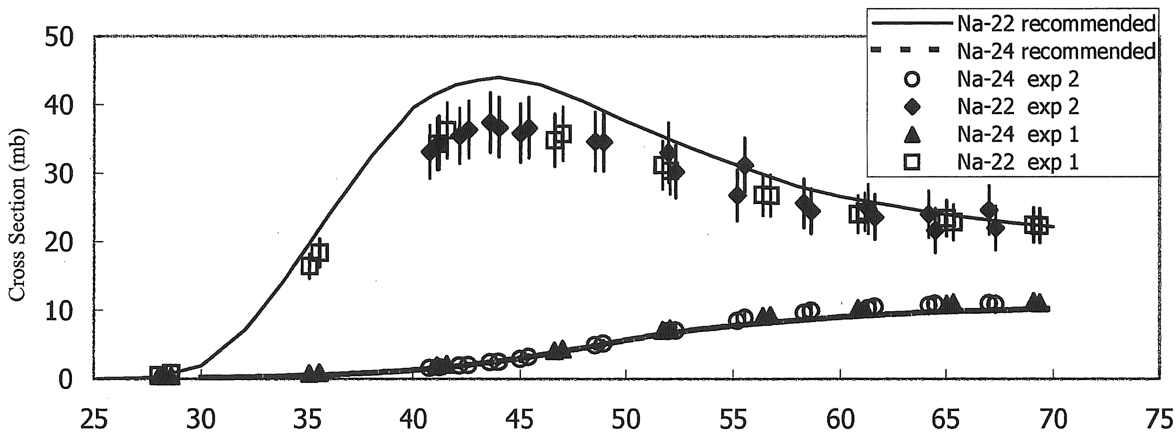

Fig.1 Excitation functions of the $^{27}Al(p,x)^{22,24}Na$ reactions.

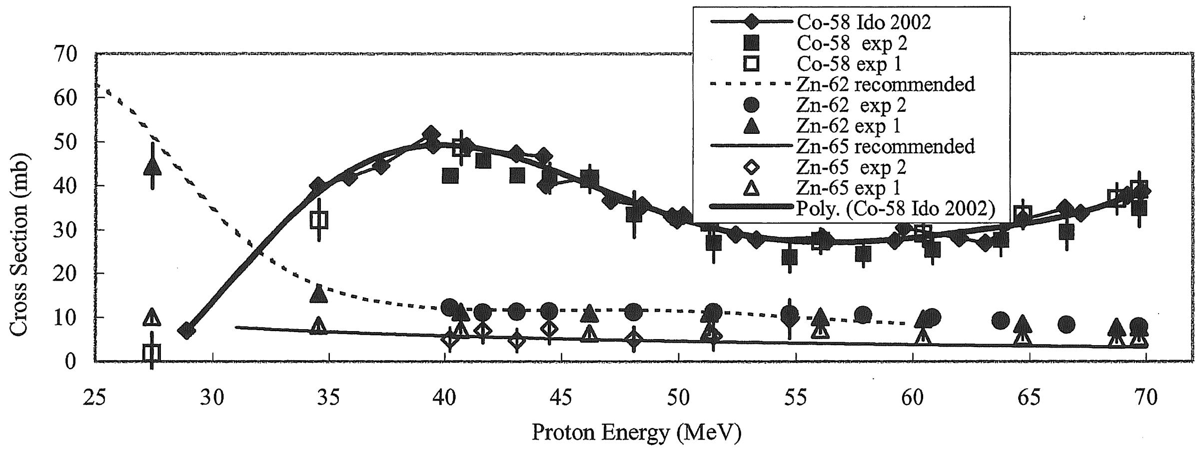

Fig.2 Excitation functions of the $^{nat}Cu(p,x)^{58}Co,^{62,65}Zn$ reactions.

IV. Results and Discussion

The measured excitation functions along with the available literature values and the theoretical values are shown in **Figs. 3** and **4**. The results of the selected reactions are described in the following subsequent sections.

1. $^{181}Ta(p,x)^{175,176,177,180}Ta$

The measured data of ^{175}T given in **Fig.3a** show the combined results of direct and "cumulative production cross-sections" that sharply increase with the increasing proton energy and consistent with C.L.Rao[1]. No literature data for ^{176}T production in our investigated energy range were reported yet. C.L.Rao[1] reported the direct formation cross-section of ^{177}Ta through chemical separation, but in our measured data the daughter, ^{177}Ta of parent, ^{177}W decay has also been considered along with direct formation. Therefore, the shape of the measured excitation function is similar to C.L.Rao[1]. For $^{181}Ta(p,x)^{180}Ta$ reaction, our measurement is the first one from 42 to 69 MeV. In this case the measured cross-section gradually decreases due to the decreasing importance of the direct interaction mechanism like the emission of deuterons by the well-known pickup process with increasing incident energy.

2. $^{181}Ta(p,x)^{173,175}Hf$

The ^{173}Hf nuclide is observed as a direct spallation product. The measured cross-sections of $^{181}Ta(p,x)^{173}Hf$ shown in **Fig.4a** are in good agreement with the model calculations at energies below 60 MeV. The probability of this reaction is largely inhibited by the Coulomb barrier in both the entrance and exit channels. ^{175}Hf is formed only directly below 42 MeV proton bombarding energies. At higher energy ^{175}Hf is formed through the decay of the parent ^{175}Ta. It has been possible to measure the ^{175}Ta formation cross-sections from 56 MeV in the present work cited earlier. Therefore the daughter ^{175}Hf of parent decay largely contributed on its direct formation and production cross-section shown in **Fig.4b** increases sharply from 56 MeV with increasing energy.

3. $^{181}Ta(p,x)^{179}Lu$

The measured excitation functions are shown in **Fig.4c** together with the results of model calculation compiled in MENDL[7]. No literature data have been reported over the investigated energy range. The calculated values are too low and do not fit the experimental data.

4. $^{181}Ta(p,x)^{178}W$

^{178}Ta (9.3 min half life) in secular equilibrium with ^{178}W has prominent γ-rays with energies 1341 keV, 1350 keV and 511 keV annihilation radiation. These lines decay with 22 day parent half-life and were used in cross-section determinations. The measured values are completely supporting the data of C.L.Rao[1] and Birattari[5], both in shape and maximum and that fact confirms the reliability of our data. At lower energies this reaction proceeds through the formation of a compound nucleus. With increasing of incident proton energy the excitation function goes through a maximum when the excitation energy is most appropriate for evaporating the requisite number of particles, then drops very rapidly as the energy becomes higher because at higher excitation energy it is much more likely that more particles will evaporate[16]. The tailing off of the excitation function at higher energies indicates that the nucleon-nucleon internal cascade mechanism becomes important[1,6].

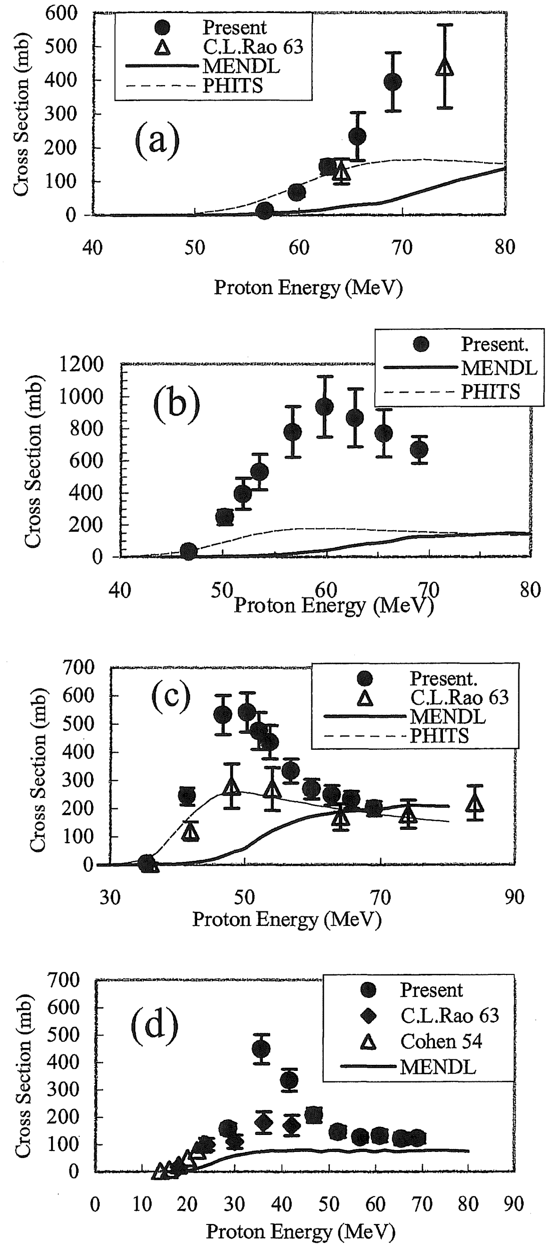

Fig.3 Excitation functions of the $^{181}Ta(p,x)$ reactions: a) $(p,x)^{175}Ta$; b) $(p,x)^{176}Ta$; c) $(p,x)^{177}Ta$; d) $(p,x)^{180}Ta$.

V. Conclusions

We have measured the excitation functions of the proton-induced activation reactions on tantalum in the energy range 28-70 MeV using the stacked foil technique. The relatively good agreement of the measured cross-sections of the monitor reactions with recommended data confirms the reliability of various techniques used in our experiment. New data were obtained for $^{181}Ta(p,x)^{176}Ta$, $^{181}Ta(p,x)^{179}Lu$ and above 42 MeV for $^{181}Ta(p,x)^{180}Ta$ reactions. The present experiment has given new data for all of the investigated reactions. Two experiments took place in the same experimental conditions and resulted in reasonable agreement. In most of the data points, our measurements support other experiments. For ^{175}Ta, ^{176}Ta, ^{177}Ta, ^{180}Ta and ^{175}Hf the single curves of theoretical calculation do not agree with our results, because the experimental data include cumulative production while the theoretical data are only direct. Besides, we have observed the contradictory results

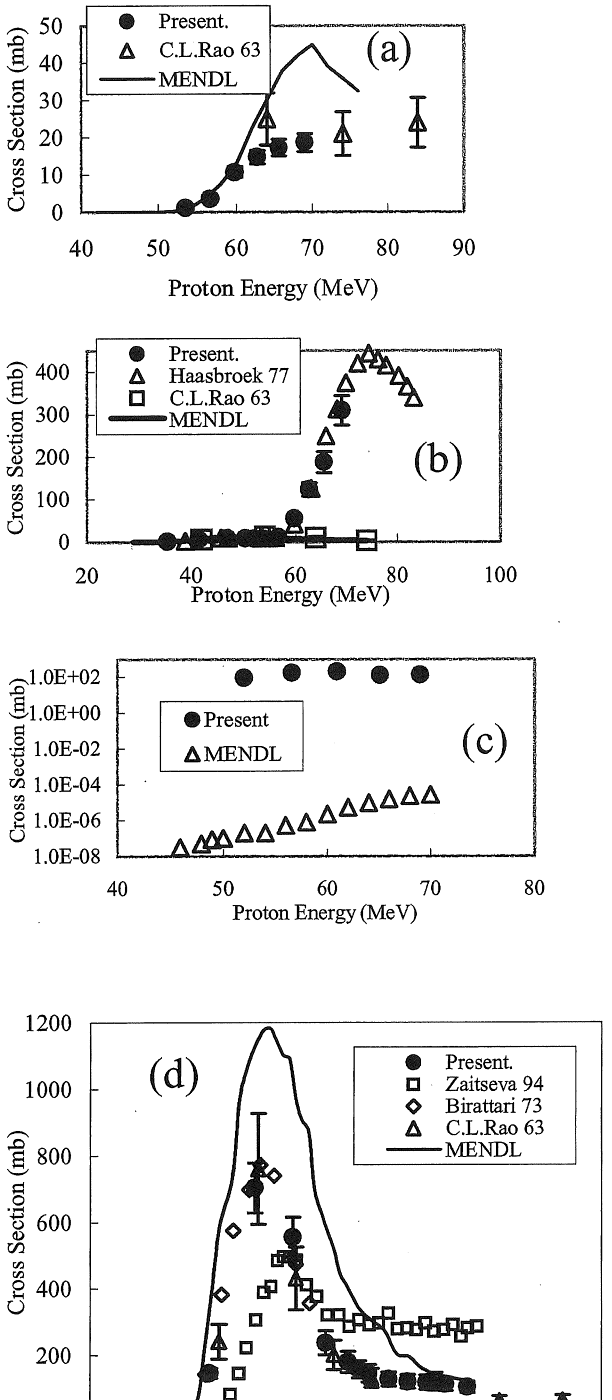

Fig.4 Excitation functions of the ^{181}Ta(p,x) reactions: a) (p,x)^{173}Hf; b) (p,x)^{175}Hf; c) (p,x)^{179}Lu; d) (p,x)^{178}W.

between MENDL and INC-GEM model calculations. The newly measured cross-sections will contribute to improve the statistical model code for reliable data calculations and allow to produce the recommended data for practical purposes.

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