Cyclotron production of ^{48}V via ^{nat}Ti(d,x)^{48}V nuclear reaction; a promising radionuclide

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Abstract. In this experimental work, we studied the excitation function of ^{nat}Ti(d,x)^{48}V nuclear reactions from 24 MeV down to threshold energy. Natural titanium foils were arranged in the popular stacked-foil method and activated with deuteron beam generated from an AVF cyclotron at RIKEN, Wako, Japan. The emitted γ activities from the activated foils were measured using an offline γ-ray spectrometry. The present results were analyzed, compared with earlier published experimental data and also with the evaluated data of Talys code. Our new measured data agree with some of the earlier reported experimental data while a partial agreement is found with the evaluated theoretical data. In addition to the use of ^{48}V as a beam intensity monitor, recent studies indicate its potentials as calibrating source in PET cameras and also as a (radioactive) label for medical applications. The results are also expected to further enrich the experimental database and also to play an important role in nuclear reactions model codes design.

1. Introduction

The use of short-lived positron emitting radionuclides in medical diagnostic procedures is nowadays a very well developed technique, most especially as imaging modality in positron emission tomography. In such procedures, the short-lived radionuclides, like ^{15}O ($T_{1/2}$= 2 min), ^{11}C ($T_{1/2}$ = 20.4 min) or ^{18}F ($T_{1/2}$ = 110 min), are usually labeled with pharmaceuticals of biological origin. By using these formed radiopharmaceuticals (e.g $^{11}CH_3I$, ^{18}FDG) with PET cameras, excellent images of heart, brain or tumors can be obtained for different kind of medical examinations. However, the short-lived nature of these positron emitters is usually a disadvantage when performing studies of slow biological processes [1]. The longer lived positron emitters (also known as non-standard positron emitters or Innovative PET radionuclides) have been reported as more suited to the study of slow metabolic activities and to labelling of organic compounds [2]. The decay properties of the longer-lived radionuclides play an important role in obtaining quality and volume of images as it allows ample time for image measurement during biological studies.

The usage of metallic titanium (Ti) in medical field is wide ranging. Since it has not been known to react with human body, titanium metal is widely used in several biological implants such as artificial hips and formation of pins for setting bones [3, 4]. On the other hand, charged particle bombardment of natural or enriched titanium isotopes produce several radionuclides useful in medical and biological fields. The production of ^{48}V via proton or deuteron has been reported by several studies. Vanadium-48 ($T_{1/2}$=15.9735 d), a positron emitting radionuclide, which decays to stable ^{48}Ti by EC (50.1%) and β^+(49.9%), find its acceptance as a monitor radioisotope mainly due to its decay characteristics and its relatively long half-life. The investigated radionuclide (^{48}V) is recently finding an increasing

potentials in different fields such as, from being radioactive tracer in biological processes [5] and material science [6] studies to its applicability in renal artery brachytherapy procedure [7]. Furthermore, ^{48}V was associated with some important biochemical characteristics such as anti-carcinogenic effect and thus tipped to be labeled with some compounds for *in vivo* studies [8]. In addition, a conducted study on 'alternative radionuclide' to the conventional use of ^{68}Ge ($T_{1/2}$ =271 d) in PET to further improve the quality of images has suggested the use of ^{48}V as a transmission source for correction of possible attenuation in the positron emission tomography technique [9]. Therefore, the need of precise measurement of excitation function for ^{48}V is very important.

An extensive survey on the reported experimental data on production of the beam monitoring and medically important ^{48}V via deuteron bombardment route indicated several studies are available. However, there exist discrepancies among the experimental literature, leading to the need of more measurements. In beam monitoring purpose and medical applications, high accuracy of radionuclide cross-sections are especially important and therefore the need for additional measurements are vital. Present study reports new experimental cross-sections of ^{48}V via deuteron bombardment on natural titanium foils.

2. Experimental setup and Analysis

Present study was performed using similar main experimental procedures to some of our previous works [10-14]. The irradiation of target foils was performed using the well-established stacked foil activation technique while measurements of activities of the ^{48}V was achieved via HPGe γ-ray spectrometry. The excitation function of ^{48}V radionuclide has been reported in the energy region of 2.5 to 23.8 MeV. Additional details relevant to present study are explained under the sub-sections below.

2.1 Targets, bombardment and activity measurements

Metallic titanium (20.32-μm thick; 99.99% purity; Goodfellow, UK) foil of natural isotopic abundance (^{46}Ti: 8.25 %; ^{47}Ti: 7.44 %; ^{48}Ti: 73.72 %; ^{49}Ti: 5.41 %; ^{50}Ti: 5.18 %) [15] was used as the primary target material. For beam monitoring and additional excitation functions measurements, several high purity (99.99% purity; Goodfellow, UK) natural metallic foils of nickel, and platinum were interleaved between the titanium foils in the stack. The target foils were weighed with high precision electronic balance for precise determination of foil thickness. A stack was prepared with uniform foil dimensions of 15 × 15 mm^2 following the size of the target holder to ensure proper focusing of the incident beam to center of the stack. The prepared stack was then held in a water-cooled target holder, which serves as a Faraday cup and then bombarded for 2.0 h with 24-MeV deuteron beam of characteristic average beam current of about 200 nA, from the AVF cyclotron of RIKEN RI Beam Factory. Through a tantalum slit in the particle exit channel of the cyclotron, the beam was collimated to 9-mm diameter onto the target foils. The whole processes of stack preparation, irradiation and activity measurements were carried out at the Nishina Center for Accelerator-Based Science, RIKEN, Wako, Saitama, Japan.
The emitted γ-rays were measured without any chemical separation using a high resolution HPGe γ-ray spectrometer (ORTEC; GEM-25185P; 55.1-mm crystal diameter and 52.0-mm thickness; operating voltage: +2000 V; relative efficiency: 25%) which was coupled to a 4096 multi-channel analyser and other associated electronics. A cooling period of 9 to 10 days were allowed before the measurements of this radionuclide based on its half-life so as to ensure interference free cross-sections. The measurements were repeated several times at different source-to-detector distances. Similarly, all measurements were done with consideration of dead time less than 10 % by adjusting the source to detector distances. The gamma analysis of the spectra was done using the Maestro (Ver. 7.01; ORTEC) gamma vision program.

The efficiencies of the detector used at various source-to-detector distances were evaluated using multi-nuclide γ-ray standard source obtained from DBA Isotopes Products Laboratories (USA). Further details of major procedure involved in the calculations of the efficiencies at various source-to-distances were earlier reported in our similar studies [14].

2.2 Determination of beam intensity, foil energies and cross-sections

The beam intensity was determined from measured activity of $^{nat}Ni(d,x)^{61}Cu$ from the front foil facing the beam, using the IAEA [16] $^{nat}Ni(d,x)^{61}Cu$ recommended monitor reaction cross-section. The intensity was considered as a constant in the stack and was used to deduce cross-sections for each foil in the stack. The uniformity of the deuteron beam intensity along the irradiated foils was further ascertained by calculating cross-sections of ^{61}Cu from the other Ti foils in the stack, and comparing with the IAEA [16] recommended values. The presence of other metallic foils of different densities and thicknesses helped in slowing down the incident deuteron beam along the stack. The degradation of the initial 24 MeV deuteron beam energy along the stacked foils was calculated using a computer program, SRIM-2003 software [17]. The average deuteron-energy on each foil was solely dependent on the foil position in the stack.

Taking into account some of the target parameters such as density, thickness and the incident bombarding particles, the cross-sections, σ (E), of the assessed radionuclide were computed using the well-known activation formula [10, 14].

Similarly, we adopted decay data of the investigated reaction products from the ENSDF library [18] retrieved via the interface of Live Chart of nuclides [19]. The Q-values and threshold energies were calculated based on the AME mass evaluation [20], extracted through the Q-tool system [21] and they are as well presented in the Table 1.

Table 1. Decay data of the assessed ^{48}V.

Nuclide	Half-life	Decay mode (%)	E_γ (keV)	I_γ (%)	Contributing reactions	Q-Value (MeV)	Threshold (MeV)
^{48}V	15.9735 d	$\varepsilon+\beta^+$:100	944.130	7.870 *7*	$^{47}Ti(d,n)^{48}V$	4.6	0.0
			983.525	99.98 4	$^{48}Ti(d,2n)^{48}V$	-7.0	7.3
			1312.106	98.2 3	$^{49}Ti(d,3n)^{48}V$	-15.2	15.8

The major sources of uncertainties in our experiment were considered during the cross-section calculation and are summarized in Table 2. An overall uncertainty in present cross-sections in the range of 0.01 - 22.81% was obtained by quadratic summing of the individual uncertainties listed in the Table 2.

Table 2: Evaluated uncertainties on cross-sections calculations.

Nuclide	Uncertainties (%)					
	γ-ray intensity (ΔIγ/ Iγ)	beam intensity	detector efficiency	thickness of target	γ-ray counting statistics	total uncertainty
^{48}V	0.040	5	4	2	0.3 - 5.9	0.01 - 22.81

3. Results and discussion

The production cross-sections of the ^{48}V radionuclide are tabulated in Table 1 while its excitation function is plotted in Fig. 1 together with earlier reported experimental measurements obtained from the EXFOR library [22] and the evaluated data in the TENDL-2014 library [23] which provides the output of the TALYS code [24]. In some cases, isotopic cross-sections reported in some earlier measurements [25-27] were normalized by multiplying with their natural isotopic abundance for comparison with our elemental cross-sections on the Figure. Similarly, the reported work in [28] was normalized based on the recommendation of the authors in their latter publication [29].

3.1. Production of independent ^{48}V

In addition to ^{48}V, there is also ^{48}Sc which was simultaneously produced from the deuteron irradiated Ti foils. Concurrently, both ^{48}V ($T_{1/2}$ = 15.9735 d) and the ^{48}Sc ($T_{1/2}$ =43.67 h,) decays to the same

stable ^{48}Ti nuclide, and thus have some shared gamma lines as indicated in Table 2. There are three possible ways to assess independent ^{48}V production cross-sections. First, it is possible to use any of the interfering gamma line of ^{48}Sc with ^{48}V for the measurement of cross-sections of the ^{48}V but a computational technique is necessary in order to latter isolate the independent ^{48}V contributions in the calculated cross-sections. This is usually technical and was thus avoided. The second possibility is to use the independent gamma line of the ^{48}V (E_γ =944.130 keV), although it has a relatively low intensity (I_γ = 7.870 %). In the third method, since ^{48}V has longer half-life compared to ^{48}Sc, a long cooling period is usually allowed before the measurement of ^{48}V, sufficient enough until the interfering ^{48}Sc radionuclide completely decay. Thus, a cooling period of at least 10 half-lives of ^{48}Sc ($T_{1/2}$ =43.67 h) is usually needed for it to completely decay, that is about 450 hours from end of bombardment of the foil. This last procedure was followed in the present study since our measurements were made 9 to 10 days from the end of irradiation.

The major nuclear reactions contributing to the formation of ^{48}V are listed in Table 1. There are several experimental studies for comparison. Figure 1 compared the present studies with some of the literature experimental data [25-34] and also with IAEA recommended data. The Figure also compared our results with the extracted data of TENDL-2014 library, which was calculated based on TALYS code. Our data agree with the recommended data of IAEA but not with some few of the experimental literature data. On the other hand, the TENDL-2014 evaluated cross-sections have overestimated the present measurement.

Table 3: Measured cross-sections for ^{nat}Ti(d,x)^{48}V

Energy (MeV)	Cross-section (mb)
23.8 ± 0.5	224.8 ± 15.2
22.3 ± 0.5	261.6 ± 17.6
20.4 ± 0.5	308.0 ± 20.7
18.6 ± 0.5	336.4 ± 22.6
16.8 ± 0.6	339.0 ± 22.8
12.8 ± 0.6	271.1 ± 18.2
6.3 ± 0.8	21.6 ± 1.5
4.6 ± 0.9	16.7 ± 1.1
2.50 ± 1.50	0.80 ± 0.01

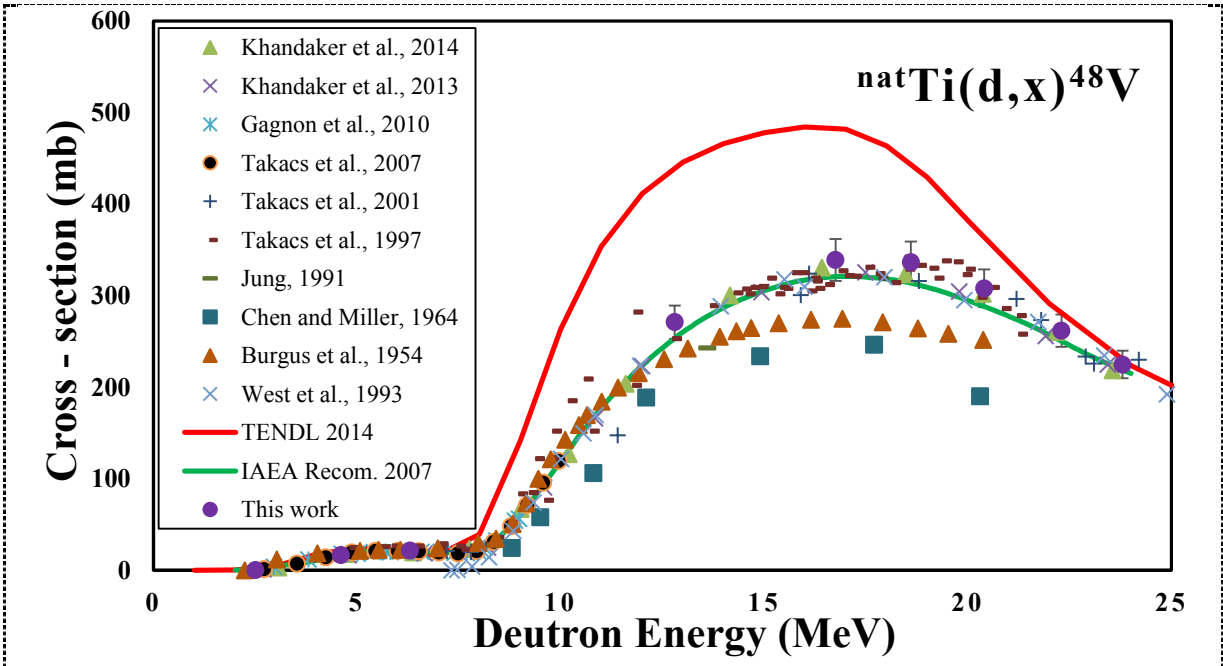

Figure 1. Excitation function of ^{nat}Ti(d,x)^{48}V reaction.

4. Conclusion

This work reports new experimental cross-sections for the ^{nat}Ti(d,x)^{48}V reaction in the energy region of 2.5 – 23.8 MeV. The measured excitation function was compared with the available experimental literature data and with the theoretical data extracted from the TENDL-2014 library. The new cross-sections confirm the previous available data in the EXFOR database with some additional insight. A large discrepancy was found between the present experiment cross-sections and the theoretical data of the TENDL-2014 library, which was based on Talys code. Present result could play an important role for improvements of prediction ability of the TALYS code.

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