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Investigation of the ^{27}Al(d,x)^{24}Na nuclear reaction for deuteron beam monitoring purpose

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Abstract. Activation cross-sections for the ^{27}Al(d,x)^{24}Na nuclear reaction was measured by using a stacked-foil activation technique combined with high purity germanium (HPGe) γ-ray spectrometry over deuteron energy range of 2–24 MeV. Measured data were critically compared with the available literature data and also with the theoretical data extracted from the TENDL data base. Accuracy of the ^{27}Al(d,x)^{24}Na cross-sections were confirmed by the simultaneous measurements of the ^{nat}Ti(d,x)^{48}V monitor reaction cross-sections. Present results reproduced well the IAEA recommended ^{nat}Ti(d,x)^{48}V reaction cross-sections, but provide slight deviation with the IAEA recommended ^{27}Al(d,x)^{24}Na cross-sections. It may be concluded that the use of ^{27}Al(d,x)^{24}Na in deuteron beam monitoring should not be a perfect choice if one has the option to use the ^{nat}Ti(d,x)^{48}V reaction.

1. Introduction

Nowadays, production of medical radionuclides has become a very important feature of nuclear technology. Precise knowledge on nuclear reaction cross-sections is not only playing a vital role in their production but also in various practical applications such as power generation, radiation dosimetry, development of nuclear reaction model code, thin layer analysis for wear measurement, basic nuclear physics as well as astrophysics. In principle, neutron or charged-particle induced nuclear reactions are used as the pathways for radionuclides production. But, the status of charged-particle induced reaction cross-sections are scanty relative to the neutron induced one, and such activities is in the developing stage. Moreover, accuracy of charged-particle induced nuclear reaction cross-sections largely depends on the precise determination of the accelerated particle beam parameters such as beam intensity. Monitoring of beam intensity can be done in several ways. The integrated beam current obtained via Faraday cup is sometimes not possible and/or not accurate enough. However, some nuclear reactions could be used with a high accuracy to monitor beam parameters. Understanding the importance of beam parameter monitoring in activation experiments, a number of reactions are recommended by the International Atomic Energy Agency (IAEA) via a coordinated research project [1]. In a number of earlier efforts, we already verified and/or reproduced [2,3] the IAEA recommended widely used ^{nat}Ti(d,x)^{48}V monitor reaction. In connection to this, we would like to verify IAEA recommended another low energy beam monitoring reaction ^{27}Al(d,x)^{24}Na. Thus, present work concerns the simultaneous activation of both ^{nat}Ti and ^{27}Al monitor foils by a deuteron beam with 24 MeV energy in order to verify the IAEA recommended ^{27}Al(d,x)^{24}Na cross-sections for beam monitoring purpose.

2. Experimental details

Present study was performed using a similar experimental procedure to our earlier works [4,5]. A well-established stacked foil activation technique combined with HPGe γ-ray spectrometry was used herein. Some specific information relevant to the present work is given as follows:

The whole process of sample preparation, irradiation and activity measurements were conducted at the Nishina Center for Accelerator-Based Science, RIKEN, Wako, Saitama, Japan. Metallic foils of samarium (24.45 μm and 26 μm; 99% purity; Goodfellow, UK; Lot no.: LS 433954 LO) with natural isotopic composition were used as the main target material in this experiment. A high purity natural titanium foil (20.50-μm thick; 99.99% purity; Nilaco, Japan; Lot no.: 453213) and aluminum foil (24.56 μm; 99.999% purity; Goodfellow, UK; lot no.: LS415355 VCS) was used for beam monitoring and beam energy degradation purposes. The thickness of the foils were determined using a typical formula: Thickness = (Weight of the sample)/(Density of sample $\times$ Area of the foil). All of the used foils were weighed with high precision electronic balance to obtain precise mass. The samples were then cut into small pieces with a dimension of 15 $\times$ 15 mm^2 following the size of the target holder. A typical stack was then prepared by placing the titanium and aluminium foils in between any two samarium foils in a regular fashion so that the full beam energy is absorbed within the stacked foil. The prepared stack was then kept

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Table 1. Decay data of the investigated radionuclides ^{24}Na and ^{48}V; bold γ-lines were used in activity determination.

Radio-nuclide	Decay Mode (%)	γ-energy, Eγ (keV)	γ-intensity, Iγ (%)	Contributing reaction	Thresh-old (MeV)
^{24g}Na (14.997 h)	β- (99.995)	**1368.626** 2754.007	99.9936 *15* 99.855 *5*	^{27}Al(d, p+α) IT decay of ^{24m}Na	5.76
Beam monitoring reaction					
^{48}V (15.9735 d)	β^+(49.9); EC(50.1)	944.130 **983.525** 1312.106	7.870 *7* **99.98** ***4*** 98.2 *3*	^{46}Ti(d,n) ^{47}Ti(d,2n) ^{48}Ti(d,3n)	0.0 7.3 15.8

in a target holder, which serves as a Faraday cup to obtain integrated beam currents/charges after irradiation. The exit point/channel of the beam line of the AVF cyclotron was collimated to 9-mm diameter through a tantalum slit so that the target receive the full beam energy properly. The stacked foils were then irradiated by a 24-MeV deuteron beam extracted from the AVF cyclotron of RIKEN, Wako, Japan for a period of 2.0 h with an average beam current of ∼120 nA. After a sufficient cooling time of about ∼2 hours, the stack was removed from the target holder, and made ready for an offline gamma-ray spectrometry.

The emitted γ-rays were measured non-destructively 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. The measurements were repeated for several times at different source-to-detector distances following a maximum dead time of <10%. A Maestro (Ver. 7.01; ORTEC) gamma vision computer program was used to analyze the acquired gamma-ray spectra. The efficiencies of the detector for each measured distance were evaluated using a multi-nuclide γ-ray point source obtained from the DBA Isotopes Products Laboratory (USA). Further details involved in the calculations of efficiencies are reported in our earlier studies [2,4].

The degradation of deuteron energy along the stacked foils was calculated by using a computer program STACK based on the energy-range relation described by Williamson et al. [6]. The mean deuteron-energy on each foil in the stack was solely depends on the foil position in the stack. Starting with the initial deuteron energy incident on the front foil of the stack, the deuteron energy (E) effective at each sample was obtained taking a mean of the ingoing and outgoing particle energies. The ^{nat}Ti(d,x)^{48}V (E_d = 23.88 MeV, σ = 217.54 mb) [1] monitor reaction recommended by the IAEA was used to determine the final beam current. The beam intensity was determined from the activity of the Ti foil placed at the front position of the stack, and considered as a constant to deduce the cross-sections for each foil in the stack. The cross-sections were determined using a well-known activation formula [7,8]. The decay data of the investigated reaction products were retrieved from the NuDat-2.6 interface [9], and they are summarized in Table 1. The Q-values and threshold energies were calculated by the Q-tool system [10], and they are also presented in Table 1.

The uncertainty in the deuteron energy is due to the uncertainties in the initial beam energy (∼1%), the target thickness (∼1%), and the beam straggling. Specifically, the uncertainty in the primary beam energy was estimated to be ±0.24 MeV, but due to the straggling effect and the finite foil thickness, it increased gradually along the stack and reached to ±0.92 MeV at the last foil. Thus, the estimated uncertainties (and/or beam spreading) of a representing energy point ranged from ±0.69 MeV to ±1.16 MeV, and they are shown in tables and figures. On the other hand, the major sources of uncertainties considered in the cross-sections are as follows: γ-ray counting statistics (0.43–1.35%), beam flux (∼5%), detector efficiency (∼4%), sample thickness (∼1%), and γ-ray intensity ($\Delta I\gamma/I\gamma$ =0.002%). All of the uncertainties were considered as independent, and consequently, they were quadratically added to obtain the total uncertainty which is in the range of 6.5–6.6%.

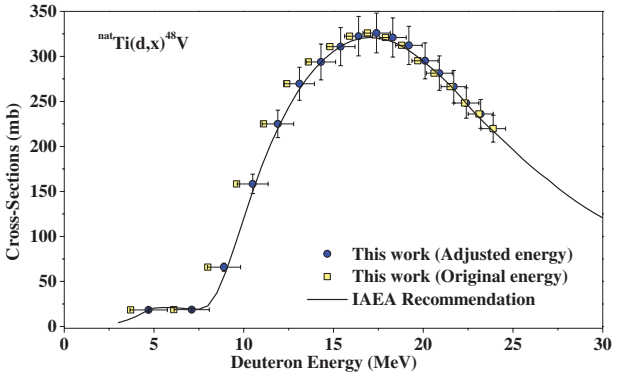

Figure 1. Excitation function of the ^{nat}Ti(d,x)^{48}V monitor reactions together with the IAEA recommended cross-sections. The mean deuteron energy for each foil in the stack was adjusted using a correction factor of 0.02 which is cumulatively increased from the first foil to the last foil.

3. Results and discussion

The deduced beam current via the ^{nat}Ti(d,x)^{48}V monitor reaction was compared with the direct beam current obtained via Farady cup and/or accelerator settings, and found about 4.16% higher value. Measurements of cross-sections in this work is based on the obtained beam current from the ^{nat}Ti(d,x)^{48}V monitor reaction. Figure 1 shows the excitation function of the ^{nat}Ti(d,x)^{48}V monitor reaction together with the IAEA recommended cross-sections. It was observed that the IAEA recommended cross-sections are slightly deviated in the downward energy region. Thus, the calculated energy via STACK program was adjusted following the IAEA recommended ^{nat}Ti(d,x)^{48}V

Table 2. Measured cross-sections of the ^{24}Na radionuclide under this study.

Deuteron energy (MeV)	Energy error (MeV)	Cross-Sections (mb)	Cross-Sections error (mb)
23.4	0.69	59.7	4.1
22.6	0.69	59.4	4.0
21.9	0.70	58.0	3.9
21.1	0.70	55.3	3.8
20.3	0.71	52.7	3.6
19.5	0.72	47.6	3.2
18.6	0.73	42.0	2.9
17.7	0.74	35.6	2.4
16.7	0.75	28.3	1.9
15.7	0.77	20.6	1.4
14.6	0.78	12.74	0.87
13.5	0.80	6.15	0.42
12.2	0.83	1.91	0.13
10.9	0.87	0.37	0.03
9.4	0.92	0.161	0.011
7.6	1.00	0.150	0.010
5.5	1.16	0.151	0.010
3.4	1.36	0.143	0.009

monitor reaction cross-sections in the horizontal scale. A correction factor of 0.02 was used for this adjustment which was cumulatively increased from the first foil to the last foil in the stacked samples. The production cross-sections of ^{24}Na radionuclide is presented in Table 2 while the excitation function is plotted in Fig. 2 together with the earlier reported experimental data obtained from the EXFOR library [11] and the evaluated data taken from the TENDL-2015 library [12]. Error-bars are omitted for the experimental data taken from the EXFOR library for better visibility. Furthermore, the recommended cross-sections by Takacs et al. [13] for ^{27}Al(d,x)^{24}Na reaction is also potted in Fig. 2 for comparison.

The population of the ground state ^{24g}Na ($T_{1/2}$ = 14.997 h) is contributed by the direct reactions presented in Table 1, and by an IT decay (b_{IT} = 99.95%) of its short-lived isomeric state ^{24m}Na ($T_{1/2}$ =20.18 ms) within our investigated energy region. Therefore, the measured production cross-sections are regarded as the sum of the ground and meta-stable states, $^{24(g+m)}$Na. The neutron rich ^{24g}Na decays to the stable ^{24}Mg via an emission of β^- particle (b_{β^-} = 99.995%) followed by the emission of 1368.626 keV γ-line (I_γ = 99.9936%) and 2754.007 keV (I_γ = 99.855%) γ-lines. The 2754.007 keV γ-line lies outside the detector calibration range, thus the intense 1368.626 keV γ-line released from the first excited state of ^{24}Mg was used to determine the cross-sections of ^{24g}Na radionuclide.

Numerous authors [13–27] have already investigated the ^{27}Al(d,x)^{24}Na reaction due to its importance in the beam monitoring applications. Figure 2 shows an overall good agreement of the present results with the earlier measurements by Zhao et al., Lebeda et al., Hagiwara et al., Bem et al., Roehm et al., Michel et al. The data reported by Tao et al., Hermanne et al., and Takacs et al. show only a partial agreement to our measured data, while the measured data by Lenk & Slobodrian, Martens & Schweimer and Batzel et al. show quite discrepant results to all other measurements. The cross-sections reported by Watson et al. are slightly higher than other experimental data. The recommended cross-sections by Takacs et al. is in agreement up to 17 MeV, and then show a systematically lower values. The IAEA recommended cross-sections show systematically lower values almost in the whole energy region. The TENDL-2015 library reproduces a similar shape of the excitation function but slightly overestimates its magnitudes in the energy region above 15 MeV.

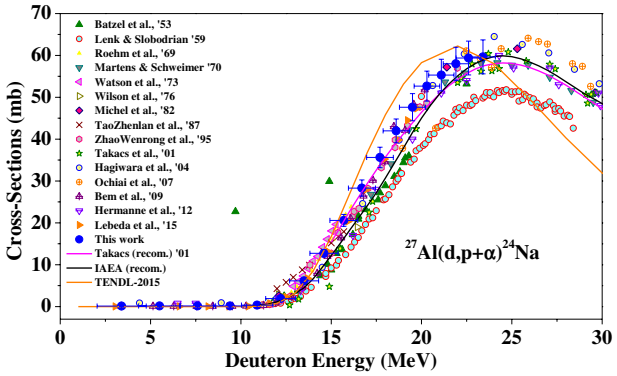

Figure 2. Excitation function for the ^{27}Al(d,x)^{24}Na nuclear reaction.

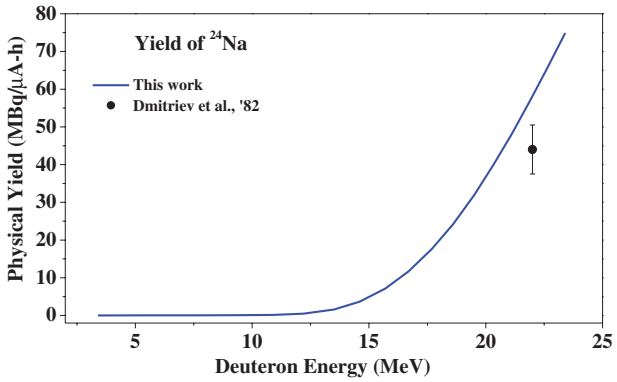

Figure 3. Physical thick target yields for the ^{24}Na radionuclide from ^{27}Al target.

Physical thick target yields [28,29] for the ^{24}Na radionuclide were deduced using the measured cross-sections and the stopping power of the Al target over the energy range from 3 to 24 MeV taking into account that the total energy is absorbed in the target. It is expressed as MBq-μA^{-1}.h^{-1}, i.e. the activity at the end of a bombardment performed at a constant 1 μA beam current on a target during 1 hour. The deduced yield for the production of ^{24}Na radionuclide is plotted in Fig. 3 as a function of deuteron energy. Dmitriev et al. [30] reported the yields for ^{24}Na by irradiating a thick aluminium target with a 22-MeV deuteron beam. Figure 3 shows that the yields reported by Dmitriev et al. for ^{24}Na at 22 MeV is smaller than our deduced values. Due to the lack of literature, we were unable to compare with any other data of directly measured thick target yields for ^{24}Na radionuclide.

4. Conclusions

Activation cross-sections of the ^{27}Al(d,x)^{24}Na reaction were measured in the energy range of 3–24 MeV using a stacked-foil activation technique with an overall

uncertainty of better than 7%. The cross-sections were normalized by using the ^{nat}Ti(d,x)^{48}V monitor cross-sections recommended by the IAEA. Measured data were critically compared with the available literature data and found an overall good agreement with some of the earlier measurements, whereas other shows a partial agreement. The IAEA recommended cross-sections for ^{27}Al(d,x)^{24}Na nuclear reaction was verified by the present investigation. The IAEA recommended cross-sections for ^{27}Al(d,x)^{24}Na reaction show systematically lower values in the whole energy region, while the extracted data from the TENDL-2015 show slightly higher values than our measured data. It is suggested that the use of IAEA recommended ^{27}Al(d,x)^{24}Na cross-sections should not be a perfect choice and/or it demands a revision to be used as beam monitoring process.

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