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Activation cross sections of deuteron-induced reactions on niobium up to 24 MeV

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Keyword

Molybdenum-93m; Deuteron irradiation; Niobium target; Cross Section; Excitation function

Abstract

We measured cross sections of deuteron-induced reactions on a niobium target up to 24 MeV using the stacked-foil activation technique and off-line γ -ray spectrometry. The production cross sections of $^{93\text{m}}\text{Mo}$, $^{92\text{m}}\text{Nb}$ and ^{89}Zr were determined. The results were compared with the experimental data studied earlier and the predictions of theoretical calculations.

1. Introduction

There are several radioisotopes of Mo, Nb and Zr suitable for medical use.. One of the potential radioisotopes for nuclear medicine among these elements is $^{93\text{m}}\text{Mo}$ [1]. This radioisotope has features of a moderate half-life ($T_{1/2} = 6.85$ h), γ -lines ($E_\gamma = 263.049$, 684.693 and 1477.138 keV with $I_\gamma = 57.4$, 99.9 and 99.1%, respectively), conversion electrons ($E_e = 243.050$ keV with $I_e = 29.7\%$) and Auger electrons ($E_e = 2.27$ keV with $I_e = 37.9\%$) [2], which were summarized in NuDat 2.7 [3]. With these decay parameters, $^{93\text{m}}\text{Mo}$ has great potential for use in medical diagnosis and/or therapy.

Several reactions to produce $^{93\text{m}}\text{Mo}$ were studied earlier, such as proton- (see e.g. [4] and references therein) and deuteron-induced reactions on Nb [5][6][7][8], α -induced reactions on Zr [9] and ^7Li -induced reactions on Y [10][11]. The experimental literature was surveyed in the EXFOR database [12]. In this paper, we focused on the deuteron-induced reactions on Nb because the cross sections are about four times larger than those of the proton-induced reactions [8]. In addition, the $^{93\text{m}}\text{Mo}$ production cross sections at around the peak energy of 17 MeV are scattered over several tens of mb [6][7]. Therefore, in order to obtain an accurate and smooth curve as a function of projectile energy, an experiment was performed to measure the cross sections up to 24 MeV. In the experiment, we could obtain production cross sections of $^{93\text{m}}\text{Mo}$, $^{92\text{m}}\text{Nb}$ and ^{89}Zr . Based on the result, the integral yield of $^{93\text{m}}\text{Mo}$ was also deduced.

2. Method

We used the standard methods, such as the stacked-foil activation method and off-line γ -ray spectrometry. The stacked target was composed of ^{93}Nb (99.9% purity, Nilaco Corp., Japan) and $^{\text{nat}}\text{Ti}$ (99.6% purity, Nilaco Corp., Japan) foils for monitoring the beam parameters. Niobium is a mono isotopic element, ^{93}Nb . The thicknesses of the Nb and Ti foils were 27.11 and 9.13 mg/cm², respectively, which were estimated from measurement of surface area and weight of larger sheets. After the thickness measurement, we cut the large sheets into 10 × 10 mm² rectangular pieces to fit into our target holder.

The target was irradiated by a 23.6 MeV deuteron beam at the AVF cyclotron of the RIKEN RI Beam Factory. The irradiation lasted for 30 minutes with an average intensity of 200.3 nA. The intensity was measured by a Faraday cup. The incident energy was 23.6 ± 0.1 MeV, which was determined by the time-of-flight method using a plastic scintillator monitor [13]. The degradation of the beam energy in the stacked target was calculated using the SRIM code [14]. The propagation of the initial energy uncertainty is also estimated through the target. These beam parameters can be assessed by the $^{\text{nat}}\text{Ti}(\text{d},\text{x})^{48}\text{V}$ monitor reaction.

The irradiated foils were separated from each other shortly after the end of bombardment. The γ -lines from the foils were measured by a high-resolution HPGe detector (ORTEC GMX-25190-P) and analyzed by dedicated software, Gamma Studio (SEIKO EG&G). The detector was calibrated using a multiple standard γ -ray point source consisting of $^{57,60}\text{Co}$, ^{88}Y , ^{109}Cd , ^{113}Sn , ^{137}Cs , ^{139}Ce and ^{241}Am radioisotopes. The reaction and decay data used for the γ -ray spectrometry were taken from NuDat 2.7 [3], LiveChart [15] and QCalc [16] (Table 1).

We derived the cross sections of the $^{\text{nat}}\text{Ti}(\text{d},\text{x})^{48}\text{V}$ monitor reaction and compared the result with the IAEA recommended values [17] to assess the beam parameters. The cross sections were obtained from measurements of the 983.5-keV γ ray (I_{γ} = 99.98%) emitted from the ^{48}V decay ($T_{1/2}$ = 15.97 d) after an adequate cooling time for decay of the co-produced ^{48}Sc ($T_{1/2}$ = 43.67 h). The result of the comparison with the IAEA recommended values is shown in Fig. 1, which shows good agreement without any adjustments. According to the comparison, the correctness of the measured beam intensity and the energy calculation in the targets was guaranteed.

Table 1: Reactions and decay data for reaction products

Nuclide	Half-life	Decay mode (%)	E_γ (keV)	I_γ (%)	Contributing reactions	Q-value (MeV)	Refs.
$^{93\text{m}}\text{Mo}$	6.85 h	IT (99.88) β^- (0.12)	263.049	57.4(11)	$^{93}\text{Nb}(\text{d},2\text{n})$	-3.4	[2]
$^{92\text{m}}\text{Nb}$	10.15 d	$\epsilon+\beta^+$ (100)	934.44	99.15(4)	$^{93}\text{Nb}(\text{d},\text{t})$	-2.6	[18]
^{89}Zr	78.41 h	$\epsilon+\beta^+$ (100)	909.15	99.04(3)	$^{93}\text{Nb}(\text{d},\alpha 2\text{n})$	-7.8	[19]
^{48}V	15.97 d		983.5	99.98(4)	$^{\text{nat}}\text{Ti}(\text{d},\text{x})$		[20]

Fig. 1. Excitation function of the $^{\text{nat}}\text{Ti}(\text{d},\text{x})^{48}\text{V}$ monitor reaction compared with the recommended values.

3. Result and Discussion

The derived cross sections are presented in Table 2 and graphically in Figs. 2-4. Only direct reactions in Table 2 are energetically possible for the three products and hence the reported cross sections are independent. The results are compared with the data measured earlier [5][6][7] and the TENDL-2017 data [21]. The data in the literature [8] were excluded because their energy range of 30-50 MeV was not overlapped with our data.

The estimated total uncertainty of the cross sections was 8.0-24.4%. It was the square root of the quadratic summation of each component; statistical uncertainty (<23.1%), target thickness (1%), target purity (1%), beam intensity (5%), detector efficiency (5%), γ -intensity (<1.1%) and peak fitting (3%).

Table 2. Measured cross sections

Energy (MeV)	$^{93}\text{Nb}(\text{d},\text{n})^{93\text{m}}\text{Mo}$ (mb)	$^{93}\text{Nb}(\text{d},\text{x})^{92\text{m}}\text{Nb}$ (mb)	$^{93}\text{Nb}(\text{d},\text{x})^{89}\text{Zr}$ (mb)
22.7 $\pm$ 0.4	64.5 $\pm$ 5.3	53.9 $\pm$ 4.4	9.1 $\pm$ 0.7
22.1 $\pm$ 0.4	70.6 $\pm$ 5.8	45.6 $\pm$ 3.8	6.4 $\pm$ 0.6
21.5 $\pm$ 0.4	81.5 $\pm$ 6.7	41.2 $\pm$ 3.4	4.5 $\pm$ 0.4
19.6 $\pm$ 0.4	100.3 $\pm$ 8.2	24.0 $\pm$ 2.0	0.83 $\pm$ 0.11
19.0 $\pm$ 0.5	108.3 $\pm$ 8.9	16.4 $\pm$ 1.4	0.36 $\pm$ 0.09
18.3 $\pm$ 0.5	114.4 $\pm$ 9.4	13.9 $\pm$ 1.2	
16.2 $\pm$ 0.5	113.2 $\pm$ 9.3	7.0 $\pm$ 0.6	
15.4 $\pm$ 0.5	107.3 $\pm$ 8.8	6.5 $\pm$ 0.6	
14.6 $\pm$ 0.6	98.6 $\pm$ 8.1	5.5 $\pm$ 0.5	
12.1 $\pm$ 0.6	62.8 $\pm$ 5.1	3.9 $\pm$ 0.4	
11.2 $\pm$ 0.7	48.7 $\pm$ 4.0	3.3 $\pm$ 0.3	
10.2 $\pm$ 0.7	33.6 $\pm$ 2.7	2.4 $\pm$ 0.3	
6.8 $\pm$ 0.9	0.94 $\pm$ 0.08		
5.3 $\pm$ 1.1	0.016 $\pm$ 0.001		
3.5 $\pm$ 1.4	0.0038 $\pm$ 0.0003		

3.1 The $^{93}\text{Nb}(d,2n)^{93\text{m}}\text{Mo}$ Reaction

The γ -line at 263.049 keV ($I_\gamma = 57.4\%$) from the $^{93\text{m}}\text{Mo}$ IT decay ($T_{1/2} = 6.85$ h) was measured after a cooling time of about 10 hours. The derived excitation function of the $^{93}\text{Nb}(d,2n)^{93\text{m}}\text{Mo}$ reaction is shown in Fig. 2 together with the earlier experimental data [5][6][7] and the TENDL-2017 data [21]. Our result shows a very smooth curve and good agreement with the other experimental data in the whole energy region. The theoretical calculation overestimates the experimental data.

Fig. 2. The excitation function of the $^{93}\text{Nb}(d,2n)^{93\text{m}}\text{Mo}$ reaction.

3.2 The $^{93}\text{Nb}(d,x)^{92m}\text{Nb}$ Reaction

The excitation function of the $^{93}\text{Nb}(d,x)^{92m}\text{Nb}$ reaction was determined from measurements of the γ -line at 934.44 keV ($I_\gamma = 99.15\%$) from the ^{92m}Nb decay ($T_{1/2} = 10.15$ d). The count of the peak was obtained after a cooling time of 32 days. The result is shown in Fig. 3 together with the experimental data measured earlier [6][7] and the calculation data [21]. Our result is in good agreement with the other experimental data. The TENDL-2017 data are consistent with the experimental data below 15 MeV, however are larger than our experimental data above 15 MeV.

Fig. 3. The excitation function of the $^{93}\text{Nb}(d,x)^{92m}\text{Nb}$ reaction.

3.3 The $^{93}\text{Nb}(d,x)^{89}\text{Zr}$ Reaction

The reaction product ^{89}Zr ($T_{1/2} = 78.41$ h) emits the γ -ray of 909.15 keV ($I_\gamma = 99.04\%$). The measurements of the intense γ -line were performed about 24 hours after the end of irradiation. The derived cross sections of the $^{93}\text{Nb}(d,x)^{89}\text{Zr}$ reaction are shown in Fig. 4 with experimental data in the literature [6][7] and the TENDL-2017 data [21]. In the whole energy region as shown in the figure, the data acceptably agree with each other.

Fig. 4. The excitation function of the $^{93}\text{Nb}(d,x)^{89}\text{Zr}$ reaction.

3.4 Integral yield of ^{93m}Mo

The integral yield of ^{93m}Mo from the $^{93}\text{Nb}(d,2n)^{93m}\text{Mo}$ reaction was deduced from the excitation function using the spline fitting and the stopping power $S(E)$ calculated on the SRIM code [14]. Physical yield [22] was derived in the unit of MBq/ μAh up to 22.7 MeV. The result together with the other experimental data [8][23] is shown in Fig. 5. The data of Ref. [8] were deduced by the excitation function measured in Ref. [6]. Our result is almost consistent with the data in [8][23]. The deviation from the data in [8] probably comes from slight difference of cross sections and stopping power calculation.

Fig. 5. The physical yield of ^{93m}Mo .

4. Summary

The cross sections of the deuteron-induced reactions on ^{93}Nb were measured up to 24 MeV. The cross sections to produce $^{93\text{m}}\text{Mo}$, $^{92\text{m}}\text{Nb}$ and ^{89}Zr were derived using the stacked-foil activation method and off-line γ -ray spectrometry. The obtained excitation functions were very smooth in the whole energy region. Our results were compared with the experimental data studied earlier and show good agreement with them. The integral yield of $^{93\text{m}}\text{Mo}$, deduced from the obtained excitation function, is also in good agreement with the previous data.

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