Annals of Nuclear Energy 63 (2014) 653–657

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Measurement of effective neutron cross section of the reaction ^{98}Mo (n, γ) ^{99}Mo in TRR nuclear reactor by using thermal neutrons

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ARTICLE INFO

Article history:
Received 17 April 2013
Received in revised form 21 August 2013
Accepted 22 August 2013

Keywords:
Activation method
Westcott's convention
Effective cross section
Molybdenum-98
Thermal neutron

ABSTRACT

The effective cross section of ^{98}Mo was measured using Westcott's convention. Diluted samples of natural MoO_3 were irradiated for 15 min in the channel D_6 of the core of the Tehran research reactor (TRR). The neutron flux at irradiation position was measured using diluted Au_2O_3 and Co_3O_4. The effective cross section was deduced as 0.526 ± 0.02b obtained from thermal flux. Index in Westcott's convention, $r\sqrt{T/T_0}$ at the irradiation position was obtained as 0.0545 ± 0.001. The contribution of the epithermal neutrons in total neutron activity of ^{99}Mo was 69%. The necessary correction factors such as thermal neutron self-shielding (G_{th}) and epithermal neutron self-shielding (G_{epi}), and gamma-ray attenuation (F_g) were taken into account. The results were presented and compared with effective cross section in evaluated nuclear data laboratory such as JENDL-4, KAERI and evaluated data by Mughabghab.

1. Introduction

^{98}Mo is used as a parent isotope for the production of ^{99m}Tc generators. The ^{99m}Tc with half life of 6.01 h is the most widely used radiopharmaceutical in nuclear medicine which is produced from disintegration of ^{99}Mo with half-life of 66 h. More than 90% of nuclear medicine diagnoses are established by using this radionuclide (Froment et al., 2002). In Iran ^{99m}Tc is used for 10,000–14,000 patients weekly in 120 nuclear medicine centers and hospitals. Usually, ^{99}Mo is produced around the world using two major pathways: separation from fission products of ^{235}U and neutron activation of ^{98}Mo. In Iran due to limitation in the use of ^{235}U, the second method was selected. In this method neutron capture on ^{98}Mo in natural molybdenum trioxide was used as a target material. The main disadvantage of the neutron capture on ^{98}Mo is the low specific activity since thermal neutron cross section for ^{98}Mo is small, 0.137 barn (Mughabghab, 2003). It is known that ^{98}Mo has neutron cross section with resonance in the epithermal energies higher than 0.4 eV (Ryabchikov et al., 2004; Shikata and Iguchi, 1986). Therefore epithermal neutrons have a great role in producing ^{99}Mo of high activities. We selected central channel D_6 at Tehran research reactor for irradiation of the samples, because of maximum thermal and epithermal neutron flux in it.

The aim of this work was experimental measurement of effective cross section of ^{98}Mo (n, γ) ^{99}Mo, which depends on the fraction of epithermal neutrons in the neutrons spectrum at the irradiation position and study contribution of the epithermal neutrons in total neutron activity of ^{99}Mo.

2. Theory

According to Westcott's convention (Westcott et al., 1958) the reaction rate per target nuclei is given by:

$$R = \varphi_0 \sigma(T) \tag{1}$$

where $\varphi_0 = n v_0$ is the neutron flux defined as the neutron density times the 2200 m/s velocity, σ (T) is the effective cross section and can be defined as

$$\sigma(T) = \sigma_0 \left[G_{th} g(T) + r\sqrt{T/T_0} S_0 G_{epi} \right] \tag{2}$$

where σ_0 is the thermal cross section for 2200 m/s neutron, g (T) is the parameter which represents the departure of the cross section from the $1/v$ law in the thermal region (g $(T) = 1$ if nuclide obeys the $1/v$ law in this energy region), G_{th} and G_{epi} are the self-shielding coefficients for the thermal and epithermal neutron, $r\sqrt{T/T_0}$ is the epithermal index which denotes the fraction of the epithermal neutron in the neutron spectrum at irradiation position (it is considered zero for a pure thermal flux) and S_0 is the parameter which represents the ratio of the resonance integral and thermal cross-section, such as

$$S_0 = \frac{2}{\sqrt{\pi}} \frac{I_1}{\sigma_0} \tag{3}$$

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http://dx.doi.org/10.1016/j.anucene.2013.08.035

where I_1 is the reduced resonance integral (Gryntakis and Kim, 1983; Katoh et al., 1999). I_1 means the quantity after subtracting the $1/v$ component above the Cd cut-off energy, E_{cd}, from the resonance integral I. A cylindrical Cd-filter of 1 mm thickness has E_{cd} = 0.55e V. The $1/v$ contribution to the resonance integral from the E_{cd} is given by

$$\Delta I(1/v) = I - I_1 = \int_{E_{cd}}^{\infty} g(T)\sigma_0\sqrt{\frac{E_0}{E}}\cdot\frac{dE}{E} = 2g(T)\sigma_0\sqrt{\frac{E_0}{E_{cd}}} \tag{4}$$

For E_0 taken as 0.0253 eV, therefore, contribution of $1/v$ component to the resonance integral is estimated as

$$\Delta I(1/v) = 0.428\ g\sigma_0 \tag{5}$$

The reduced resonance integral can be defined as

$$I_1 = I - 0.428\ g\sigma_0 \tag{6}$$

Eq. (1) with Eq. (2) is rewritten in a simplified form (Nakamura et al. 2003):

$$R = \varphi_0\sigma(T) = \sigma_0[\phi_1 G_{th} g(T) + \phi_2 S_0 G_{epi}] \tag{7}$$

For the irradiation without a Cd cover sheet,

$$R' = \varphi'_0\sigma(T) = \sigma_0[\phi'_1 G_{th} g(T) + \phi'_2 S_0 G_{epi}] \tag{8}$$

For the irradiation within Cd cover sheets. R and R' represent the reaction rates for irradiation without a Cd covert and within Cd cover, respectively. φ_1 and φ'_1 are the neutron flux components in the thermal energy region and φ_2 and φ'_2 are in the epithermal energy region. The values of $\varphi_{1,2}$ and $\varphi'_{1,2}$ at the irradiation of each sample are determined by solving the simultaneous equations of Eqs. (7) and (8) for monitors Au and Co. The nuclear data of flux monitors (diluted Au_2O_3 and Co_3O_4) are given in Table 1 (Mughabghab, 2003). The reaction rate is measured by an HPGe detector and can be evaluated as

$$R = \frac{N_p M e^{\lambda t_d}}{N_a t_m W \varepsilon\theta\gamma(1 - e^{-\lambda t_{irr}})} \tag{9}$$

where t_d is the time interval between the termination of irradiation and the start of counting; t_m is the counting interval time; t_{irr} is the irradiation time; λ is the decay constant; N_p is the net number of counts in the full-energy peak; W is the weight of the sample; M is the atomic weight; N_a is Avogadro's number; ε is the photo peak counting efficiency; θ is the isotopic abundance of the target nuclide; γ is the gamma emission probability. The correction factor for gamma-ray attenuation, F_g, for cylindrical samples at a given γ-ray energy was approximated as follows (Karadag et al., 2003):

$$F_g = \frac{\mu t}{1 - e^{-\mu\cdot t}} \tag{10}$$

where μ is the linear attenuation coefficient (cm^{-1}) and t is the sample thickness in cm, it is possible to obtain μ/ρ which is the total mass attenuation coefficient (cm^2/g) for the compound materials, which are taken from the XCOM database (Berger et al., 1999).

3. Experimental procedure

The samples were irradiated in the channel D_6 of the core of the Tehran research reactor. The reactor was of the swimming pool type and the reactor power was 5 MW. Fig. 1 shows the arrangement of the reactor core, which has 6 vertical irradiation channels. Thermal and epithermal neutron fluxes were calculated by using the MCNP Monte Carlo transport code. As Fig. 2 illustrates the maximum thermal neutron flux is 4.74×10^{13} n/cm^2 s and the maximum epithermal neutron flux is 2.72×10^{12} n/cm^2 s in energy range 4–1000 eV.

The samples are in oxide form and all diluted to Al_2O_3 powder up 95.93% in order to minimize errors due to the self-shielding effects, because Al has a low neutron absorption cross-section. The samples were filled in the quartz ampoules with a wall thickness of 1 mm, external diameter of 6 mm and height of 20 mm. For determination values R', neutron flux and Cd-ratio (R_{cd}) of a monitor nuclide, Cd capsules, 1 mm in thickness, 20 mm in diameter and 30 mm in height, were used. The Cd-ratio is the ratio of the activity of the bare sample to the activity of cadmium covered sample. The neutron flux exposed to each sample was determined by measurement activities of diluted Au_2O_3 and Co_3O_4 samples irradiated simultaneously with the natural MoO_3. The samples were irradiated for 15 min. For each run, three samples were put into Cd capsules and other three samples were irradiated without Cd capsule. To avoid the effect of Cd cover to neutron flux exposed to the bare samples, the assigned distance between the samples was 50 mm. The correction factor for thermal and epithermal neutron fluxes due to the distance between the two sample series were estimated to be 0.9799 and 0.9874, respectively, which were obtained from MCNP Monte Carlo transport code.

The gamma ray measurements were performed after proper cooling time. The detector used for measurement of the gamma ray from the irradiated samples was high purity germanium with a diameter of 59.2 mm and a thickness of 30 mm. Its efficiency was 80% at 1.33 MeV relative to a 76.2 mm diameter × 76.2 mm length NaI detector. The energy resolution of the detector was 1.8 keV full width half maximum (FWHM) at the 1.33 MeV gamma peak of ^{60}Co. The full peak efficiency calibration of high purity germanium detector was performed using a mixed standard radionuclides solution containing ^{152}Eu.

4. Data evaluation

4.1. Neutron self-shielding correction factors

Neutron self-shielding correction factors must be introduced to be taken into account. These factors depend on the geometry and dimension of the sample. The self-shielding factor is defined as the ratio of specific activity of the irradiated target A to specific activity of an irradiated infinite diluted target A_0. Hence, neutron self-shielding factor, G, for a sample is defined as follows:

$$G = \frac{A}{A_0} \tag{11}$$

Table 1
Nuclear data used for of flux monitors and ^{98}Mo.

Sample	Nuclear reaction	Half-life	g (300 K)	σ_0 (b)	I_0 (b)	S_0[a]	Energy (MeV)	I_γ (%)
Al_2O_3–2.854%Co_3O_4	^{59}Co(n, γ)^{60}Co	5.271 years	1.0004	37.18 ± 0.06	75.9 ± 2.0	1.82 ± 0.06	1.173	99.90
							1.333	99.98
Al_2O_3–2.123%Au_2O_3	^{197}Au (n, γ)$_{198}$Au	64.68 h	1.0054	98.65 ± 0.09	1550 ± 28	17.25 ± 0.32	0.4118	95.5
Al_2O_3–4.068%MoO_3	^{98}Mo (n, γ)^{99}Mo	65.94 h	1.006	0.137 ± 0.005	6.9 ± 0.3	56.36 ± 3.0	0.739.5	12.13

[a] Evaluated by using Eq. (3).