Annals of Nuclear Energy 38 (2011) 1693–1697

Contents lists available at ScienceDirect

Annals of Nuclear Energy

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

Measurements of activation cross sections for ^{175}Lu(n, α)^{172}Tm, ^{176}Lu(n, α)^{173}Tm and ^{175}Lu(n, p)$^{175m+g}$Yb reactions induced by neutrons around 14 MeV

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ARTICLE INFO

Article history:
Received 1 April 2010
Received in revised form 2 April 2011
Accepted 11 April 2011
Available online 2 May 2011

Keywords:
Neutrons
Nuclear reaction
Cross section
Activation technique
Lutetium

ABSTRACT

The cross sections for the ^{175}Lu(n, α)^{172}Tm, ^{176}Lu(n, α)^{173}Tm and ^{175}Lu(n, p)$^{175m+g}$Yb reactions have been measured in the neutron energy range of 13.5–14.8 MeV using the activation technique. The first data for ^{175}Lu(n, α)^{172}Tm reaction cross sections are presented. In our experiment, the fast neutrons were produced via the ^{3}H(d, n)^{4}He reaction on K-400 Neutron Generator at Chinese Academy of Engineering Physics (CAEP). Induced gamma activities were measured by a high-resolution (1.69 keV at 1332 keV for ^{60}Co) gamma-ray spectrometer with high-purity germanium (HPGe) detector. Measurements were corrected for gamma-ray attenuations, random coincidence (pile-up), dead time and fluctuation of neutron flux. The neutron fluences were determined by the cross section of ^{93}Nb(n, 2n)^{92m}Nb or ^{27}Al(n, α)^{24}Na reactions. The neutron energy in the measurement was by the cross section ratios of ^{90}Zr(n, 2n)$^{89m+g}$Zr and ^{93}Nb(n, 2n)^{92m}Nb reactions. The results were discussed and compared with experimental data found in the literature and with results of published empirical formulae.

1. Introduction

Experimental data of neutron-induced reactions in the energy range around 13–15 MeV are needed to verify the accuracy of nuclear models used in the calculation of cross sections. Furthermore, the data are of considerable importance for practical applications, such as for integral calculations on the first wall, blanket and shield of a conceptual fusion power reactor. The data for gas production via neutron induced reactions are of great importance in the domain of fusion reactor technology, particularly of nuclear transmutation rates, nuclear heating and radiation damage due to gas formation. A lot of experimental data on neutron induced cross sections for fusion reactor technology applications have been reported and great efforts have been devoted to compilations and evaluations (CINDA-A, 2000; Mclane et al., 1988). The variations in the cross sections with the neutron energy are also of great interest for studying the excitation of nuclei to different energy levels and subsequent decay to ground state, either directly or through different energy levels including metastable state. We chose to study the neutron-induced reaction cross sections of the lutetium (Lu) isotopes mainly for three reasons. First, lutetium has the highest atomic number of all the rare earth elements (REE). It is comprised of two isotopes ^{175}Lu and ^{176}Lu and is followed by the element hafnium. Due to its importance to *s*-process studies, lutetium has attracted considerable scientific interest. Second, The reaction cross sections of ^{176}Lu(n, α)^{173}Tm and ^{175}Lu(n, p)$^{175m+g}$Yb around the neutron energies 14 MeV were only obtained by three laboratories (Sato et al., 1975; Qaim, 1976; Coleman et al., 1959), but all measurements were obtained before 1980. Furthermore, there were large discrepancies in those data. The large discrepancies are probably due to use of different methods, energy resolution of detectors, target materials and adopting different nuclear parameters. Third, for ^{175}Lu(n, α)^{172}Tm reaction of lutetium isotopes, the cross sections have been not reported. Thus it is necessary to measure then again and obtain excitation functions around the neutron energies of 14 MeV. About the cross sections and isomeric cross section ratios of ^{175}Lu(n, 2n)^{174m,g}Lu in the neutron energies of 13.5–14.8 MeV for lutetium have been reported by authors (Vallis, 1966; Qaim, 1974; Frehaut and Mosinski, 1974; Dilg et al., 1968; Laurec et al., 1981; Wille and Fink, 1960; Nethaway, 1972; Veeser et al., 1977; Bayhurst et al., 1975; Luo et al., 2007b).

In the present work, the cross sections of the ^{175}Lu(n, α)^{172}Tm, ^{176}Lu(n, α)^{173}Tm and ^{175}Lu(n, p)$^{175m+g}$Yb were measured in at neutron energy 13.5–14.8 MeV and a gamma-ray counting technique was applied using high-resolution gamma-ray spectrometer and data acquisition system. Pure Lu_2O_3 was used as the target material. The reaction yields were obtained by absolute measurement of the gamma activities of the product nuclei using a coaxial high-purity germanium detector. The neutron energies in this

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doi:10.1016/j.anucene.2011.04.014

measurement was determined by cross section ratios for the ^{90}Zr(n, 2n)$^{89m+g}$Zr and ^{93}Nb(n, 2n)^{92m}Nb reactions (Lewis and Zieba, 1980).

2. Experimental

Cross sections were measured by activation and identification of the radioactive products. This technique is very suitable for investigating low-yield reaction products and closely space low-lying isomeric states, provided their lifetimes are not too short. The details have been described over the years in many publications (Bostan and Qaim, 1994; Cserpák et al., 1994; Luo Junhua et al., 2005; Nesaraja et al., 2003; Rahman and Qaim, 1985). Here we give some salient features relevant to the present measurements.

2.1. Samples and irradiations

About 3 g of Lu_2O_3 powder of natural isotopic composition (>99.99% pure) was pressed at 10 ton/cm^2, and a pellet, 0.2 cm thick and 2.0 cm in diameter was obtained. Three such pellets were prepared. Monitor foils of Nb (99.99% pure, 0.2 mm thick) and Al (99.999% pure, 0.04 mm thick) of the same diameter as the pellets were then attached in front and at the back of each sample.

Irradiation of the samples was carried out at the K-400 Neutron Generator at Chinese Academy of Engineering Physics (CAEP) and lasted 128 min with a yield ~4–5 × 10^{10} n/s. Neutrons were produced by the T(d, n)^{4}He reaction with an effective deuteron beam energy of 134 keV and beam current of 230 μA. The tritium–titanium (T–Ti) target used in the generator was 2.18 mg/cm^2 thick. The neutron flux was monitored by a uranium fission chamber so that corrections could be made for small variations in the yield. The groups of samples were placed at 0°, 90° or 135° angles relative to the beam direction and centered about the T–Ti target at distances of ~3–5 cm. Cross sections for ^{93}Nb(n, 2n)^{92m}Nb or ^{27}Al (n, α)^{24}Na reactions (Wagner et al., 1990) were selected as monitors to measure reaction cross section of the ^{175}Lu(n, α)^{172}Tm, ^{176}Lu(n, α)^{173}Tm and ^{175}Lu(n, p)$^{175m+g}$Yb.

2.2. Determination of the incident neutron energy

In the D–T reaction (Q value of 17.6 MeV), induced by deuterons of energy E_d, the kinetic energy E_n of the neutrons emitted at angle θ can be estimated (Curtis, 1969) from the following expression:

$$(E_n)^{\frac{1}{2}} = \frac{(M_d M_n E_d)^{\frac{1}{2}} \cos\theta + \left(M_d M_n E_d \cos^2\theta + \{M_\alpha + M_n\}[M_\alpha Q + E_d(M_\alpha - M_n)]\right)^{\frac{1}{2}}}{M_\alpha + M_n} \quad (1)$$

where M_d, M_n and M_α are the masses of deuteron, neutron and alpha particle, respectively. The effective D–T neutron energy at irradiation position was determined by the Nb/Zr method (Curtis, 1969; Lewis and Zieba, 1980; Nethaway, 1978; Pavlik et al., 1982). The measured neutron energy was shown in Fig. 1 together with the calculation using Eq. (1). The uncertainty in the neutron energy at ~3 to 5 cm was estimated to be 200 keV from a consideration of the sample sizes, d$^+$ beam diameter of about 3–4 mm, and the uncertainty in the Nb/Zr method (Lewis and Zieba, 1980).

2.3. Measurement of radioactivity

The gamma-ray activity of ^{172}Tm, ^{173}Tm, ^{175g}Yb, ^{92m}Nb and ^{24}Na was determined by a high-purity germanium (HPGe) detector (ORTEC, model GEM 60P, Crystal diameter: 70.1 mm, Crystal length: 72.3 mm, made in USA) with a relative efficiency of ~68% and an energy resolution of 1.69 keV at 1332 keV for ^{60}Co. The efficiency of the detector was pre-calibrated using various standard gamma sources. One of the gamma-ray spectra is shown in Fig. 2. The decay characteristics of the product radioisotopes and the natural abundances of the target isotopes under investigation are summarized in Table 1 (Browne and Firestone, 1996).

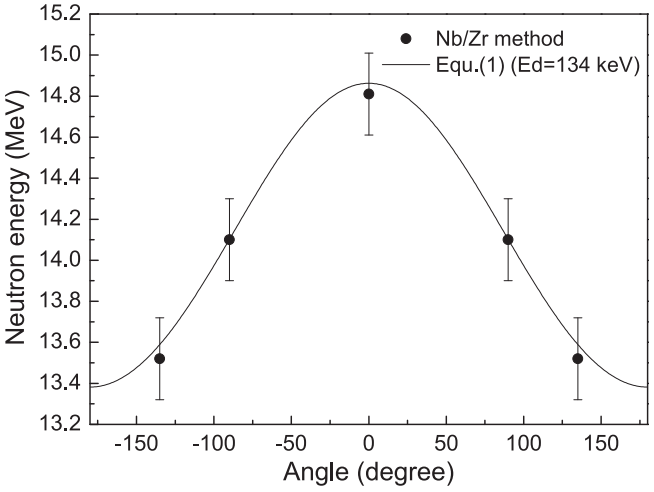

Fig. 1. Angular dependence of D–T neutron energy. The neutron energies were calculated by choosing the incident deuteron energy E_d = 134 keV. The open circles show experimental data determined by the Nb/Zr method (Lewis and Zieba, 1980).

2.4. Calculation of cross sections and their uncertainties

The measured cross sections can be calculated by the following formula (cf. Luo et al., 2007a):

$$\sigma_x = \frac{[S\varepsilon I_\gamma \eta KMD]_0}{[S\varepsilon I_\gamma \eta KMD]_x} \frac{[\lambda AFC]_x}{[\lambda AFC]_0} \sigma_0 \quad (2)$$

where the subscript m represents the term corresponding to the monitor reaction and subscript x corresponds to the measured reaction. ε is the full-energy peak efficiency of the measured characteristic gamma-ray, I_γ the gamma-ray intensity, η the abundance of the target nuclide, M the mass of sample, $D = e^{-\lambda t_1} - e^{-\lambda t_2}$ the counting collection factor, t_1, t_2 the time intervals from the end of the irradiation to the start and end of counting, respectively, A the atomic weight, C the measured full-energy peak area, λ the decay constant and F is the total correction factor of the activity:

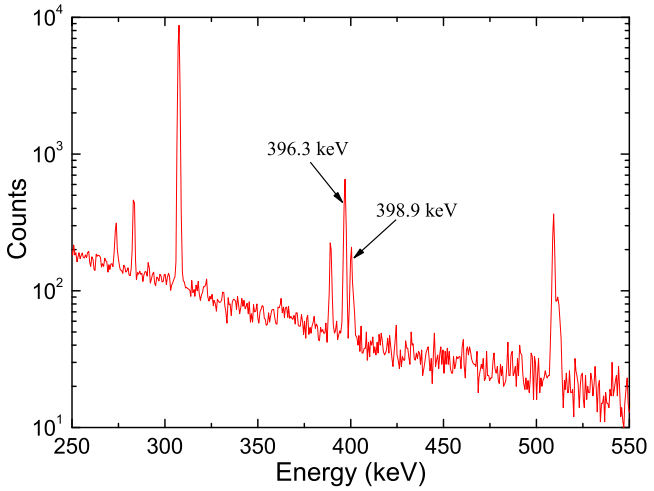

Fig. 2. The γ-ray spectra of about 3 h after the end of the irradiation.