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Measurement of Activation Cross Sections of Erbium Irradiated by Proton Beam

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Erbium is one of the rare earth elements which are used to produce therapeutic radioisotopes by charged particle induced reactions, such as proton, deuteron and alpha particle-induced reactions. In this study, production cross sections of ^{165}Tm and ^{166}Tm were measured for erbium samples irradiated by a proton beam with $E = 45$ MeV provided by the MC-50 cyclotron of KIRAMS (Korea Institute of Radiological and Medical Sciences). A HPGe detector was used to measure γ-rays for an activation cross section measurement. A stacked foil technique was adopted to measure the activation cross sections at different proton energies using copper foils as degraders and monitor foils to measure the proton beam current. The SRIM code was used to calculate the energy of the proton beam penetrating each foil. The measured data are compared with other available experimental data and theoretical cross sections calculated by TALYS code.

I. INTRODUCTION

Erbium is one of the rare earth elements which are used to produce therapeutic radioisotopes by charged particle induced reactions using proton, deuteron and alpha particle beams. Charged particle-induced reactions have advantages in specific activity and carrier level of end-products compared to the (n,γ) reaction. The radionuclide ^{165}Tm is one of the important isotopes produced through the ^{nat}Er(p,x) reaction since it decays to ^{165}Er with a half-life of 30.06 h. The radionuclide ^{165}Er can be used for Auger-electron therapy since it decays by electron capture without gamma radiation with a decay time of 10.36 h. The radionuclide ^{165}Er can be produced directly by ^{165}Ho(p,n)^{165}Er reaction [1].

In this study, activation cross sections were measured for erbium samples irradiated by a proton beam, specifically the production cross section of ^{165}Tm as well as ^{166}Tm. Similar work has been performed by Tarkanyi *et al.* [2–5] who measured the activation cross sections of proton and deuteron induced reactions on erbium and Kiraly *et al.* [6] who measured the activation cross sections of alpha particle induced reactions on erbium. Tarkanyi *et al.* [2, 3] measured the cross sections of ^{nat}Er(p,x)^{165}Tm and ^{nat}Er(p,x)^{166}Tm for the proton energy range 0 to 35 MeV and 60 to 70 MeV. The current work reports on cross sections measurements for proton energies from 19 to 45 MeV.

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II. EXPERIMENTAL METHOD

The MC-50 cyclotron of KIRAMS (Korea Institute of Radiological and Medical Sciences) provided a 45 MeV proton beam which was incident on a natural Er sample. A conventional stacked foil activation technique was used to measure the production cross sections of Tm radioisotopes as a function of proton energy.

Decay γ-ray energies have to be selected for cross section measurements. The selected gamma energies are between 200 and 1400 keV considering the availability of measured efficiencies. We also selected gamma energies having intensities greater than 2% and 5% for ^{165}Tm and ^{166}Tm respectively. The selected gamma energies were checked for doublet with gamma energies of other radioisotopes produced by irradiation. TALYS code [7] calculation results and decay data were used for checking the doublet. Among the selected gammas, the 242.917 keV and 778.814 keV gammas were used for ^{165}Tm and ^{166}Tm, respectively.

Erbium foils were purchased from Goodfellow. We measured the thickness of Er foils by measuring the mass and dimensions of Er foils. The measured thickness was 22.44 ± 0.02 μm. A total of 16 erbium foils having dimension of 1.1 cm x 1.1 cm were prepared for the irradiation. The production cross section, σ of the Tm radioisotopes is determined from

$$\sigma = \frac{\lambda C}{I_p t \rho \{1 - e^{-\lambda t_i}\} e^{-\lambda t_c} \{1 - e^{-\lambda t_m}\} I_\gamma \epsilon}, \qquad (1)$$

where λ is the decay constant (s^{-1}), C is the net counts in the photo-peak, I_p is the proton beam intensity (protons/s), t is the thickness of foil (cm), ρ is the atomic density (atoms/cm^{-3}), t_i is the irradiation time (s), t_c is the cooling time (s), t_i is the measurement time (s), I_γ is the gamma-ray intensity, and ϵ is the detection efficiency. The erbium foils were irradiated by a proton beam for one hour through a collimator having a diameter of 1 cm. Since the half-lives of ^{165}Tm and ^{166}Tm are 30.06 h and 7.70 h respectively, a cooling time of about 1 day was adopted.

Gamma-rays were measured with a p-type coaxial ORTEC HPGe detector with an efficiency of 10%. The Gamma Vision 5.1 program was used for the analysis of the spectra. Decay data was taken from the NNDC nuclear data [8]. The detection efficiency was measured by using 8 different γ-rays from a standard ^{152}Eu source.

The reactions contributing to the production of Tm radioisotopes are listed in Table I. Natural erbium consists of 6 isotopes. The abundances are 0.14%, 1.60%, 33.50%, 22.87%, 26.98% and 14.91% for ^{162}Er, ^{164}Er, ^{166}Er, ^{167}Er, ^{168}Er and ^{170}Er, respectively. The Q-values and threshold energies were calculated by Q-value Calculator program at the NNDC in BNL [9].

TABLE I. Contributing reactions of ^{165}Tm and ^{166}Tm.

Nuclide	Contributing reactions	Q-value (MeV)
^{165}Tm	^{164}Er(p,γ)	4.3
	^{166}Er(p,2n)	-10.8
	^{167}Er(p,3n)	-17.3
	^{168}Er(p,4n)	-25.1
	^{170}Er(p,6n)	-38.3
^{166}Tm	^{166}Er(p,n)	-3.8
	^{167}Er(p,2n)	-10.3
	^{168}Er(p,3n)	-18.0
	^{170}Er(p,5n)	-31.3

The proton beam intensity was determined using the monitor reaction ^{nat}Cu(p,x)^{62}Zn. A 50 μm thick natural Cu foil was placed after each Er foil to monitor proton beam intensities throughout the entire energy range. The production cross section of ^{62}Zn was taken from Ref. [10]. The 596.56 keV γ-ray from the decay of ^{62}Zn was used to deduce the yield and derive the proton beam current. The proton beam current was considered as constant throughout all the foils. The average value of all 16 beam intensities was used for the analysis. A 100 μm thick natural Cu foil was also placed after each Er and 50 μm thick Cu foils to degrade the energy of the proton beam. The proton beam energy at each foil was calculated using the SRIM code [11].

III. MODEL CODE CALCULATIONS

The measured cross sections of ^{nat}Er(p,x)^{165}Tm and ^{nat}Er(p,x)^{166}Tm were compared with theoretical calculation predictions from the TALYS code. In this work,

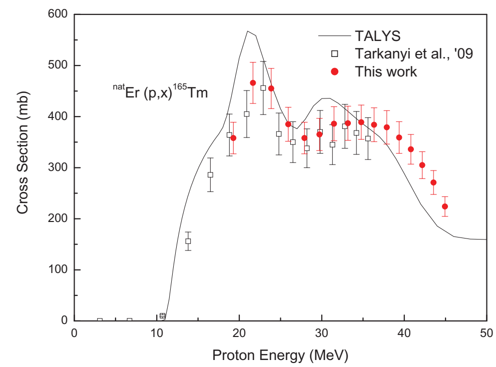

FIG. 1. Measured cross sections of the ^{nat}Er(p,x)^{165}Tm reaction in comparison with theoretical calculations.

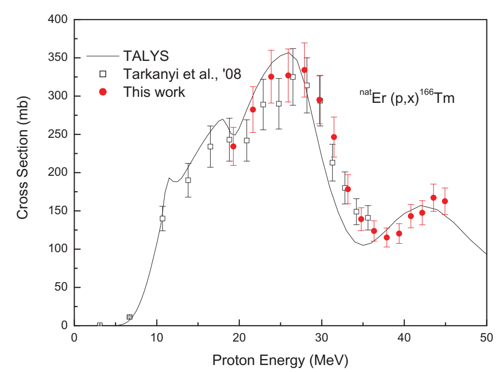

FIG. 2. Measured cross sections of the ^{nat}Er(p,x)^{166}Tm reaction in comparison with theoretical calculations.

TALYS-1.2 was used with the default parameter set. The TALYS calculations provide not only the total cross section but also the cross section of each reaction channel contributing to the total cross section.

IV. RESULTS AND DISCUSSION

Table II shows the proton energy at each erbium foil. The uncertainty of proton beam energy is due to the energy straggling and foil thickness. Eq. (1) was used to estimate the proton beam current from the counts of 596.56 keV gammas which was measured from the irradiated Cu foils. The average current measured from 16 Cu foils is 96.0 nA.

The cross section of ^{nat}Er(p,x)^{165}Tm and ^{nat}Er(p,x)^{166}Tm were estimated by using counts of 242.917 keV (I_γ=35.5%) and 778.814 keV (I_γ=19.1%) gammas respectively. The main source of cross section uncertainty is the uncertainty of proton beam current.

Fig. 1 shows the measured cross section of ^{nat}Er(p,x)^{165}Tm as a function of proton energy. Our results are lower than the theoretical predictions

TABLE II. Measured production cross sections of ^{165}Tm and ^{166}Tm.

Proton energy (MeV)	Cross sections (mb)	
	^{165}Tm	^{166}Tm
44.9 ± 0.1	224 ± 19	163 ± 17
43.6 ± 0.2	271 ± 23	167 ± 18
42.2 ± 0.2	305 ± 26	148 ± 16
40.8 ± 0.2	336 ± 29	143 ± 15
39.4 ± 0.3	359 ± 31	120 ± 13
37.9 ± 0.3	379 ± 33	115 ± 12
36.3 ± 0.3	384 ± 33	124 ± 13
34.8 ± 0.4	389 ± 34	139 ± 15
33.2 ± 0.4	387 ± 33	178 ± 19
31.5 ± 0.4	386 ± 33	246 ± 26
29.7 ± 0.5	365 ± 32	295 ± 31
27.9 ± 0.5	358 ± 31	334 ± 35
25.9 ± 0.6	385 ± 33	327 ± 35
23.9 ± 0.6	455 ± 39	325 ± 34
21.7 ± 0.7	466 ± 40	282 ± 30
19.3 ± 0.7	358 ± 31	234 ± 25

from the TALYS code in the energy range of 20 - 35 MeV, but higher in the range of 35 - 45 MeV. Our measured data was compared with the data reported by Tarkanyi *et al.* [2]. Both data agree well within the uncertainties.

Fig. 2 shows the measured cross section of ^{nat}Er(p,x)^{166}Tm as a function of proton energy. Although the agreement between our results and TALYS calculation results are better than the case of ^{165}Tm, our data is higher than the TALYS results in the energy range of 30 - 35 MeV. Our measured data agree well with the data reported by Tarkanyi *et al.* [3].

V. CONCLUSIONS

Erbium is one of the rare earth elements used to produce therapeutic radioisotopes by charged particle induced reactions. The cross sections of ^{nat}Er(p,x)^{165}Tm and ^{nat}Er(p,x)^{166}Tm were measured for proton energies from 19 to 45 MeV using a conventional stacked foil activation technique.

The measured cross sections of ^{nat}Er(p,x)^{165}Tm and ^{nat}Er(p,x)^{166}Tm were compared with theoretical calculations values from the TALYS code. The measured data and TALYS calculation results show similar tendencies in excitation functions. A good agreement is shown between our data and other experimental data.

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