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Measurement of excitation functions in alpha-induced reactions on yttrium

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ABSTRACT

The excitation functions of ^{89g,m,90,91m,92m}Nb,^{88,89}Zr, and ^{87g,m,88,90m,91m}Y from alpha-induced reactions on ^{89}Y were measured from their respective threshold to 45 MeV by using a stacked-foil activation technique at the MC-50 cyclotron of the Korean Institute of Radiological and Medical Sciences. The results were compared with the earlier reported data as well as with the theoretical values obtained from the TENDL-2013 library based on the TALYS1.6 code. Our measurements in the energy region from the threshold energy to 45 MeV are in general good agreement with the other experimental data and calculated results. The integral yields for thick target of the produced radionuclides were also deduced from their measured cross sections and the stopping power of ^{89}Y. The measured excitation functions find importance in various practical applications including nuclear medicine and improvement of nuclear model calculations.

1. Introduction

The role of the cyclotron in the production of medically promising radioisotopes and fulfilling the particular needs of users is increasing. Cyclotrons are efficiently used to produce the radioisotopes due to compact size and to easy operation. Other than that, cyclotrons produce less harmful waste, minimum nuclear accidents, no nuclear proliferation risk, and are easy to handle as compared with nuclear reactors. From radioisotopes production point of view, medical cyclotrons are the best route to produce short lived and β^+ emitter radionuclides. Recent advancements in the medical sciences and multiple applications of radioisotopes in research and development have widened the interdisciplinary fields. Physicists, chemists, physicians, industrialists, and agriculturalists are getting benefits of radioisotopes by applying them in the respective fields of research [1–3].

The ^{89}Y nuclide as a target material for alpha activation with suitable energy can be used for the production of medically important radioisotopes (e.g. ^{90}Nb, ^{88,89}Zr, ^{87,88,90}Y) [4]. The radionuclide ^{90g}Y ($T_{1/2}$ = 2.67 d; E_{β^-} = 2.28 MeV; I_{β^-} = 100%) is one of the most widely used therapeutic radio-nuclides for targeting cancer cells or as the active ingredient used in treating liver tumors [5,6]. Since ^{90g}Y decays entirely by β^- emission, an accurate calculation of dosimetry is difficult. Therefore, a corresponding γ- or β^+-emitter (^{87}Y or ^{88}Y versus ^{90}Y) is needed for estimating optimal therapeutic doses for human. The radionuclide ^{87g}Y ($T_{1/2}$ = 79.8 h; E_γ = 388.5 keV (82.2%) and E_γ = 484.8 keV (89.8%)) used as a γ-ray emitter can be used in combination with the single photon emission computed tomography [7]. The long-lived ^{88g}Y ($T_{1/2}$ = 106.63 d; E_γ = 898.04 keV (93.7%) and E_γ = 1836.06 keV (99.2%)) radionuclide is used to investigate slow metabolic processes in animals [8] and also frequently used as a calibration source of γ-ray detectors. The ^{88}Zr radionuclide can be used to produce significant amount of ^{88}Y radioisotope through the ^{88}Zr/^{88}Y generator. Due to its reasonable half-life the radionuclide is also used to study animal bio distribution. Besides the conventional β^+-emitters such as ^{11}C, ^{15}O, and ^{18}F, the metallo-radionuclide ^{89}Zr ($T_{1/2}$ = 78.41 h; E^+_β = 0.9 MeV; I^+_β = 22.7%) has been suggested as a suitable positron emitting radionuclide for labeling antibodies [9] and as a potential candidate for the immuno-PET [10]. The use of ^{89}Zr is constantly evolving and it is expected that in the near future this radionuclide will be widely applied in a PET imaging. The emission of β^+ (E_{mean} = 0.66 MeV and E_{max} = 1.5 MeV) from ^{90}Nb makes it especially promising for the immuno-PET and clinical applications [11,12].

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Only limited excitation functions for the alpha-induced reactions on ^{89}Y are available in the various energy ranges [13–19]. In the present work, we measured the excitation functions of ^{89g,m}Nb, ^{90}Nb, ^{91m}Nb, ^{92m}Nb, ^{88}Zr, ^{89}Zr, ^{87g,m}Y, ^{88}Y, ^{90m}Y, and ^{91m}Y in the ^{89}Y(α,x) reactions by the stacked-foil activation and off-line γ-ray spectrometric technique in the energy range between their respective reaction threshold and 45 MeV at the MC-50 cyclotron of the Korean Institute of Radiological and Medical Sciences (KIRAMS), Korea. The experimental results measured in this work were compared with the literature data [13–16] and also theoretical values obtained from TENDL-2013 library [20] based on the computer code TALYS-1.6 [21]. The integral yields for thick target of the investigated radioisotopes were calculated from the excitation function and the electronic stopping power.

2. Experimental details and data analysis

The excitation functions of residual radioisotopes by irradiating yttrium metallic foils with alpha particles were measured as a function of alpha energy from their respective thresholds up to 45 MeV by using a well-established stacked-foil activation technique combined with the off-line gamma spectrometry. The stacked-foil activation technique, activity determination, and data evaluation procedures are described elsewhere [22–29]. Some important features relevant to the present work are described below.

A high-purity yttrium (99.9% purity, 25-μm thickness) and natural copper (>99% purity, 10-μm thickness) metal foils with a size of 1.0 cm × 1.0 cm were stacked alternatively. Natural copper foils were used as a flux monitor and an energy degrader. The sequence of the yttrium and copper foils in a stack was designed on the basis of threshold values for the products from ^{89}Y(α,x) and ^{nat}Cu(α,x) reactions. We made two stacked-foils in order to measure cross-sections for different input alpha energies. The first stack consists of eleven sets of Y-Cu foils and five Y foils are attached in the stack. The second stack was made by adding a 25-μm thick Al foil in front of the first stack. This additional Al foil was used as an energy degrader to reduce the input alpha energy in each foil.

The stacked-foil was irradiated by a 45 MeV collimated alpha beam with a 10 mm diameter and about 200 nA beam for 30 min at the external beam line of the MC-50 cyclotron at KIRAMS [30]. The beam energy and current were kept constant during the irradiation. The irradiation geometry was designed so that the foils in the sample get the maximum beam. After irradiation and an appropriate cooling time, the induced activities of the irradiated foils were measured by using a γ-ray spectrometry. The γ-ray spectrometer was an energy- and efficiency-calibrated n-type coaxial HPGe detector couple to a PC-based multi-channel analyzer with the associated electronics to determine the photo-peak area of the γ-ray spectrum by using the Gamma Vision 5.0 (EG&G Ortec) program.

The energy resolution of the detector was 1.9 keV full width at half maximum (FWHM) at the 1332.5 keV γ-ray photo-peak of ^{60}Co. The energy and the efficiency calibration of the γ-ray spectrometer was determined at the different distances using the standard gamma source ^{152}Eu having γ-rays in the energies range of 121.8–1408.0 keV. The relative detection efficiency was 20% at 1332.5 keV relative to that of a standard 7.6 cm diameter, 7.6 cm-long NaI(Tl) detector. The detection efficiency curves as a function of the photon energy were measured at several distances from the end-cap of the detector to keep the dead time below 10% and to avoid pileup effects.

The activity measurements of the irradiated samples were started at 3 h after the end of the bombardment (EOB). The measurement of the activated sample and monitor foils were repeated several times to follow the decay of the radionuclides and thereby to identify the possible interfering nuclides. The intensity of alpha beam was determined by using the IAEA recommended monitor reactions, ^{nat}Cu(α,x)^{65}Zn (σ_{max} = 220.9 mb at E_α = 10.5 MeV), ^{nat}Cu(α,x)^{66}Ga (σ_{max} = 133.84 mb at E_α = 42.1 MeV), and ^{nat}Cu(α,x)^{67}Ga (σ_{max} = 52.94 mb at E_α = 42.1 MeV) [31] from the measured activities induced in the monitor foil. The use of multiple monitor foils decreases the probability of introducing unknown systematic errors during an activity measurement. It was also considered that the loss of beam flux was very small and very hard to deduce practically. Therefore we considered the measured beam intensities for each foil were almost constant. The alpha energy degradation along the stacked foils was estimated by using the computer program SRIM-2013 [32]. The estimated average energy loss in each foil was 2–4 MeV.

The production cross-sections for the ^{89}Y(α,x) reactions at the i-th foil, $\sigma(E_i)$ were determined in the alpha energy ranges of by using the well-known activation formula [25–26].

$$\sigma(E_i) = \frac{\lambda \cdot C(E_i)}{\varepsilon(E_\gamma) \cdot I_\gamma \cdot \rho \cdot t \cdot \phi \cdot (1 - e^{-\lambda t_m}) \cdot e^{-\lambda t_c} \cdot (1 - e^{-\lambda t_i})} \quad (1)$$

where λ is the decay constant (s^{-1}), $C(E_i)$ is the net counts under the photo-peak area at the i-th sample, $\varepsilon(E_\gamma)$ is the detection efficiency of the HPGe-detector, I_γ is the γ-ray intensity, ρ is the atomic density, t is the foil thickness (cm), ϕ is the beam intensity (s^{-1}), t_c is the cooling time (s), t_m is the counting time (s), and t_i is the irradiation time (s). The decay data such as the half-life ($T_{1/2} = \ln 2/\lambda$) and γ-ray emission probability (I_γ in Eq. (1)) as well as the γ-ray energy (E_γ) used in the determination of the γ-ray detection efficiency were taken from the ENSDF evaluation [33–38] obtained from the NuDat 2.6 database [39] and are summarized in Table 1. The Q-values and threshold energies calculated on the basis of the atomic mass evaluation by Wang et al. [40] combined with the Q-tool system [41] are also presented in Table 1. Intense and independent characteristic γ-lines were used to quantify the radionuclides. In some cases, two or more characteristic γ-rays were used to check the obtained results.

The uncertainty of the alpha energy for each representing energy point in the stack depends on the irradiation circumstances and the position of the foil in the stack. These are due to the initial beam energy, the thickness and homogeneity of target foils, and the beam straggling. The estimated uncertainty of a representing point in the alpha energy ranges from ± 0.53 up to ±1.33 MeV, which is shown in tables and figures. Total uncertainties of cross-sections were obtained according to the rules of error propagation assuming that all uncertainties are independent. Moreover, some of the sources of uncertainties are common to all data, while others affect each reaction or energy point individually. However, the combined uncertainty in each cross-section was estimated by considering the following uncertainties; statistical uncertainty of the γ-ray counting (4 ∼ 25%), uncertainties in the alpha beam intensity (5 ∼ 6%), in the efficiency calibration of the detector (3 ∼ 4%), due to the sample thickness (1 ∼ 2%), and in gamma intensity (∼1%). The overall uncertainties of the measured cross-sections were in the range of 8 ∼ 26%.

Comparison of reliable experimental cross-sections with model calculated ones, without any adjustment of model parameters, may reveal information on accuracies or deficiencies of various reaction mechanisms included in the conventional reaction models. Therefore, we compare our experimental production cross-sections with the TENDL-2013 library [20], which compiles reaction cross-sections based on the TALYS 1.6 code [21].