Measurements of production cross-sections of medical radioisotopes via charged-particle-induced reactions

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Production reactions of medical radioisotopes are studied for nuclear medicine. One of such radioisotopes is ^{169}Yb and its production cross-sections of four charged-particle-induced reactions were experimentally determined. Physical yields were derived from the measured cross sections. The comparison of the physical yields indicates that the deuteron-induced reaction on ^{169}Tm is preferable for the ^{169}Yb production.

1. Introduction

Radioisotopes can be used for a variety of applications, e.g., radiotherapy and diagnosis in nuclear medicine. There are basically several reactions on stable nuclei to produce each radioisotope. Investigations of such reactions are necessary to find better reactions with less byproducts and with higher cost effectiveness. Production cross-sections of the radioisotopes are thus important nuclear data. However, there still exist a lack of data and data with large errors [1]. It is indispensable to obtain more accurate and reliable data for the application. Recent technical development of accelerators and detectors enables us to reach such data.

We study charged-particle-induced reactions among the possible reactions for the production. The charged-particle-induced reactions have an advantage to be able to produce radioisotopes with atomic numbers different from those of targets. We expect chemical separation of the products from the targets and obtain the objective radioisotopes with high specific activity.

One of the medical radioisotopes we focused on is ^{169}Yb ($T_{1/2}$ = 32.018 d, EC = 100%). It is an Auger electron and X-ray emitter and suitable for brachytherapy [2]. To produce ^{169}Yb, there are several reactions, such as proton-, deuteron- and alpha-induced reactions on thulium and alpha-induced reaction on erbium (Fig. 1). We performed experiments to determine cross sections of the four reactions [3–5]. The production cross-sections were compared with previous studies and theoretical model calculation in the TENDL library [6]. Physical yields of the products for practical use

^{167}Yb 17.5 min	^{168}Yb Stable	**^{169}Yb 32.0 d**	^{170}Yb Stable
^{166}Tm 7.70 h	^{167}Tm 9.25 d	^{168}Tm 93.1 d	^{169}Tm Stable
^{165}Er 10.4 h	^{166}Er Stable	^{167}Er Stable	^{168}Er Stable

Fig. 1. Nuclear chart around ^{169}Yb.

were also be derived from the measured cross sections. The results are expected to contribute to nuclear medicine.

2. Method

The experiments were performed at RIKEN, Japan and ATOMKI, Hungary. The well-developed methods, stacked-foil activation technique and high-resolution gamma-ray spectrometry, were adopted. The targets consisted of thin metallic foils for objective and monitor reactions. The stacked targets were irradiated with beams of the charged particles. The beam intensities were measured by a Faraday cup. Gamma rays emitted from the irradiated foils without chemical separation were measured by HPGe detectors. Nuclear data required for deduction of cross sections were retrieved from online databases [7,8].

The cross sections of the monitor reactions were compared with the IAEA recommended values [9] to assess the beam parameters and target thicknesses. According to the comparison of the monitor reactions, only the beam intensities were corrected within the uncertainties. The corrected intensities and measured thicknesses were used to deduce the production cross-sections of ^{169}Yb.

The production cross-sections of ^{169}Yb were derived from measured net counts of the 177.21-keV gamma line (I_γ = 22.28%). The more intense gamma lines at 63.12 (I_γ = 43.62%) and 197.96 keV (I_γ = 35.93%) were unselected because of possible interference with X rays and the 198.25-keV gamma line from the ^{168}Tm decay ($T_{1/2}$ = 93.1 d).

3. Result and discussion

We measured cross sections of the four reactions, proton- [3], deuteron- [5] and alpha-induced reactions on ^{169}Tm and alpha-induced reaction on ^{nat}Er [4]. Physical yields were derived from the measured cross sections and compared with each other. The most appropriate reaction for the ^{169}Yb production among them was discussed based on the comparison.

3.1. ^{169}Tm(p,n)^{169}Yb reaction

The cross sections of the ^{169}Tm(p,n)^{169}Yb reaction were determined as shown in Fig. 2 [3]. The result is compared with the previous experimental data [10–12] and the TENDL-2019 values [6]. The peak amplitude and position are consistent with Birattari et al. [10]. The data of Spahn et al. [11] are two times larger than ours. The shape of the TENDL-2019 data is different from the experimental data.

3.2. ^{169}Tm(d,2n)^{169}Yb reaction

The excitation function of the ^{169}Tm(d,2n)^{169}Yb reaction was measured [5]. The result is shown in Fig. 3 together with the earlier measured experimental data [13,14] and the TENDL-2019 values [6]. The peak position is in good agreement with the previous experimental data and theoretical calculation although our result shows slightly higher than the other experimental data.

3.3. ^{169}Tm(α,x)^{169}Yb reaction

We performed two experiments to measure the cross sections of the ^{169}Tm(α,x)^{169}Yb reaction. The cooling times were longer than 86.9 d and 36.5 d for the first and the second experiments. During the cooling times, the co-produced parent nucleus ^{169}Lu ($T_{1/2}$ = 34.06 h) had entirely decayed to ^{169}Yb.

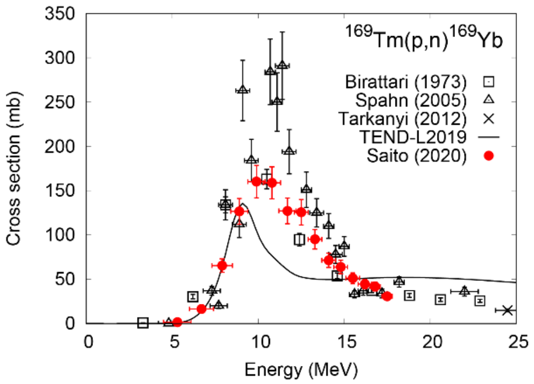

Fig. 2. Excitation function of the ^{169}Tm(p,n)^{169}Yb reaction with the previous data [13,14] and the TENDL-2019 values [6].

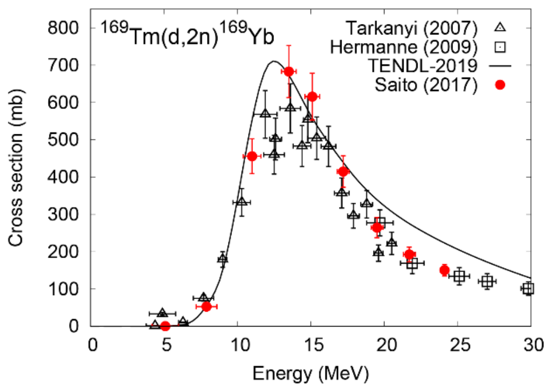

Fig. 3. Excitation function of the ^{169}Tm(d,2n)^{169}Yb reaction with the previous data [13,14] and the TENDL-2019 values [6].

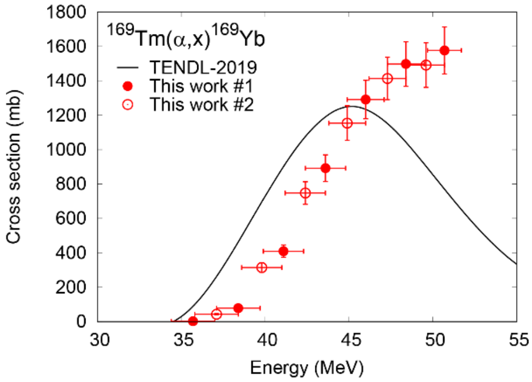

Fig. 4. Excitation function of the ^{169}Tm(α,x)^{169}Yb reaction with the TENDL-2019 values [6].

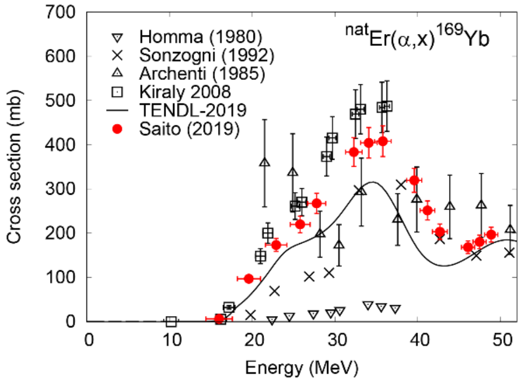

Fig. 5. Excitation function of the ^{nat}Er(α,x)^{169}Yb reaction with the previous data [15–18] and the TENDL-2019 values [6].

Therefore, the cumulative cross sections could be obtained. The results are shown in Fig. 4 with the TENDL-2019 values [6]. The TENDL-2019 values are very different from our result. There is no previous study found in a literature survey.

3.4. ^{nat}Er(α,x)^{169}Yb reaction

The cross sections of the ^{nat}Er(α,x)^{169}Yb reaction were experimentally determined [4]. The result is shown in Fig. 5 in comparison with the previous studies [15–18] and the TENDL-2019 values [6]. Both data of Király et al. [17] and TENDL-2019 have nearly the same peak position as ours at around 35 MeV while the amplitudes are different. The other experimental data differ significantly in both the shape and amplitude from our results.

3.5. Physical yield of ^{169}Yb

Physical yields of ^{169}Yb in the proton-, deuteron-, alpha-induced reactions on ^{169}Tm and alpha-induced reaction on ^{nat}Er were derived from the measured cross sections. The results are shown in Fig. 6 and found that the deuteron-induced reaction on ^{169}Tm is preferable to produce ^{169}Yb. We can obtain ^{169}Yb without any radioactive impurities using chemical separation because co-produced ^{168}Yb and ^{170}Yb are stable.

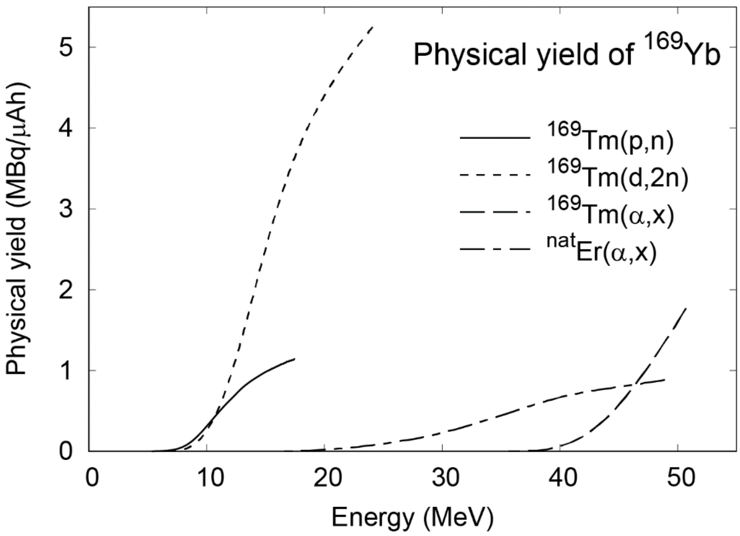

Fig. 6. Physical yields derived from the measured cross sections.

4. Conclusion

The production reactions of the medical radioisotope ^{169}Yb were investigated. The cross sections of four charged-particle-induced reactions were measured in RIKEN, Japan and ATOMKI, Hungary. Physical yields were determined from the measured cross sections and compared with each other. According to the comparison, the deuteron-induced reaction on ^{169}Tm is the most appropriate for the production.

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