The REMIX Project: Early Results on the Determination of the XS for the Production of Theranostic-Relevant Isotopes of Terbium by Deuteron Irradiation of Dysprosium Foils

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INTRODUCTION

REMIX project pursuits the goal of studying and optimizing the production routes of ^{47}Sc and terbium isotopes using light charged particles. It has a duration of three years and was funded by INFN-CSN5 in 2021 [1]. The MI section of this project is devoted to the optimization of the production of terbium radionuclides using deuteron and alpha particle beams.

Terbium family presents four radioisotopes that could be exploited for nuclear medicine applications [2]. ^{155}Tb ($T_{1/2}$ = 5.32 d) and ^{152}Tb ($T_{1/2}$ = 17.5 h) can be employed for SPECT and PET respectively. The decay properties of ^{161}Tb ($T_{1/2}$ = 6.89 d) make its employment suitable in metabolic radiotherapy, due to its intense Auger emission. Auger electron-emitting isotopes are high LET therapeutic agent and have the possibility to destroy cancer cells by depositing the energy of their ionizing radiation over a short range [3]. Lastly, ^{149}Tb ($T_{1/2}$ = 4.1 h) exhibits theranostic features. It can be used in targeted radiotherapy with alpha particle, and due to its combined β^+ emission, it can be employed for PET imaging at the same time.

In this contribution we present early results relative to the production of ^{155}Tb and ^{161}Tb via deuteron induced nuclear reactions on dysprosium targets with the natural isotopic abundance. The cross-sections for the co-production of ^{156}Tb and ^{160}Tb contaminants are reported too.

At the moment, there is only one previous work relative to this subject, whose results will be compared with our experimental data [4].

MATERIALS AND METHODS

The cross-sections (XSs) were determined by using the stacked-foils technique. Two different stacks have been assembled and irradiated, each of them containing four ^{nat}Dy targets (purity>99%, purchased from GoodFellow). After each Dy target, natural aluminum foils were placed with catcher and degrader role, but they were also used, together with titanium targets, as monitor foils.

Irradiations were carried out by accelerating deuterons with the high intensity and high energy cyclotron (AVF IBA-C70XP, K =70) of ARRONAX research center in Saint-Herblain, France. The two irradiations were performed with an energy interval of about 1.5 MeV to cover the energy range between 24 MeV and 32 MeV, with a constant current of about 150 nA for a duration of one hour each. The stacks were irradiated in air at about 7 cm from the end of the beam line, where a Kapton foil (thickness 50 μm) is used to close the line.

To evaluate the current stability during the experiment, a beam dump combined with a current integrator was used. The energy of the incident deuteron beam on each foil of the stack was calculated by performing a simulation with SRIM software [5].

Intensity and energy of the beam were verified by comparing our experimental cross-sections of the monitor reactions, ^{27}Al(d,αp)^{24}Na and ^{nat}Ti(d,x)^{48}V, with the ones recommended by IAEA [6].

The produced radioisotopes activity within each target was measured at LASA laboratory, Segrate (MI), using high resolution gamma-ray spectrometry technique with four HPGe detectors. The relevant nuclear data used are reported in Table 1.

TALYS 1.96 [7] calculations were performed using the default options for the input parameters, in order to compare the experimental XSs with the theoretical ones.

Table 1. Nuclear data of ^{155}Tb, and ^{161}Tb and the co-produced terbium contaminants: ^{156}Tb and ^{160}Tb [8].

Nuclide	Half-life	E_γ [keV]	I_γ [%]
^{155}Tb	5.32 d	105.305 ± 0.003	25.1 ± 0.5
		180.103 ± 0.001	7.45 ± 0.15
		262.322 ± 0.002	5.29 ± 0.50
^{156}Tb	5.35 d	199.213 ± 0.001	40.9 ± 0.1
		356.426 ± 0.005	13.6 ± 0.3
		534.318 ± 0.011	66.59 ± 0.16
		1222.36 ± 0.04	31.0 ± 1.6
^{160}Tb	72.3 d	298.580 ± 0.002	26.1 ± 0.6
		879.383 ± 0.003	30.1 ± 0.6
		1177.962 ± 0.004	14.9 ± 0.3
^{161}Tb	6.89 d	74.5671 ± 0.0002	10.2 ± 0.5

EXPERIMENTAL RESULTS

Activation XSs of the terbium radioisotopes produced via the ^{nat}Dy(d,x)^{1xx}Tb reactions are reported in Figs. 1-4 compared with the theoretical calculations.

When the direct production of a radioisotope cannot be isolated from the contribution of the radioactive decay of the co-produced father, the reaction will be marked with (cum).

The production XS of ^{155}Tb is reported in *Figure 1*. Both the direct reactions and the decay chain ^{155}Ho (48 min) →^{155}Dy (9.9 h) →^{155}Tb (5.32 d) contribute to the production of this radioisotope. There is a slight disagreement with TALYS simulation and the previous experimental data [4].

In *Figure 2* we report the XS relative to the reactions ^{nat}Dy(d,x)^{156}Tb. The data are compatible with the experimental results of the work of Tárkányi et al. [4]. There is a discrepancy of about a factor 2 between the theoretical

results and our experimental data.

The XS relative to the production of ^{160}Tb is displayed in *Figure 3*. There is a major discordance with TALYS simulation, while there is accordance with the previous experimental results [4] up to about 30 MeV.

Lastly, the XS of the production of the therapeutic ^{161}Tb is shown in *Figure 4*. TALYS code do not properly reproduces the experimental results, while there is agreement with the other experimental points [4].

In *Figure 5* is displayed the XS of ^{nat}Ti(d,x)^{48}V that is one of the monitor reactions used to evaluate the correctness of the intensity and the energy of the beam. The agreement with the recommended cross section is high [6].

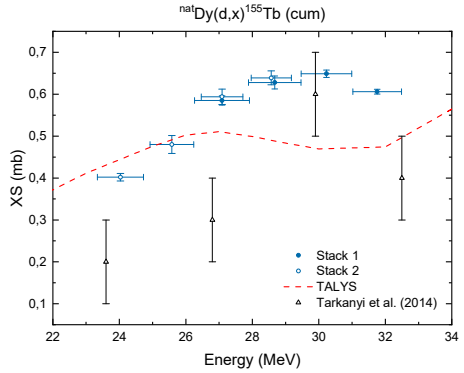

Figure 1. Experimental and theoretical XS for ^{nat}Dy(d,x)^{155}Tb (cum).

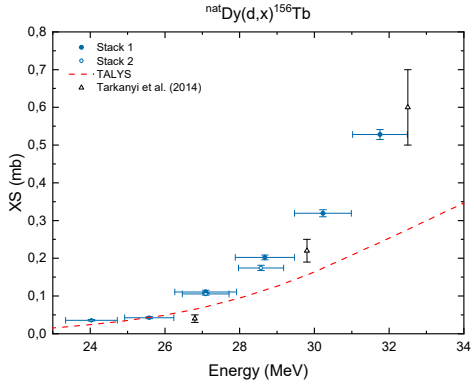

Figure 2. Experimental and theoretical XS for ^{nat}Dy(d,x)^{156}Tb.

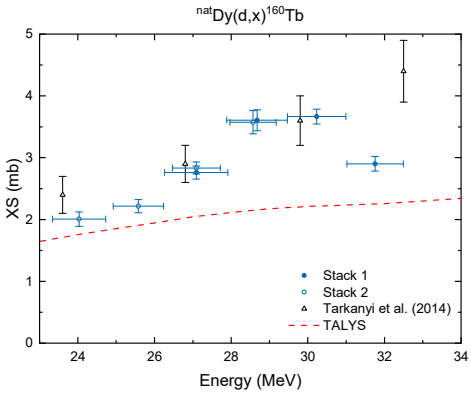

Figure 3. Experimental and theoretical XS for ^{nat}Dy(d,x)^{160}Tb.

CONCLUSIONS

In this report we presented the early results of the experimental XSs of the ^{nat}Dy(d,x)^{1xx}Tb reactions. The XSs were determined in the energy range between 24 MeV and 32 MeV.

Only one previous dataset was found in literature and there is general accordance with the results of this experiment [4].

The theoretical calculations need to be improved because the reproducibility of the experimental data is limited.

The experimental XSs of the production of the medical isotopes of terbium presented in this contribute are low. Nevertheless, a complete determination of the XSs curves down to lower energies will allow to determine the energy range where the specific activity and the radionuclidic purity of the radioisotope of interest are higher and to compare them with the results of the other production routes that will be investigated in the near future.

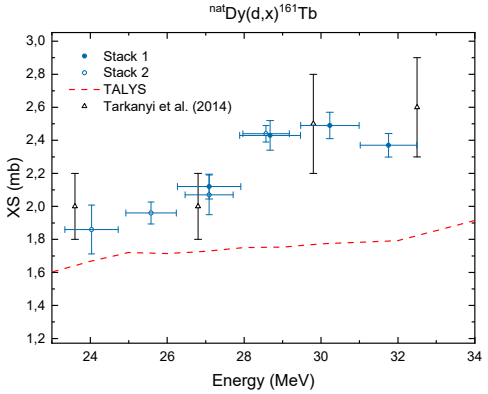

Figure 4. Experimental and theoretical XS for ^{nat}Dy(d,x)^{161}Tb.

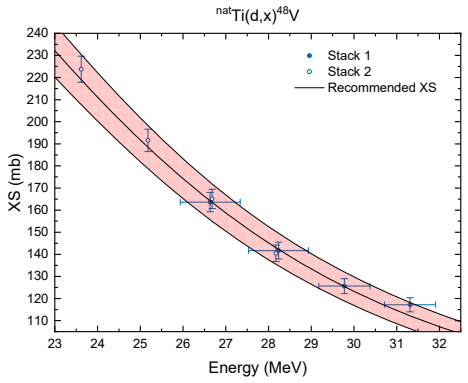

Figure 5. Experimental XS for ^{nat}Ti(d,x)^{48}V and IAEA recommended XS [6].

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