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⁴⁷Sc production with proton beams on isotopically enriched ⁴⁸Ti and ⁴⁹Ti targets

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Abstract. The theranostic emerging radionuclide 47 Sc is under the spotlight of the scientific community due to its medically favorable decay characteristics ($t_{1/2}$ =3.3492 d, E_{γ} =159.381 keV, $E_{\beta^-,mean} = 162.0$ keV) for both the diagnostic and treatment procedures. In the framework of the LARAMED (LAboratory of RAdionuclides for MEDicine) program at INFN-LNL (National Institute of Nuclear Physics-Legnaro National Laboratories), the investigation of the possible 47 Sc production routes using a 70 MeV proton beam is one of the goals of the REMIX (Research on Emerging Medical radIonuclides from the X-sections) project, funded by INFN for the years 2021-2023. Since the LARAMED bunkers are currently under completion, experiments are performed in collaboration with the GIP ARRONAX facility (Nantes, France), where a 70 MeV multi-particle cyclotron is operational. In this work, the cross-section measurements using enriched ^{48,49}Ti targets are reported and the results presented in comparison to the previous literature data, when available. The ⁴⁷Sc excitation functions are analyzed in relation to the contaminants' ones since the co-produced radionuclides can affect the dose delivered to a patient. ⁴⁶Sc cross-section curves are mainly taken into account since ⁴⁶Sc cannot be chemically separated from ⁴⁷Sc and its half-life ($t_{1/2}$ =83.79 d) is longer than the ⁴⁷Sc one ($t_{1/2}$ =3.3492 d).

1. Introduction

Theranostics, the new standpoint of nuclear medicine, benefits from the physical properties of some radionuclides to perform both the diagnosis and treatment of human diseases. The radionuclides employed in such procedures should have similar chemical characteristics to label the same biologically active molecule. For this reason, it is advantageous if the theranostic pair encompasses two radioisotopes of the same element or even only one radioisotope if the decay characteristics are suitable for both the diagnosis and the therapy. The latter is the case of 4^{7} Sc, which emits a γ -ray at 159.381 keV (see table 1) suitable for SPECT (Single Photon Emission Computed Tomography) imaging, accompanying a low-energy β -particle (E_{β -.mean}=162.0 keV, $I_{\beta^{-}} = 100\%$ [1]) that can be employed for RadioNuclide Therapy (RNT).

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The scientific community interest in the 47 Sc, highligthed in the IAEA (International Atomic Energy Agency) CRP (Coordinated Research Project) "Therapeutic Radiopharmaceuticals Labelled with New Emerging Radionuclides (67 Cu, 186 Re, 47 Sc)" [2], requires the investigation of the possible production routes of this radionuclide. In the context of LARAMED [3, 4], at INFN-LNL, two projects focused on the study of production cross-sections of 47 Sc and its contaminants. The first one, the PASTA (Production with Accelerator of Sc-47 for Theranostic Applications) project [5, 6, 7, 8], funded by INFN for the years 2017-2018, considers the use of protons on nat V and enriched 48 Ti. The second one, the REMIX (Research on Emerging Medical radIonuclides from the X-sections) project, funded by INFN for the years 2021-2023, is considering the proton-induced reactions on enriched 49 Ti and 50 Ti.

In this work, the ⁴⁸Ti(p,x)⁴⁷Sc cross-section is reported in comparison to the ⁴⁶Sc excitation function to search for a possible energy interval where the ⁴⁷Sc production is maximized while the ⁴⁶Sc one minimized. Among all the co-produced contaminants that can potentially contribute to the dose delivered to a patient, particular attention is given to ⁴⁶Sc which has two high branching ratio γ -rays highly impacting on the total dose (E_{γ 1}=889.381 keV, I_{$\gamma1$}=99.984 %, E_{$\gamma2$}=1120.545 keV, I_{$\gamma2$}=99.987 %). Moreover, ⁴⁶Sc cannot be chemically separated from ⁴⁷Sc or waited for its decay since its half-life (t_{1/2}=83.79 d) is longer than the ⁴⁷Sc one (t_{1/2}=3.3492 d). Also preliminary values of the ⁴⁹Ti(p,x)^{47,46}Sc cross-sections are presented.

2. Materials and methods

Thin (about 1 μ m) enriched ^{48,49}Ti targets, manufactured at INFN-LNL, were used for crosssection measurements. The ⁴⁸Ti enriched metallic powder, purchased from TRACE Sciences International (Richmond Hill, Ontario, Canada) with an isotopical enrichment purity of 99.32%, was deposited on a 25 μ m thick Al foil using the HIVIPP (HIgh energy VIbrational Powder Plating) technique [9, 10]. The ⁴⁹Ti powder instead, supplied by Oak Ridge (Oak Ridge, Tennessee, USA) with an isotopical enrichment of 96.25%, was delivered in a sponge-like shape and required a cryogenic milling process before the deposition on Al supports. The targets were characterized by Elastic Backscattering Spectroscopy (EBS) analysis to quantify the exact amount of enriched powder deposited and the level fo impurities.

Since the LARAMED bunkers are still under completion, targets were irradiated with the tunable 35-70 MeV proton beams delivered by the IBA cyclotron at the GIP ARRONAX facility (Saint Herblain, Nantes, France) [11]. The samples were arranged in stacked-foil targets including Ni monitor and Al thick degrader layers to bombard several enriched Ti targets in each irradiation run. In total, 7 targets of ⁴⁸Ti and 12 targets of ⁴⁹Ti have been bombarded. Beam current of at least 100 nA was used and the irradiation runs lasted about $1\div1.5$ h. After a proper cooling time from the End Of Bombardment (EOB), each enriched Ti target was measured with a HPGe detector calibrated in efficiency and energy to obtain the activity of all produced radionuclides. For several days after the irradiation, additional γ -spectra of each foil were collected (at least 5 acquisitions) to follow the decay of all the produced radionuclides and to check eventual γ -interferences. The γ peaks identified in the spectra to quantify the activity of ⁴⁷Sc and ⁴⁶Sc are reported in Table 1.

3. Experimental results and discussion

3.1. 48 Ti results

The cross-section values for the production of ${}^{47}Sc$ and ${}^{46}Sc$ obtained using protons on ${}^{48}Ti$ are graphed together with the few previous literature data available [12, 13] and a theoretical curve produced by TALYS 1.95 code [14] run with default parameters. In figure 1(a), the trend reported by our new data is in agreement with the literature and TALYS estimations. However, our obtained values have a difference up to 20% with the Gadioli *et al* [12] and the corrected Levkovski^{*} [13, 15] results. It has to be noted that the theoretical estimation

Table 1. Half-life, decay mode, energy and intensity of the γ -ray used in the analysis for the radionuclides considered [1].

Radionuclide	$t_{1/2}$ (d)	Decay mode	$E_{\gamma} (keV)$	$\mathrm{I}_{\gamma}~(\%)$
$\frac{^{47}\mathrm{Sc}}{^{46}\mathrm{Sc}}$	$\begin{array}{c} 3.3492 \ (6) \\ 83.79 \ (4) \end{array}$	$egin{array}{c} eta^- \ (100\%) \ eta^- \ (100\%) \end{array} \ eta^- \ (100\%) \end{array}$	$\begin{array}{c} 159.381 \ (15) \\ 889.277 \ (3) \end{array}$	$\begin{array}{c} 68.3 \ (4) \\ 99.984 \ (1) \end{array}$

largely overestimates all experimental values. The ${}^{46}Sc$ cross-section values, in figure 1(b), are in agreement with the Gadioli et al [12] data and with TALYS, in the energy range investigated.

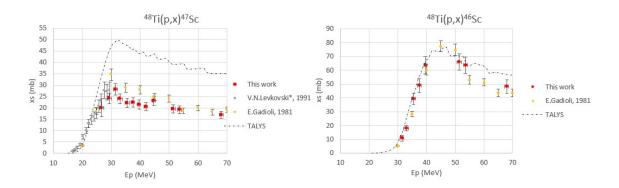


Figure 1. Cross-sections of ${}^{48}\text{Ti}(p,2p){}^{47}\text{Sc}$ reaction (a), and of ${}^{48}\text{Ti}(p,x){}^{46}\text{Sc}$ reaction (b).

3.2. ⁴⁹ Ti results

The preliminary cross-sections of ⁴⁶Sc and ⁴⁷Sc induced by protons on ⁴⁹Ti targets are graphically represented in figure 2. The results are still preliminary because the EBS analysis are not yet completed. Our measurement, in figure 2(a), is the first for the ⁴⁹Ti(p,x)⁴⁷Sc reaction so only the results about ⁴⁶Sc, in figure 2(b), are shown with a previous set of data by Levkovski^{*} [13].

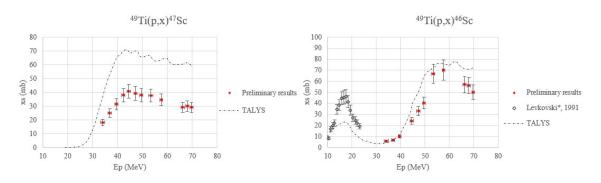


Figure 2. Cross-sections of ${}^{49}\text{Ti}(p,x){}^{47}\text{Sc}$ reaction (a), and of ${}^{49}\text{Ti}(p,x){}^{46}\text{Sc}$ reaction (b).

Similarly to the previously presented ${}^{48}\text{Ti}(p,x){}^{47}\text{Sc}$ data, our ${}^{47}\text{Sc}$ cross-section points are well below the theoretical values while the ⁴⁶Sc values are in good agreement with the TALYS estimations.

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4. Conclusions

The aim of the PASTA and REMIX projects is to determine the optimal irradiation conditions to maximize the production of 47 Sc while minimizing the co-production of radioisotopic contaminants. To achieve a precise knowledge about the cross-sections, different target materials (nat V and Ti isotopes) have been irradiated and some measurements are still ongoing. In this work, proton-induced nuclear reactions on enriched metallic 48,49 Ti targets are presented. Particular attention is given to 46 Sc since it cannot be decreased by extending the post-irradiation decay time. From a preliminary evaluation of the results it seems that these nuclear reactions cannot be used for the 47 Sc production due to the presence of other isotopic contaminants. Anyways, further investigations concerning thick targets yields and dosimetric calculations have to be done to evaluate the feasibility of these 47 Sc production routes.

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