

# First production of the medical radionuclide $^{103}\text{Pd}$ from $^{99}\text{Tc}$ and determination of the $^{99}\text{Tc}(^7\text{Li},^3\text{n})^{103}\text{Pd}$ nuclear reaction cross-sections between 20 and 34 MeV

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## Research Article

**Keywords:** Technetium-99, Palladium-103, Lithium-7 ion beam, Electrodeposition, Thin film, In-beam  $\gamma$ -ray spectroscopy, Rutherford backscattering (RBS)

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reaction cross-sections between 20 and 34 MeV**

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## Abstract

$^{103}\text{Pd}$  is a possible candidate for radiation therapy using Auger-Meitner electrons. The cross-sections of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  nuclear reaction were measured for the first time in the energy range from 20 MeV to 34 MeV using the 10 MV FN-Tandem Accelerator at the Institute of Nuclear Physics, University of Cologne. The  $^{99}\text{Tc}$  target was prepared by electrodeposition of ammonium pertechnetate onto gold foil. The thickness of the technetium layer was determined by Rutherford backscattering (RBS). The number of produced  $^{103}\text{Pd}$  nuclei was quantified with the HORUS setup by measuring prompt  $\gamma$ -rays emitted from excited  $^{103}\text{Pd}$  nuclei in parallel to irradiation (in-beam  $\gamma$ -ray spectroscopy). The maximum of the excitation function was found at an energy of 30 MeV. At higher projectile energies, contributions from the competing  $^{99}\text{Tc}(^7\text{Li},4\text{n})^{102}\text{Pd}$  reaction channel are likely to affect the observed cross-sections.

## Keywords

Technetium-99 · Palladium-103 · Lithium-7 ion beam · Electrodeposition · Thin film · In-beam  $\gamma$ -ray spectroscopy · Rutherford backscattering (RBS)

## Introduction

The radionuclide  $^{99}\text{Tc}$  is only rarely used as target material in nuclear reactions, although it accumulates in large quantities in spent nuclear fuel and its further use could contribute to a reduction of long-lived radioactive waste. Owing to its long half-life of  $2.111 \cdot 10^5$  years [1] and its low beta energy, the handling of  $^{99}\text{Tc}$  even in gram amounts does not require heavy shielding. Various neutron-induced reactions on  $^{99}\text{Tc}$  have been investigated, such as  $^{99}\text{Tc}(\text{n},\gamma)^{100}\text{Tc}$  (Furutaka et al. [2], Qaim [3]),  $^{99}\text{Tc}(\text{n},\text{p})^{99}\text{Mo}$  (Golchert et al. [4], Reimer et al. [5], Qaim [3]),  $^{99}\text{Tc}(\text{n},2\text{p})^{98}\text{Nb}$  (Qaim [3]),  $^{99}\text{Tc}(\text{n},\alpha)^{96}\text{Nb}$  (Golchert et al. [4], Reimer et al. [5], Qaim [3]), and  $^{99}\text{Tc}(\text{n},^3\text{He})^{97}\text{Nb}$  (Qaim [3]). In contrast charged-particle-induced reactions on technetium have been studied to a much lesser extent. The  $^{99}\text{Tc}(\text{p},3\text{n})^{97}\text{Ru}$  reaction was investigated by Zaitseva et al. [6] up to 100 MeV proton energy. Titarenkov et al. [7] reported irradiations of  $^{99}\text{Tc}$  with high energy protons between 0.1 GeV and

2.6 GeV energy. However, no experimental data for irradiations with heavier projectiles like  $\alpha$ -particles or  $^7\text{Li}$  ions can be found in the literature. The  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction investigated in this work leads to the formation of the medically relevant radionuclide  $^{103}\text{Pd}$  (half-life: 16.99 d), which decays by electron capture to  $^{103\text{m}}\text{Rh}$  (half-life: 56.1 min). Because the associated  $\gamma$ -ray intensities are very low, the activity of  $^{103}\text{Pd}$  cannot be quantified accurately by conventional post-irradiation  $\gamma$ -ray spectrometry. The weak  $\gamma$ -emission is accompanied by high emission yields of conversion electrons (CE) and Auger-Meitner electrons (AME). In radiotherapy, this allows for the irradiation of small tumours (e.g. metastases) without applying radiation dose to the surrounded tissue. These electrons are used in nuclear medicine in prostate brachytherapy [8,9]. Current research is focusing on the use of  $^{103}\text{Pd}$ , alongside  $^{177}\text{Lu}$  and  $^{161}\text{Tb}$ , in the targeted radionuclide therapy (TRT) [10] to treat prostate cancer.  $^{103}\text{Pd}$  has mainly been produced by proton irradiation either via the  $^{\text{nat}}\text{Ag}(\text{p},\text{x})^{103}\text{Pd}$  reaction as investigated e.g. by Fassbender et al. [11], Uddin et al. [12] Katcoff et al. [13], or via the  $^{103}\text{Rh}(\text{p},\text{n})^{103}\text{Pd}$  reaction described e.g. by Sudar et al. [14] and Hermanne et al. [15]. A comprehensive evaluation of charged-particle induced reactions leading to  $^{103}\text{Pd}$  can be found in [16]. Compared with these established methods, production of  $^{103}\text{Pd}$  via the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction offers relatively simple chemical separation of the product nuclide from the target material. Under oxidative conditions technetium is present as an anion, whereas the other commonly used target metals (Ag, Rh) and Pd occur as cations in acidic solution. Consequently,  $^{103}\text{Pd}$  can be separated from Tc by cation-exchange chromatography more straightforwardly than from Ag or Rh based targets. In this study, however, no chemical separation was carried out, as the Tc target is to be reused for other experiments. A major limitation of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  production route is the limited availability of accelerators capable of delivering  $^7\text{Li}$  ion beams with sufficiently high beam currents. This capability is typically not provided by commercial cyclotrons.

In this work, the production of  $^{103}\text{Pd}$  via the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction is investigated for the first time. The excitation function of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction was measured in the energy region from 20 MeV to 34 MeV by irradiating a thin film of  $^{99}\text{Tc}$  deposited on gold foil. This would enable a simple chemical processing of the target material because it can selectively be etched from the target with various acids without dissolving the gold foil.

The irradiations were performed at the 10 MV FN-Tandem Accelerator at the Institute of Nuclear Physics, University of Cologne. The number of produced  $^{103}\text{Pd}$  nuclei was determined with the HORUS [17,18] setup by measuring  $\gamma$ -rays emitted from excited  $^{103}\text{Pd}$  nuclei in parallel to irradiation (in-beam  $\gamma$ -ray spectroscopy).

## Experimental

### *Target preparation*

The  $^{99}\text{Tc}$  target was prepared by electrodeposition from of an aqueous ammonium pertechnetate solution, following the procedure reported by Voltz and Holt [19]. The electrolyte consisted of the  $^{99}\text{Tc}$  stock solution (ammonium pertechnetate in 0.1 M  $\text{NH}_4\text{OH}$ , Amersham, England; 17.6 MBq/mL, 0.28 mol/L) and ammonium sulfate (for molecular biology,  $\geq 99.0\%$ , Sigma, Germany) solution (1 mol/L). The ammonium pertechnetate solution was filtered through a PTFE syringe filter before use. The pH of the electrolyte was adjusted to 1 by the addition of concentrated sulfuric acid (Suprapur<sup>®</sup>, min. 96 %, Merck, Germany). The electrolysis cell (Figure 1), fabricated from PLA, featured a circular opening with a diameter of 10 mm and at its base a gold foil (99.9 %, 200 mg, 50  $\mu\text{m}$ , Goodfellow, Germany) of 15 mm diameter was placed. The gold foil (anode) was in electrical contact with the steel base and served as the working electrode. A platinum wire as the cathode and an Ag/AgCl (in 3 M KCl solution) reference electrode were immersed in the electrolysis solution from the top side. The electrolysis cell was mounted on a Peltier cooling element operated at 5 V DC, to suppress the forming of hydrogen. The Peltier element was coupled to an ice bath to ensure efficient heat dissipation. The three electrodes were connected to a potentiostat (Ossila T2006) and a potential of  $-0.85\text{ V}$  was applied for 60 minutes. Following this electrolysis, a shiny silver-grey metallic deposit was obtained.

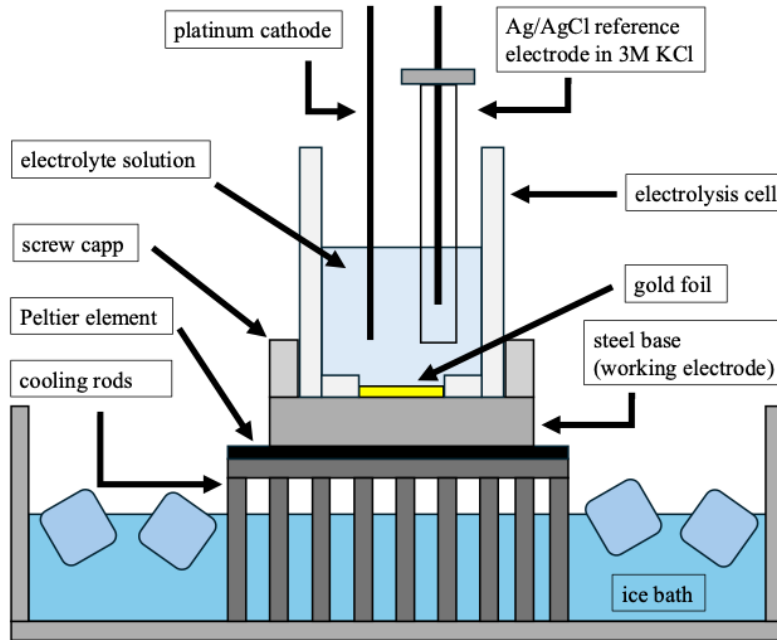


**Figure 1.** Sketch of the electrodeposition setup used for the production of the  $^{99}\text{Tc}$  target.

#### *Measurement of target thickness*

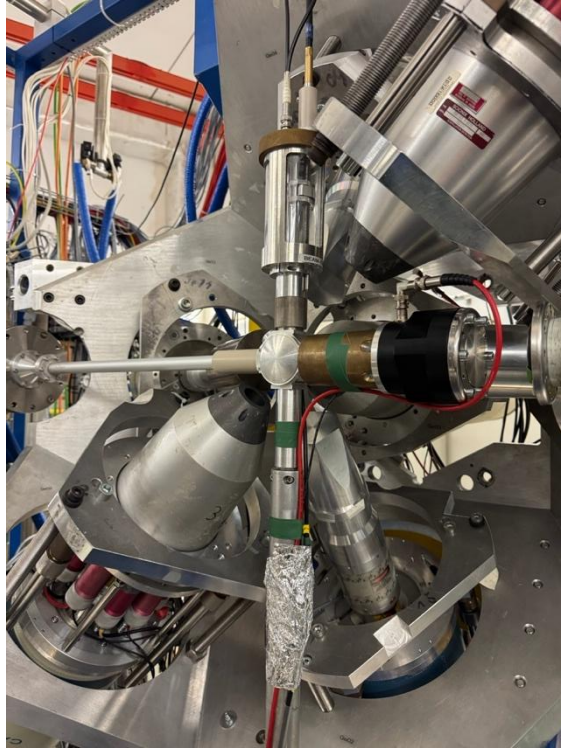
The thickness of the technetium layer on the Tc/Au target was determined by Rutherford backscattering (RBS) at the Cologne AMS 6 MeV Tandatron Accelerator of the Institute of Nuclear Physics, University of Cologne, using a beam of 7 MeV  $^9\text{Be}$  ions. Measurements were performed on four different positions on the target surface. The spectra were evaluated with the software *SIMNRA* [20]. The target was modelled within *SIMNRA* as a two-layer system consisting of pure technetium and pure gold, respectively. All spectra were fitted with the same values for energy resolution. Likewise, the same roughness corrections were applied to all simulations. The electronic stopping powers were calculated using the Ziegler-Biersack model. The RBS analysis yielded consistent areal densities for the technetium layer at all four measurement positions ( $4.3 \cdot 10^{18}$ ,  $4.55 \cdot 10^{18}$ ,  $4.4 \cdot 10^{18}$  and  $4.55 \cdot 10^{18}$  atoms/cm<sup>2</sup>, respectively), indicating uniform electrolytic deposition of the technetium on the gold foil. A marginally improved fit quality within *SIMNRA* was obtained by applying a correction factor of 0.85 to the stopping power of beryllium in gold.

However, this adjustment had no significant effect on the derived layer thicknesses. The overall uncertainty of the *SIMNRA* fitting was conservatively estimated to be approximately 5 %. The determined average areal density of  $4.45 \cdot 10^{18}$  atoms/cm<sup>2</sup> is corresponding to a technetium layer thickness of  $0.63 \pm 3$   $\mu$ m, assuming the bulk density of metallic technetium. This value is in good agreement with independent estimates based on the decrease in activity of the technetium solution during electrolytic deposition. The latter was assessed by liquid scintillation counting (LSC) of the electrolytic solution before and after deposition. However, this approach was considered less accurate, as it does not account for potential technetium losses due to adsorption of Tc e.g. on the surface of the electrolysis cell or the reference electrode.

#### *Irradiation and measurement setup*

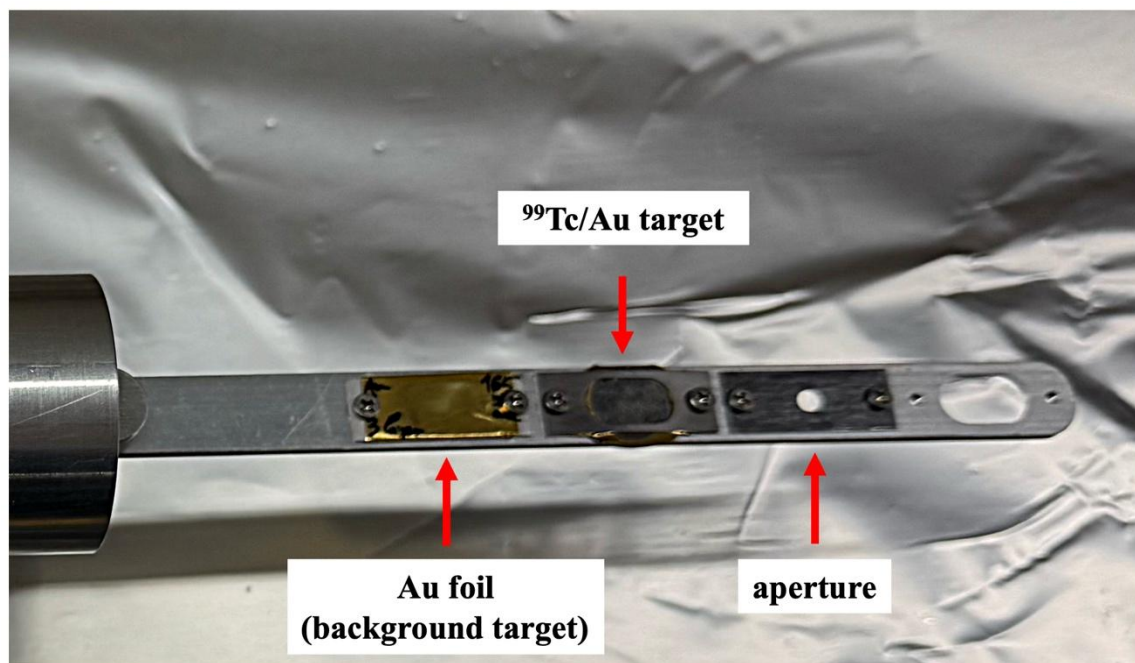
To determine the excitation function of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction, the  $^{99}\text{Tc}$  target was irradiated at the 10 MV FN-Tandem Accelerator Laboratory at the University of Cologne, using a  $^7\text{Li}$  beam in the 3+ charge state at beam energies from 20 MeV to 34 MeV with average beam currents of 5 nA to 10 nA. The prompt  $\gamma$ -rays emitted from excited  $^{103}\text{Pd}$  nuclei were detected during the irradiation (in-beam  $\gamma$ -ray spectroscopy) with the HORUS  $\gamma$ -ray spectrometer [17] (see Figure 2). The experimental setup consisted of 14 High-Purity-Germanium detectors (HPGe) positioned at five different angles (35°, 45°, 90°, 135°, and 145°) relative to the beam axis. Prior to irradiation, the detectors were energy and efficiency calibrated using a  $^{226}\text{Ra}$  standard source with a summed full-energy-peak efficiency of 0.1 % to 0.4 % at 1.12 MeV and of 0.2 % to 1.0 % at 242 keV. An electrically isolated target chamber was installed within the HORUS spectrometer, enabling direct measurement of the accumulated beam charge and thus the determination of absolute reaction cross-sections [18]. To suppress  $\gamma$ -ray background originating from scattered beam particles interacting with the aluminium chamber walls, the interior surfaces of the chamber are coated with a thin tantalum layer. Particular attention was paid to preventing unintended electrical contact between chamber components and ground. In addition, the chamber was equipped with a motorized, electrically isolated target ladder, allowing convenient switching between target and aperture positions without compromising the electrical

isolation of the setup. Besides an empty frame and an 3 mm aperture for beam focusing, the ladder was equipped with the  $^{99}\text{Tc}/\text{Au}$  target itself and a thick gold target for later gold background subtraction (Figure 3). Both gold layers ( $\text{Tc}/\text{Au}$  target and Au target) were selected to be sufficiently thick to completely stop the unreacted  $^7\text{Li}$  ion beam on target position. Irradiation times were typically 30 to 45 minutes for the  $\text{Tc}/\text{Au}$  target and approximately 10 minutes for the Au target, yielding excellent counting statistics in each detector and at each beam energy. Depending on the beam energy and target, total HPGe count rates ranged from 5 kcps to 20 kcps. Detector dead times were monitored online by the digital data acquisition system and varied between 1.8 % and 28.5 %. The incident beam current was measured using a digital current-integration system that independently recorded the currents collected on the target ladder and on the target chamber. To suppress the loss of secondary electrons, a bias voltage of  $-400\text{ V}$  was applied to an aperture located at the entrance of the target chamber. This voltage reflected  $\delta$ -electrons generated by the beam back onto the chamber, resulting in a negative current contribution on the chamber. The total number of incident beam particles for each irradiation was determined from the sum of the positive charge accumulated on the target ladder and the negative charge accumulated on the chamber, taking into account the charge state of the beam ions.



176

177 **Figure 2.** Part of the HORUS  $\gamma$ -ray spectrometer. Beam direction from the left side. Target  
178 chamber and target ladder in the middle surrounded by HPGe  $\gamma$ -ray detectors.  
179



180

181 **Figure 3.** Target ladder used for the irradiations. The Au foil (left) for background  
182 correction, the <sup>99</sup>Tc/Au target (middle) and the aperture (right) can be seen.

## *Results and discussion*

For the determination of the cross-sections, it was assumed that  $^{103}\text{Pd}$  is exclusively formed in an compound state which decays via a  $\gamma$ -ray cascade through any of the first (few) excited states and that direct population of the ground state does not occur, which is considered a reasonable assumption for the used projectile energies and reaction channel. The excited nuclear states of  $^{103}\text{Pd}$  de-excite through a cascade of  $\gamma$ -ray transitions involving the excited states at 243.96 keV (nuclear spin state  $7/2^+$ ), 531.97 keV ( $7/2^+$ ), and 718.02 keV ( $9/2^+$ ), each of which ultimately de-excites to the ground state ( $5/2^+$ ) [21]. Intensity correction factors were derived based on the level scheme reported in [21], taking into account the other decay branches of states tagged into the respective ground state  $\gamma$ -ray decay, to be 1.02 for the 243.96 keV transition, 1.11 for the 531.97 keV transition and 1.07 for the 718.02 keV transition. The gold background  $\gamma$ -ray spectra were normalized to the prompt gamma line at 279 keV which occurs in the Tc/Au target as well as the Au target and then subtracted from the cumulated  $\gamma$ -ray spectra of the irradiated Tc/Au target (Figure 4).

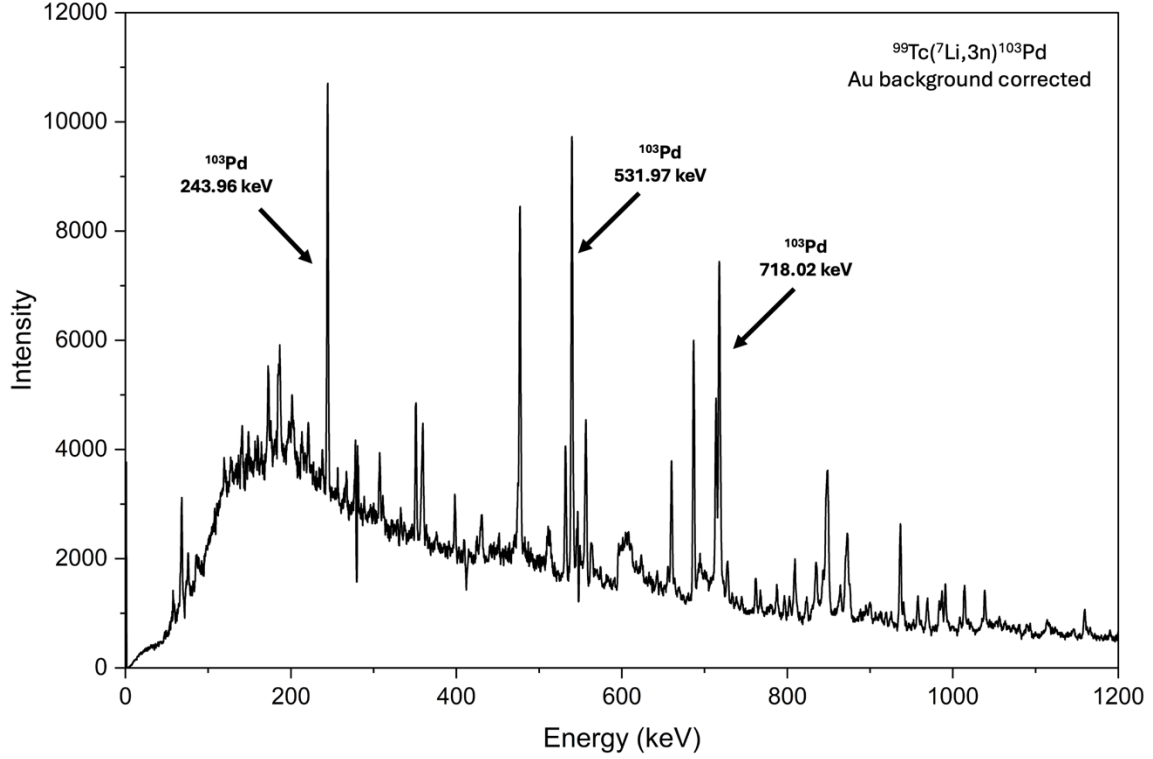


**Figure 4.** Cumulated in-beam prompt  $\gamma$ -ray spectrum of  $^{103}\text{Pd}$  (Au background corrected) from irradiation of  $^{99}\text{Tc}/\text{Au}$  with 26 MeV  $^7\text{Li}^{3+}$ . Only the most prominent peaks of  $^{103}\text{Pd}$  are marked here, which were used in the evaluation.

The number of nuclei decays  $N$  at energy  $E$  from each of these states was calculated according equation 1 from the measured  $\gamma$ -ray peak areas  $A_{\text{peak}}$  from the Au background corrected  $\gamma$ -ray spectrum considering the detector efficiency  $\varepsilon_{\text{det}}$  at energy  $E$ , the intensity of the  $\gamma$ -ray emission  $I_{\gamma}$ , the  $\gamma$ -ray emission correction factor  $c_{\gamma}$ , and dead time correction factor  $t_{\text{dead}}$ .

$$N(E) = \frac{A_{\text{peak}}}{\varepsilon_{\text{det}} \cdot I_{\gamma} \cdot c_{\gamma} \cdot (1 - t_{\text{dead}})} \quad (\text{Eq. 1})$$

Summation over all three transitions yields the total number of produced  $^{103}\text{Pd}$  nuclei  $N_{\text{total}}$ , because it is assumed that all transitions from higher states inevitably cross one of these three states before decaying into the ground state. The total cross-section value  $\sigma$  for each

detector was calculated according to equation 2 considering the number of  $^{99}\text{Tc}$  target nuclei  $N_{\text{Tc}}$ , the  $^7\text{Li}^{3+}$  particle flux  $\phi$  per  $\text{cm}^2$  and the conversion factor  $10^{27} \frac{\text{mb}}{\text{cm}^2}$  for conversion of square centimetre to millibarn.

$$\sigma = \frac{N_{\text{total}} \cdot 10^{27} \frac{\text{mb}}{\text{cm}^2}}{N_{\text{Tc}} \cdot \phi} \quad (\text{Eq. 2})$$

The so obtained total cross-sections for each detector angle are given in Table 1.

**Table 1.** Total cross-section values of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction determined from the  $35^\circ$ ,  $45^\circ$ ,  $90^\circ$ ,  $135^\circ$  and  $145^\circ$  detector rings (mean values of the cross-sections for each detector angle). Given uncertainties are the standard deviations.

| Energy (MeV) | Cross-section (mb) for detector angle $35^\circ$ | Cross-section (mb) for detector angle $45^\circ$ | Cross-section (mb) for detector angle $90^\circ$ | Cross-section (mb) for detector angle $135^\circ$ | Cross-section (mb) for detector angle $145^\circ$ |
|--------------|--------------------------------------------------|--------------------------------------------------|--------------------------------------------------|---------------------------------------------------|---------------------------------------------------|
| 20           | 46.1 $\pm$ 0.8                                   | 52.8 $\pm$ 3.1                                   | 40.2 $\pm$ 2.6                                   | 53.7 $\pm$ 2.6                                    | 50.6 $\pm$ 0.5                                    |
| 22           | 94.7 $\pm$ 3.6                                   | 105 $\pm$ 4                                      | 83.7 $\pm$ 3.7                                   | 109 $\pm$ 1                                       | 97.4 $\pm$ 6.7                                    |
| 24           | 145 $\pm$ 11                                     | 156 $\pm$ 16                                     | 117 $\pm$ 7                                      | 150 $\pm$ 3                                       | 138 $\pm$ 13                                      |
| 26           | 195 $\pm$ 8                                      | 216 $\pm$ 10                                     | 159 $\pm$ 5                                      | 210 $\pm$ 8                                       | 190 $\pm$ 8                                       |
| 28           | 241 $\pm$ 8                                      | 257 $\pm$ 24                                     | 191 $\pm$ 10                                     | 251 $\pm$ 7                                       | 222 $\pm$ 16                                      |
| 30           | 257 $\pm$ 7                                      | 282 $\pm$ 28                                     | 209 $\pm$ 15                                     | 287 $\pm$ 1                                       | 239 $\pm$ 28                                      |
| 32           | 273 $\pm$ 4                                      | 261 $\pm$ 42                                     | 189 $\pm$ 16                                     | 273 $\pm$ 9                                       | 241 $\pm$ 33                                      |
| 34           | 244 $\pm$ 5                                      | 278 $\pm$ 6                                      | 183 $\pm$ 18                                     | 275 $\pm$ 21                                      | 201 $\pm$ 22                                      |

For each beam energy, the angular dependence for the calculated cross-sections from Table 1 were fitted with a fourth-degree polynomial function. The resulting fit functions were subsequently integrated numerically over the full solid angle, considering a sphere around the irradiated target covered by the detectors. The absolute total cross-sections for each energy are shown in Table 2 and are plotted in Figure 6. The uncertainties of the cross-sections were calculated from the standard deviations of the mean cross-sections by error propagation.

**Table 2.** Total cross-section values of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction calculated based on the fitted experimental cross-section values from Table 1 by integration over the full solid angle. (\*) The data point at 34 MeV probably contains a contribution from the  $^{99}\text{Tc}(^7\text{Li},4\text{n})^{102}\text{Pd}$  reaction channel.

| Energy (MeV) | Total Cross-section (mb) |
|--------------|--------------------------|
| 20           | $70.1 \pm 7.0$           |
| 22           | $144 \pm 14$             |
| 24           | $207 \pm 36$             |
| 26           | $285 \pm 26$             |
| 28           | $341 \pm 47$             |
| 30           | $374 \pm 63$             |
| 32           | $363 \pm 83$             |
| 34           | $347 \pm 53$ (*)         |

The excitation function of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction, shown in Figure 6, exhibits a steady increase with beam energy and reaches its maximum at approximately 30 MeV. To the best of our knowledge, this represents the first experimental cross-section measurement of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction, and therefore no literature data are available for direct comparison. Above 30 MeV, the cross-section decreases gradually with increasing energy. However, the value determined at 34 MeV was neglected in the excitation function fit in Figure 6, as it may contain contributions from the competing  $^{99}\text{Tc}(^7\text{Li},4\text{n})^{102}\text{Pd}$  reaction, which sets in roughly at this energy. At this energy, the 243.96 keV and 531.97 keV lines from  $^{103}\text{Pd}$  vanish in the  $\gamma$ -ray spectrum, while the 719.38 keV  $\gamma$ -ray line originating from  $^{102}\text{Pd}$  may overlap with the 718.02 keV  $\gamma$ -ray line from  $^{103}\text{Pd}$ , potentially leading to an overestimation of the deduced  $^{103}\text{Pd}$  production cross-section.

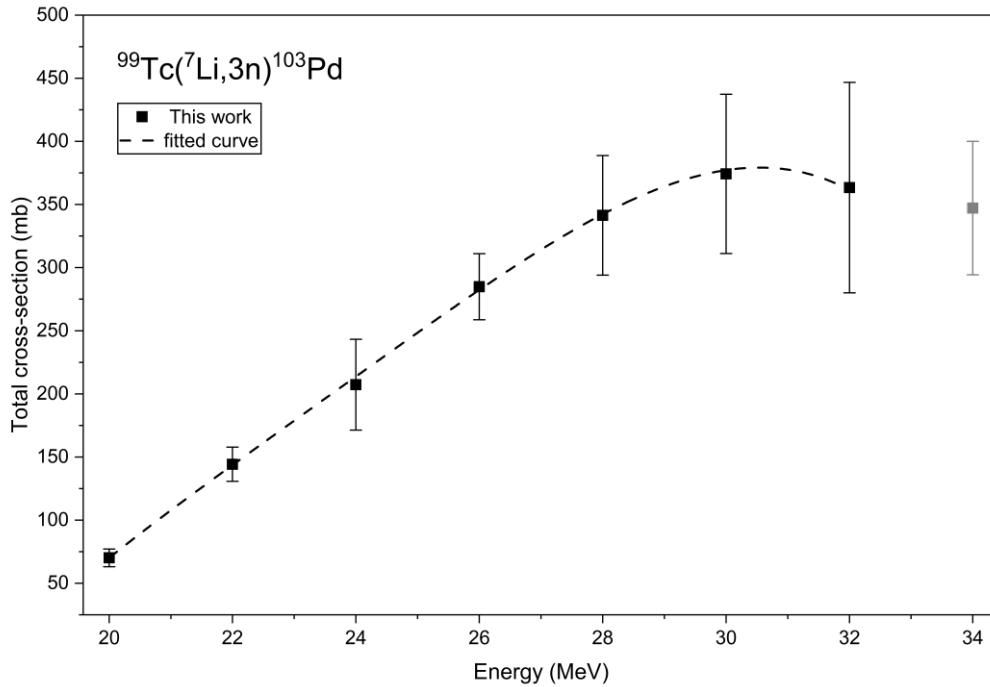


**Figure 6.** Total cross-sections of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction calculated based on the fitted experimental cross-section values by integration over the full solid angle. The  $^{99}\text{Tc}(^7\text{Li},4\text{n})^{102}\text{Pd}$  reaction probably contributes to the cross-section value at 34 MeV (grey colour).

## Conclusions

This study demonstrates that the medical radionuclide  $^{103}\text{Pd}$  can be successfully produced by irradiating a  $^{99}\text{Tc}$  target with  $^7\text{Li}^{3+}$  ions. The formation of  $^{103}\text{Pd}$  was quantified by in beam  $\gamma$ -ray spectroscopy, in which prompt  $\gamma$ -rays emitted during the de-excitation of reaction product nuclei were measured simultaneously with the irradiation. The excitation function of the  $^{99}\text{Tc}(^7\text{Li},3\text{n})^{103}\text{Pd}$  reaction was determined for beam energies between 20 MeV to 32 MeV reaching a maximum at approximately 30 MeV before decreasing at higher energies. The value at 34 MeV probably contains a contribution from the  $(^7\text{Li},4\text{n})$  reaction channel. In our view, the described irradiation strategy has two advantages for the production of medical  $^{103}\text{Pd}$ : 1) it enables to re-use  $^{99}\text{Tc}$  from fission which otherwise must be considered waste, 2) it allows for a simple future chemical separation scheme for medical  $^{103}\text{Pd}$  because Tc forms  $\text{TcO}_4^-$  anions from which the desired product  $^{103}\text{Pd}$  can be separated as  $\text{Pd}^{2+}$  cations using an ion exchange column.

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## **Author contributions**

A. Dragoun: Conceptualization, investigation, target preparation, data curation, visualization, writing – original draft, writing – review & editing. M. Weinert: Investigation, HORUS measurements, validation. A. Bohn: RBS measurements; B. Neumaier: Supervision. J. Ermert: Supervision, writing – review & editing. E. Strub: Investigation, validation, RBS evaluation, conceptualization, supervision, writing – review & editing.

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## **Declarations**

### **Data availability statement**

The recorded gamma-ray spectra and RBS measurements are available from the corresponding author upon reasonable request.

### **Conflict of interests**

The authors declare they have no competing interests.

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