Abstract 037

Cyclotron production of ^{99m}Tc via the ^{100}Mo(p,2n)^{99m}Tc reaction

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Introduction: In light of the current world-wide shortage of reactor-produced ^{99}Mo/^{99m}Tc, there is a growing interest in exploring the large-scale cyclotron production of ^{99m}Tc via the ^{100}Mo(p,2n)^{99m}Tc reaction (a method first proposed by Beaver and Hupf, *J Nucl Med*, 1971, 12: 739). In producing ^{99m}Tc, knowledge of the cross sections and theoretical yields are essential for optimizing the high-current irradiation conditions and verifying the processing/recovery techniques. A review of the existing published cross section data for the ^{100}Mo(p,2n)^{99m}Tc reaction however reveals large discrepancies in these measured values.

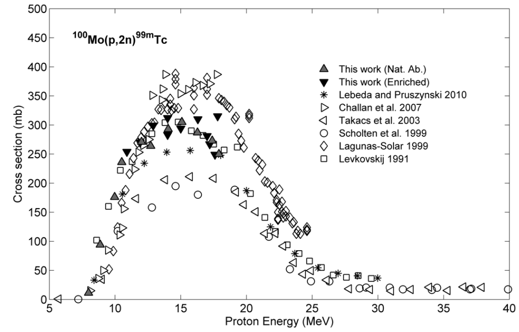
Aim: Given the large cross section discrepancies in the current literature, the goal of this work was to re-evaluate the cross sections for the ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo reactions.

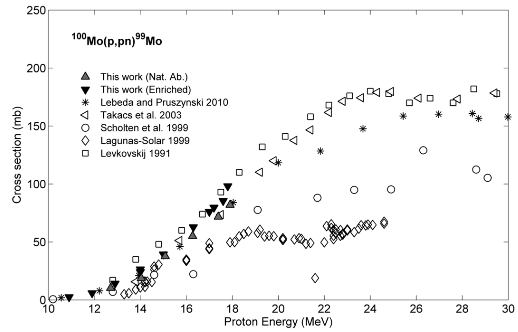
Methods: The ^{99m}Tc and ^{99}Mo cross sections were evaluated using both natural abundance (7.5 mg/cm^2) and ^{100}Mo enriched (97.42%, 7.4–11.1 mg/cm^2) foils. Foils were individually irradiated with proton energies up to 18 MeV for 600 seconds on the Edmonton PET Centre's TR 19/9 variable energy cyclotron (Advanced Cyclotron Systems Inc., Richmond, BC). A copper foil was in place for all irradiations for the purpose of monitoring the beam energy and irradiation current. Unless otherwise noted, all decay data were obtained from the NuDat 2.5 database.

The molybdenum foils were assayed multiple times (from a few hours to several days post-EOB) using an HPGe detector (sample distance ≥ 25 cm, dead time < 7%). The detector was calibrated using standard sources of ^{22}Na, ^{54}Mn, ^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba and ^{137}Cs. The ^{99}Mo activity was determined using a weighted average of the 181 keV and 739 keV peaks. In determining the ^{99m}Tc activity, the non-resolved 140/142 keV peak (89.06%/ 0.02%) was measured. Two additional contributing sources to the 140 keV peak were subtracted prior to evaluation of the direct ^{99m}Tc cross section. Firstly, as ^{99}Mo decays to ^{99m}Tc, the ^{99}Mo associated ^{99m}Tc activity at the start of counting was determined from the measured ^{99}Mo activity using the radioactive parent-daughter relationship described by Attix (*Introduction to Radiological Physics and Radiation Dosimetry*, 2004, pp. 105–106) with the branching ratio to ^{99m}Tc taken as 87.6% (Alfassi et al., *Appl Radiat Isot*, 2005 63: 37). Next, as ^{99}Mo gives rise to a 140 keV (4.52%) gamma ray upon decay, this peak contribution was calculated from the measured ^{99}Mo activity of each respective foil. Cross sections were calculated using the standard activation formula (Krane, *Introductory Nuclear Physics*, 1988, pp. 169–170) with values normalized to 100 percent ^{100}Mo enrichment.

Thick target yields were calculated from the measured ^{99m}Tc cross sections assuming 100 percent ^{100}Mo and fitting the cross-section data with a 2nd order polynomial. Values are reported for 18→10 MeV, and 22→10 MeV (cross sections extrapolated to 22 MeV from a polynomial curve fit).

Results: The following figures compare the evaluated cross sections for the direct production of ^{99m}Tc and ^{99}Mo to previously published cross section data. For the purpose of comparison, we have normalized the ^{99m}Tc data of Challan et al. (*Nucl Rad Phys*, 2007, 2: 1) to 100 percent ^{100}Mo by dividing by 9.63%. For both reactions, our results are in good agreement to values published by Levkovskij (1991). Good ^{99m}Tc cross section agreement is also noted up to E_p ~12 MeV when comparing with Lagunas-Solar (*IAEA-TECDOC-1065*, 1999) and Challan et al. We believe that the elevated ^{99m}Tc cross sections for Lagunas-Solar for E_p > ~12 MeV may be attributed to the incomplete subtraction of the ^{99}Mo 140 keV peak contributions due to underestimated ^{99}Mo cross sections. Although Challan et al. mention that they have corrected for the growth and decay of the metastable and ground states, it is unclear if the ^{99m}Tc 140 keV peaks were corrected to account for ^{99}Mo→^{99m}Tc contributions post-EOB. The absence of such a correction would similarly explain the elevated ^{99m}Tc cross sections for E_p > ~12 MeV. While the ^{99}Mo cross sections are in good agreement, the ^{99m}Tc cross sections measured in this work are significantly higher than values published by Takács et al. (*J Radioanal Nucl Chem*, 2003, 257: 195) and slightly higher, by approximately 2σ, than values presented by Lebeda and Pruszynski (to be published in *Appl Radiat Isot*). An overall disagreement was noted for both reactions when comparing with the published cross sections of Scholten et al. (*Appl Radiat Isot*, 1999, 51: 69).





E [MeV]	Foil	^{99m}Tc [mb]	^{99}Mo [mb]
8.0 ± 0.2	Nat. Ab.	11.3 ± 1.2	–
8.9 ± 0.4	Nat. Ab.	94.1 ± 8.3	–
10.0 ± 0.2	Nat. Ab.	176 ± 15	–
10.5 ± 0.6	Nat. Ab.	236 ± 21	–
12.0 ± 0.3	Nat. Ab.	271 ± 24	–
12.7 ± 0.3	Nat. Ab.	264 ± 23	10.3 ± 6.2
14.0 ± 0.3	Nat. Ab.	293 ± 26	18.7 ± 5.4
15.1 ± 0.4	Nat. Ab.	305 ± 27	37.7 ± 6.0
16.3 ± 0.5	Nat. Ab.	287 ± 25	55.1 ± 7.6
17.4 ± 0.7	Nat. Ab.	273 ± 24	71.8 ± 9.6
17.9 ± 0.8	Nat. Ab.	250 ± 22	82 ± 10
10.9 ± 0.6	Enriched	254 ± 22	2.47 ± 0.22
11.9 ± 0.2	Enriched	270 ± 24	5.82 ± 0.51
12.9 ± 0.5	Enriched	299 ± 26	14.1 ± 1.2
14.0 ± 0.2	Enriched	283 ± 25	24.1 ± 2.1
14.0 ± 0.2	Enriched	312 ± 27	26.4 ± 2.3
15.0 ± 0.2	Enriched	294 ± 26	39.4 ± 3.4
16.3 ± 0.2	Enriched	310 ± 27	62.7 ± 5.5
17.0 ± 0.3	Enriched	281 ± 24	76.1 ± 6.6
17.2 ± 0.8	Enriched	267 ± 23	79.8 ± 7.0
17.6 ± 0.8	Enriched	249 ± 22	85.4 ± 7.5
17.8 ± 0.4	Enriched	315 ± 27	98.2 ± 8.6

Throughout this experiment the beam current and detector efficiency were carefully monitored and we are confident with the 140 keV peak area corrections performed in this experiment as the evaluated ^{99m}Tc cross sections were consistent, independent of the time post-EOB upon which they were evaluated (i.e. calculated within a few hours post-EOB or >24 hours post-EOB).

Thick target yields were determined to be 14.2 mCi (526 MBq)/μAh for 18→10 MeV, and 18.2 mCi (674 MBq)/μAh for 22→10 MeV. These yields are higher than the value of 11.2 mCi (415 MBq)/μAh for 22→12 MeV reported by reported by Scholten et al., and are in good agreement with the value of 17 mCi (629 MBq)/μAh for 25→5 MeV given by Takács et al.

As we are not only interested in optimizing the yield of ^{99m}Tc, but also the purity, future work is planned to experimentally evaluate the ^{100}Mo(p,2n)^{99g}Tc cross sections. Preliminary calculations using cross sections modelled with Empire–II suggest that a ^{99m}Tc/$^{99m+99g}$Tc ratio of 18% is possible for a 3 hour irradiation at 22→10 MeV. In comparison, assuming a 24 hour elution frequency and 5% retention, the ^{99m}Tc/$^{99m+99g}$Tc ratio calculated for the standard generator setup is 26% (Alfassi et al., *Appl Radiat Isot*, 2005 63: 37).

Conclusion: We have presented updated cross sections for the ^{100}Mo(p,2n)^{99m}Tc and the ^{100}Mo(p,pn)^{99}Mo reactions. Results of this work suggest that production of large quantities of ^{99m}Tc via a cyclotron may be a viable alternative to the current reactor-based production strategy.

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