Production cross-sections of medical radioisotopes from proton bombardment of ^{nat}Mo

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This In 2009 a research program was started at the Cyclotron Institute to study the production of medical radioisotopes using beams of protons, alphas and helium-3 from the K500 superconducting cyclotron. Radioactive nuclides are mainly used in medicine for diagnostic (PET and SPECT imaging) and therapeutic (radiopharmaceuticals and brachytherapy) purposes.

There have been 8 experiments performed as part of this program. The present work will focus on results of the analysis of the first experiment in the series. The motivation for it was two-fold. First, it was a study of the excitation function of ^{99m}Tc, commonly used in SPECT imaging, and of the potential for medical applications of the other residual radioisotopes. Second, it was a test on the effectiveness of the setup and the viability of the production mechanism as an alternative to nuclear reactors and generators.

The analysis was based on the activation technique. A stack of Mo, Cu and Al foils was irradiated with a beam of protons with energy of 40 MeV/n. The gammas emitted from the activated foils were measured after a short cooling down period with a HPGe detector. A detailed description of the experimental set-up, procedure and the analysis steps can be found in a previous work [1]. The Cu and Al foils were used for energy degrading and beam normalization purposes. Figure 1 shows the beam energy after each foil as well as the order in the stack.

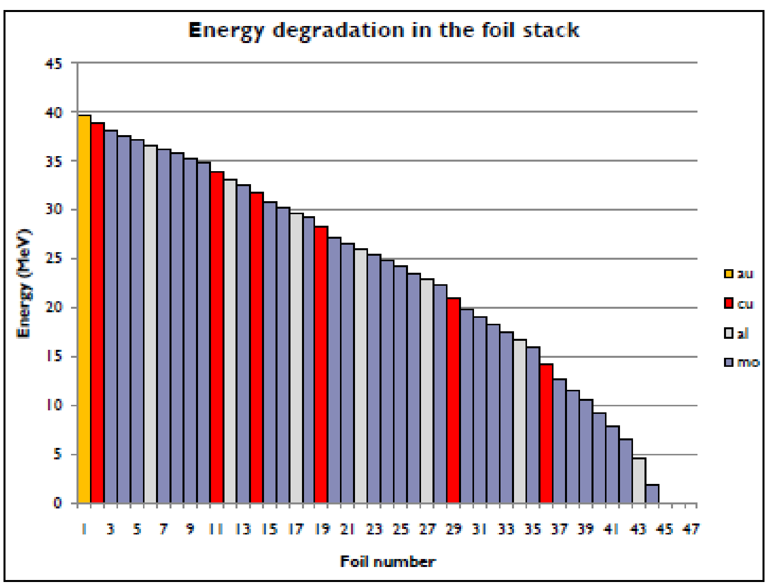

FIG. 1. Energy degradation through the stack and foil composition.

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The beam normalization was done solely with the Cu foils as Al didn't yield sufficient data. Figures 2 and 3 show the excitation functions for the $^{nat}Cu(p,x)^{65}Zn$ and $^{nat}Cu(p,x)^{62}Zn$. The blue dots indicate the recommended values dictated by the International Atomic Energy Agency. Our data showed good agreement for the $^{nat}Cu(p,x)^{65}Zn$ reaction. However, for the $^{nat}Cu(p,x)^{62}Zn$ reaction, our numbers were not very consistent with the recommended data. This turned out to be the main source of uncertainty for our measurements.

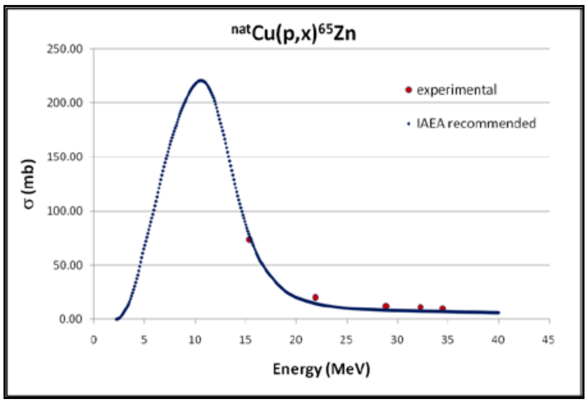

FIG. 2. Comparison between experimental and IAEA recommended cross-section values for $^{nat}Cu(p,x)^{65}Zn$.

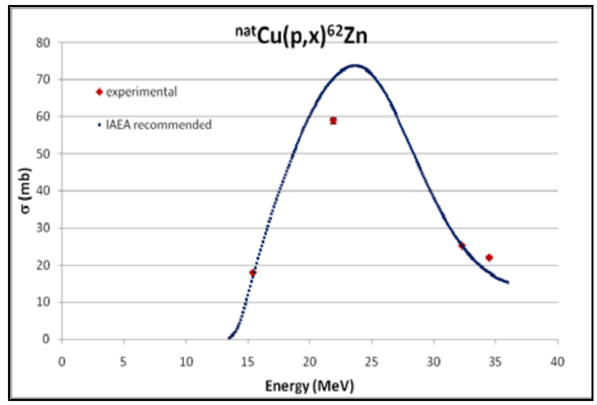

FIG. 3. Comparison between experimental and IAEA recommended cross-section values for $^{nat}Cu(p,x)^{62}Zn$.

The cross-sections for the various nuclear reactions $^{nat}Mo(p,x)$ were determined with the usual activation formula [2] and then compared with available previously published results and with calculations done with the code TALYS [3]. Given the space restrictions, here results will be shown only for the medical radioisotopes ^{99m}Tc and ^{94g}Tc.

A. $^{nat}Mo(p,x)^{99m}Tc$ - The radionuclide ^{99m}Tc is used mainly in single photon emission computed tomography (SPECT). It has a half-life of ~6 h and low energy gamma emission preferable for medical imaging. The production cross-section was determined through the analysis of the 140.5 keV gamma peak. There are two main processes contributing to its production. One is the reaction $^{100}Mo(p,2n)^{99m}Tc$ and the other is $^{100}Mo(p,pn)^{99}Mo \rightarrow {}^{99m}Tc$. Figure 4 shows the results compared to previous published data and simulated values. In the energy range 12 – 22 MeV results are consistent with the calculations but lower than previous data. Above 22 MeV the contributions from ^{99}Mo (140.51 keV) and ^{90}Nb (141.18 keV) couldn't be separated well enough so the cross-section values are higher than the calculation.

B. $^{nat}Mo(p,x)^{94g}Tc$ - The radionuclide ^{94g}Tc has a half-life of ~5 h and a mean positron energy of 358.3 keV. It has been studied for potential applications in positron emission tomography (PET) together with the isomeric state ^{94m}Tc. The latter has a half-life of 52 min, so it couldn't be studied here as it was mostly decayed by the time measurements started. The gamma line used in the analysis was the independent 702.6 keV line. Figure 5 shows the results, again compared with previous research and calculations. There is good agreement within the error bars with previous works. The calculations yielded higher values as TALYS considered the ground and isomeric states together. The red arrows and labels indicate the contributing production reactions.

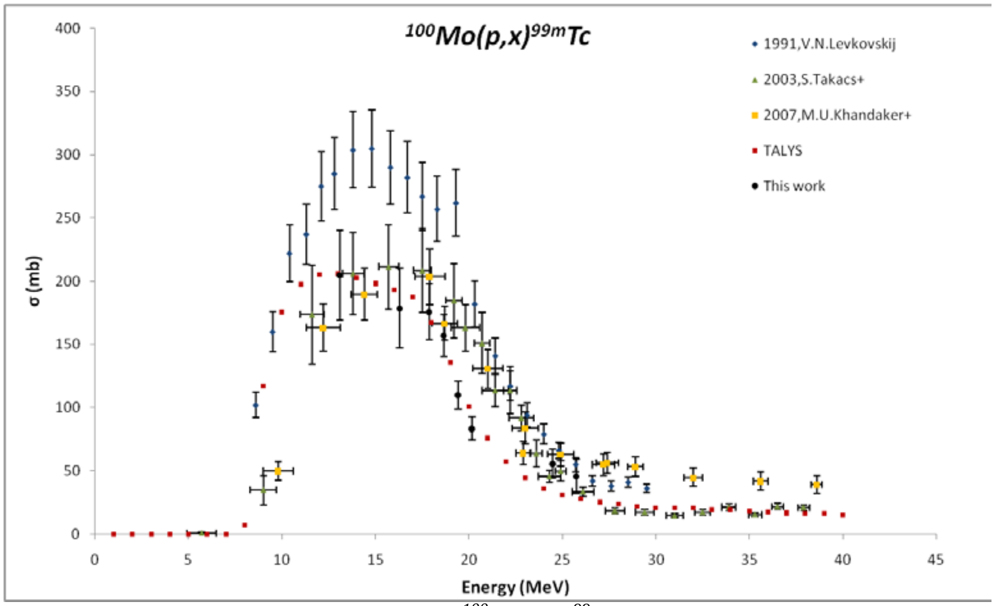

FIG. 4. Excitation function of the reaction $^{100}Mo(p,x)^{99m}Tc$.

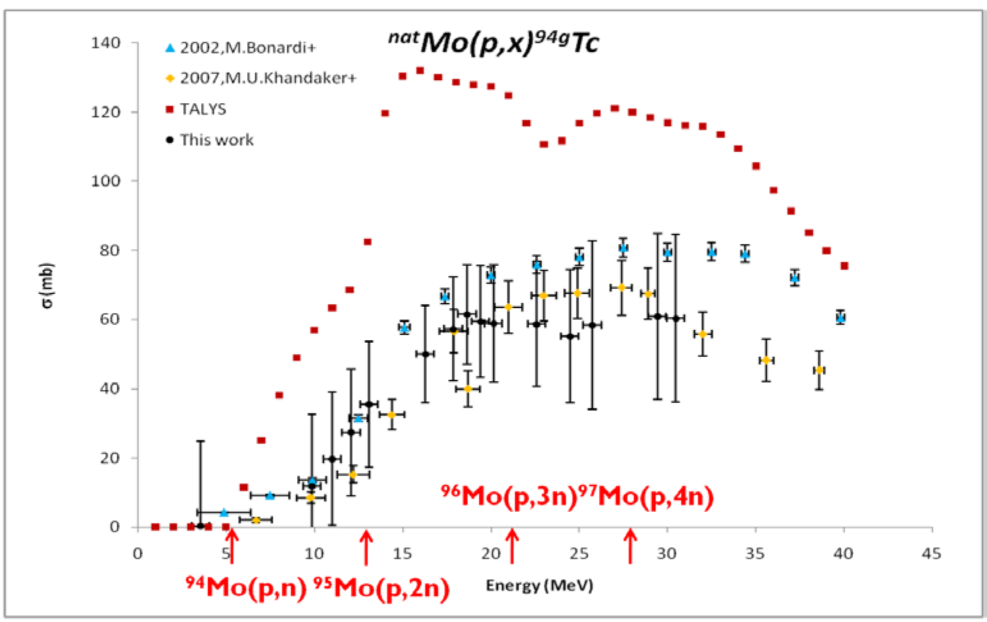

FIG. 5. Excitation function of the reaction $^{nat}Mo(p,x)^{94g}Tc$.

To summarize, Mo foils were irradiated with protons to test the production of medical radioisotope ^{99m}Tc using a cyclotron beam as an alternative method to the currently used ^{99}Mo/^{99m}Tc generator. The measured excitation function for ^{99m}Tc was in relatively good agreement with model calculations. However, there are significant discrepancies among available works including this one, which indicates the need for more data before this production method can be considered viable.

The ^{96}Tc and ^{95m}Tc excitation functions, not presented here, were measured to test the reliability of the data. They were found slightly higher than the calculations but were in good agreement with previous works. Both are being used in radiochemical and animal studies. The radioisotope ^{94g}Tc was studied for its potential application in PET studies and was found in good agreement with previous works.

The main source of uncertainty in the experiment came from beam normalization, ~20%. Other sources included statistical uncertainties of up to ~9% and uncertainties from gamma line intensity (2%), detector efficiency (1-4%) and foil thickness determination (2%).

As a test experiment, it proved that the used set-up was working and it revealed improvements necessary to be made for the following experiments in order to minimize uncertainties and increase the reliability of the data.

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