Nuclear Data for Charged-particle Monitor Reactions and Medical Isotope Production
1st RCM – 3-7/12/12

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Proposed activities:

1) Evaluation of the monitor reactions $^{27}Al(p,x)^{24}Na$ and $^{27}Al(p,x)^{22}Na$ up to 200 MeV;

2) Evaluation of γ, n, p + $^{100}Mo \rightarrow {}^{99}Mo$, ^{99m}Tc;

3) Evaluation of p + ^{232}Th production of alpha emitters;

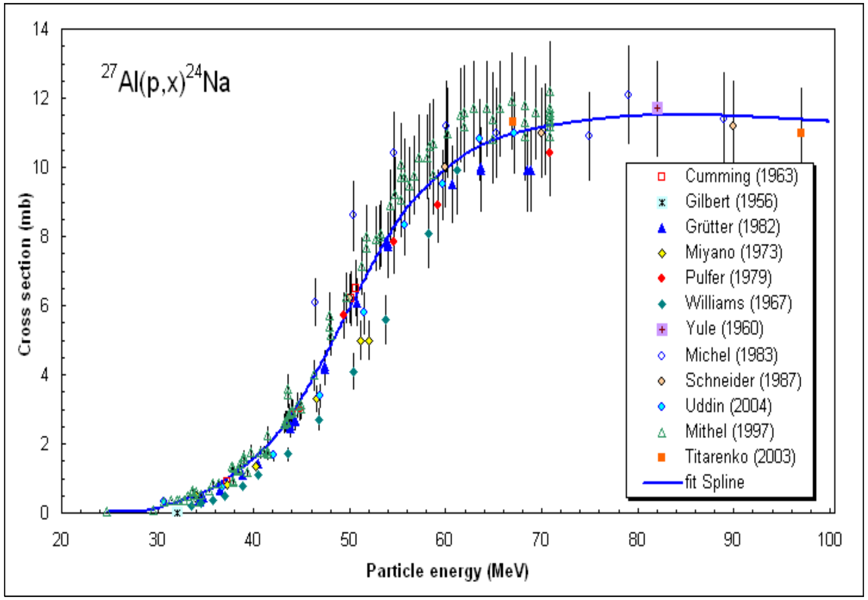
4) Development and testing of the DDHMS module of the EMPIRE-3.1 system in photon and nucleon-induced reactions, including both double differential and residual production cross sections.

– > All our activities are theoretical. They involve both model development and data evaluation.

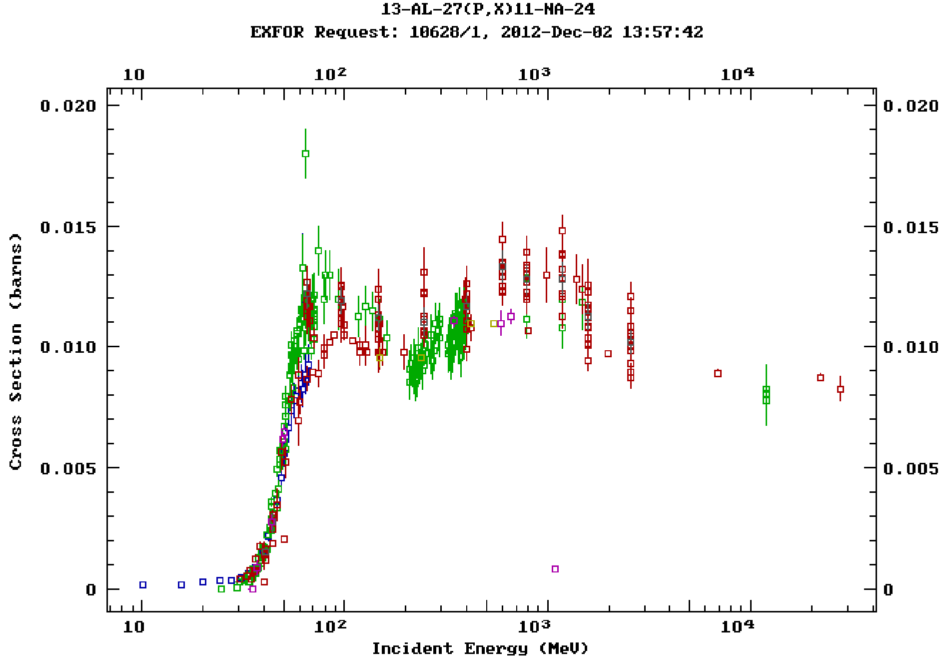
Evaluation of the monitor reaction $^{27}Al(p,x)^{24}Na$ (β-, 15 h) up to 200 MeV;

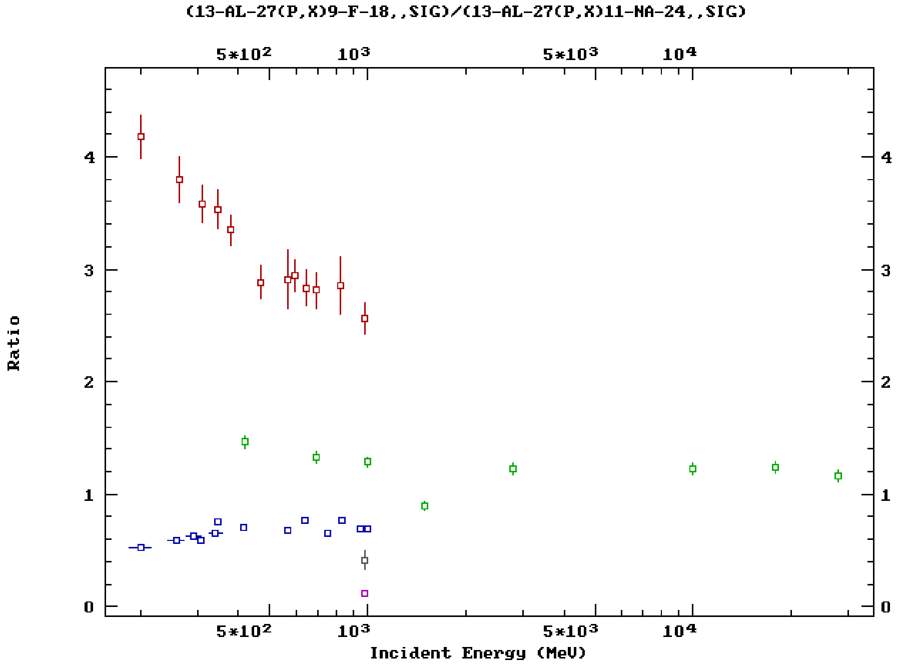
Here, x = n + 3p, d + 2p.

IAEA recommended monitor cross section (cubic spline) --->



EXFOR (p,x) data and cross section ratio data

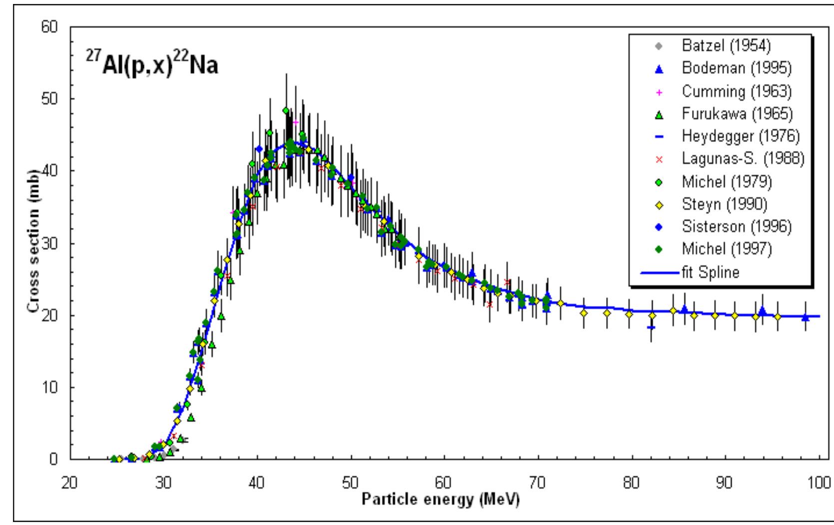




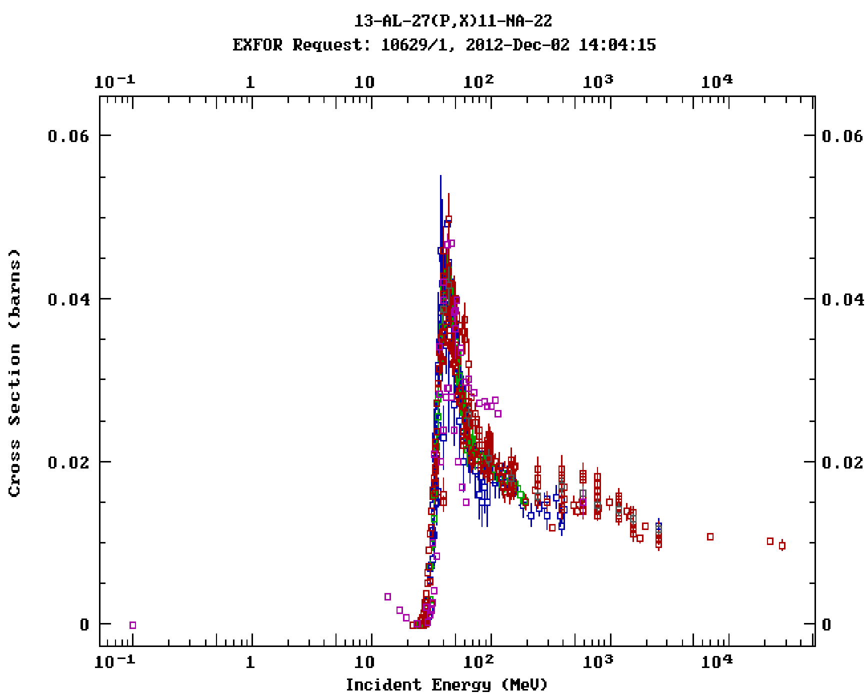
Evaluation of the monitor reaction ^{27}Al(p,x)^{22}Na (β+, 2.6 y)   up to 200 MeV;

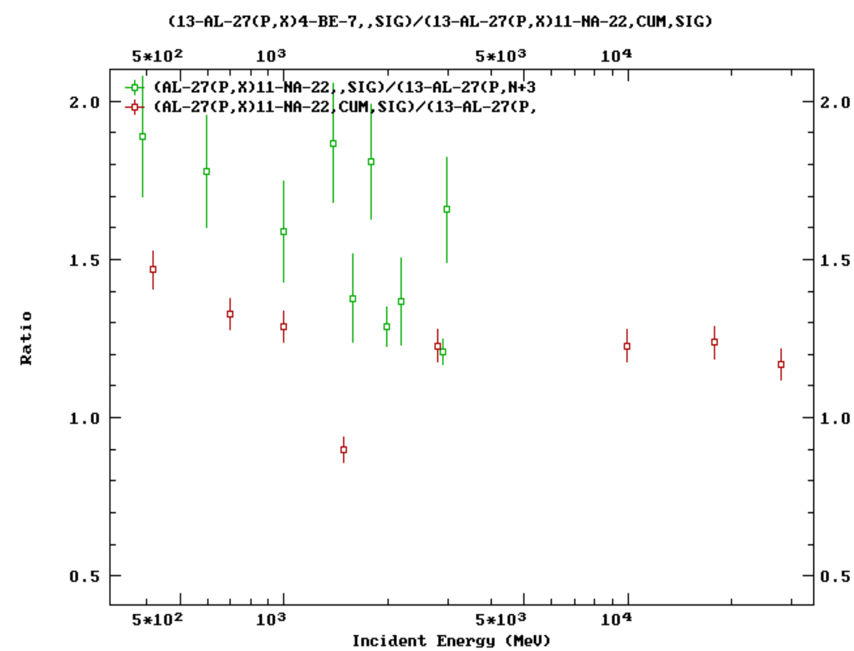
Here, x = 3n + 3p, d + 2n + 2p, 2d + n + p, 3d, α + n + p, α + d, ^{6}Li.

IAEA recommended
monitor cross section   --->
(cubic spline)

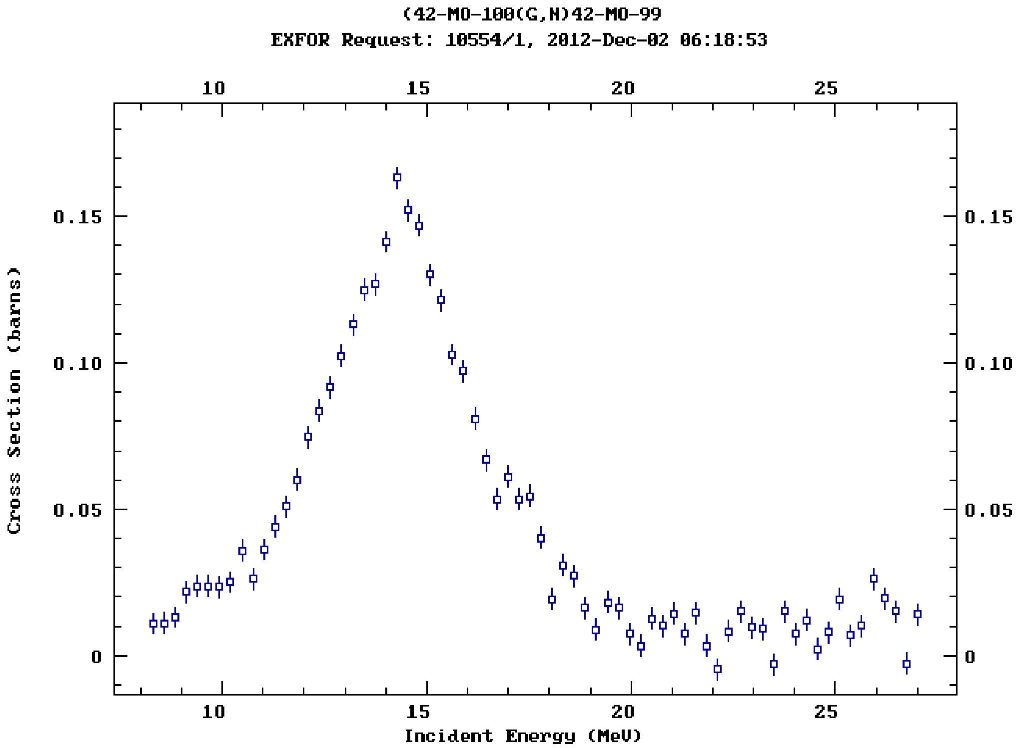


EXFOR (p,x) data and
cross section ratio data





Evaluation of $\gamma + {}^{100}Mo \rightarrow {}^{99}Mo$

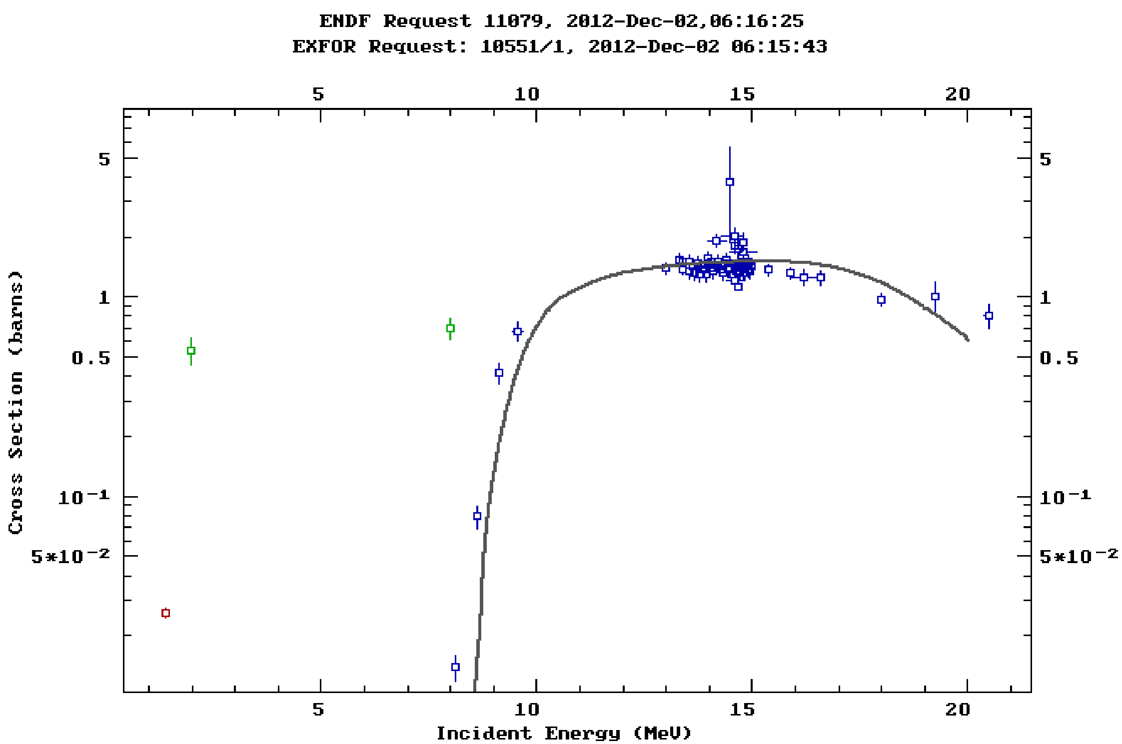


The cross sections are small → for the moment, this γ-induced reaction would seem to be more interesting for testing models than innovating technology.

Evaluation of n + $^{100}Mo \rightarrow {}^{99}Mo$

Neutron-induced fission of 235U has been the main source of ^{99}Mo up until now.

An alternative neutron-induced production route is given by the (n,2n) reaction.
The data is shown here with the ENDFB-VII.1 evaluation.

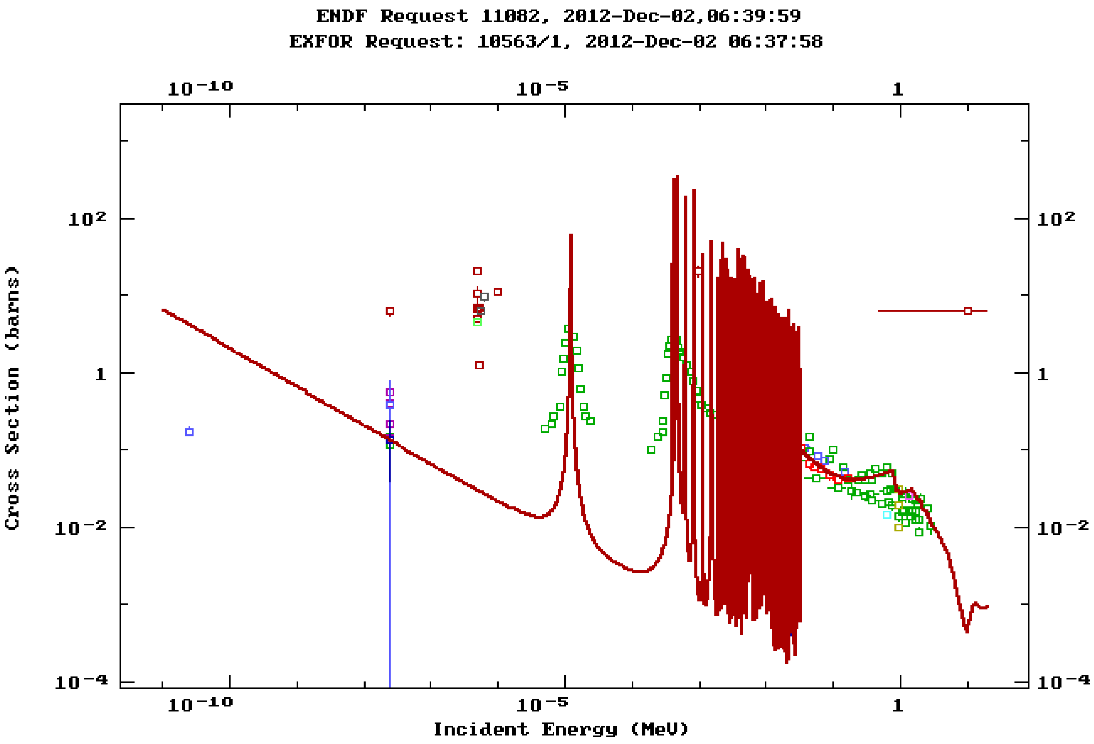


However, the energy range for activation is above that of a thermal reactor, greatly limiting any possible application of the process.

Evaluation of n + ^{98}Mo → ^{99}Mo

An alternative might be neutron-induced capture, which has the (slight) advantage that ^{98}Mo is the most abundant isotope.

Here the available data for the cross section are shown together with the ENDFB-VII.1 evaluation.



The cross section at thermal energy is about 100 mb and thus would appear to be too small to be of technological interest.

Evaluation of p + $^{100}Mo \rightarrow ^{99}Mo$, ^{99m}Tc

Direct production of ^{99}Mo and, in particular, of ^{99m}Tc have been shown to provide viable alternatives to reactor production of ^{99}Mo. These routes were not investigated seriously until recently, when shortages of reactor produced ^{99}Mo occurred. A list of references:

1) Beaver, J.E.; Hupf, H.B., "Production of ^{99m}Tc on a Medical Cyclotron: a Feasibility Study". Journal of Nuclear Medicine 12 (1971) 739.

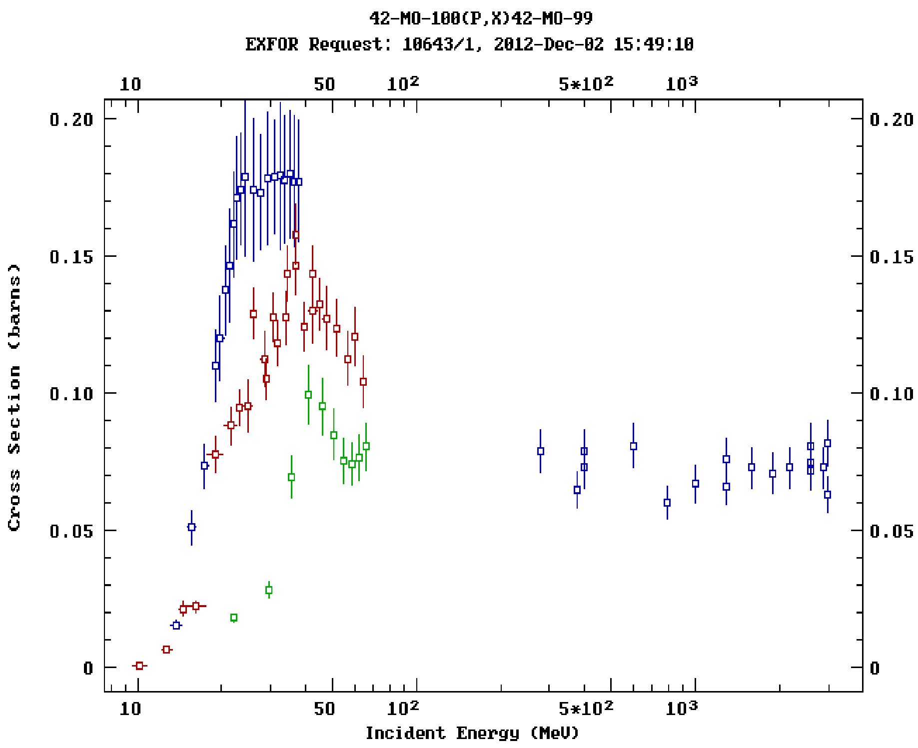
2) Scholten, Bernhard; Lambrecht, Richard M.; Cogneau, Michel; Vera Ruiz,, Hernan; Qaim, Syed M.; "Excitation functions for the cyclotron production of ^{99m}Tc and ^{99}Mo". Applied Radiation and Isotopes 51 (1999) 69.

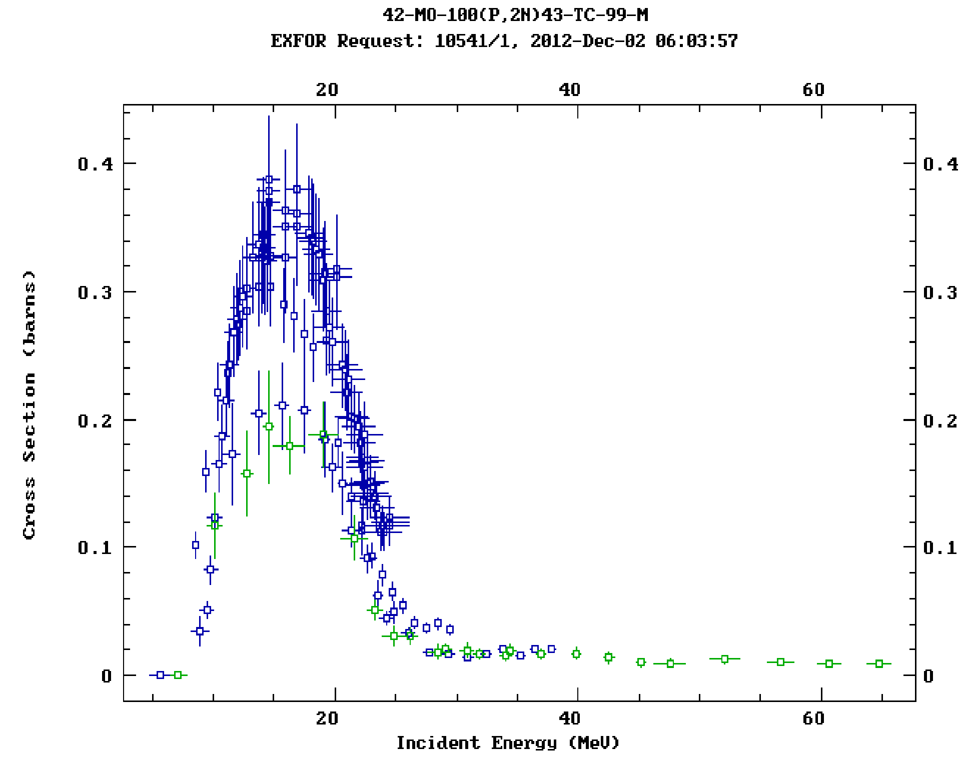
3) Takács, S.; Szűcs, Z.; Tárkányi, F.; Hermanne, A.; Sonck, M. ; "Evaluation of proton induced reactions on ^{100}Mo: New cross sections for production of ^{99m}Tc and ^{99}Mo". Journal of Radioanalytical and Nuclear Chemistry 257 (2003) 195.

4) Guérin, B.; Tremblay, S.; Rodrigue, S.; Rousseau, J.A.; Dumulon-Perreault, V.; Lecomte, R.; van Lier, J.E.; Zyuzin, A. et al. (April 2010). "Cyclotron production of ^{99m}Tc: an approach to the medical isotope crisis". Journal of Nuclear Medicine 51(2010) 13N.

5) Celler, A.; Hou, X.; Bénard, F.; Ruth, T.; "Theoretical modeling of yields for proton-induced reactions on natural and enriched molybdenum targets". Physics in Medicine and Biology 56 (2011) 5469.

Evaluation of p + ^{100}Mo → ^{99}Mo, ^{99m}Tc



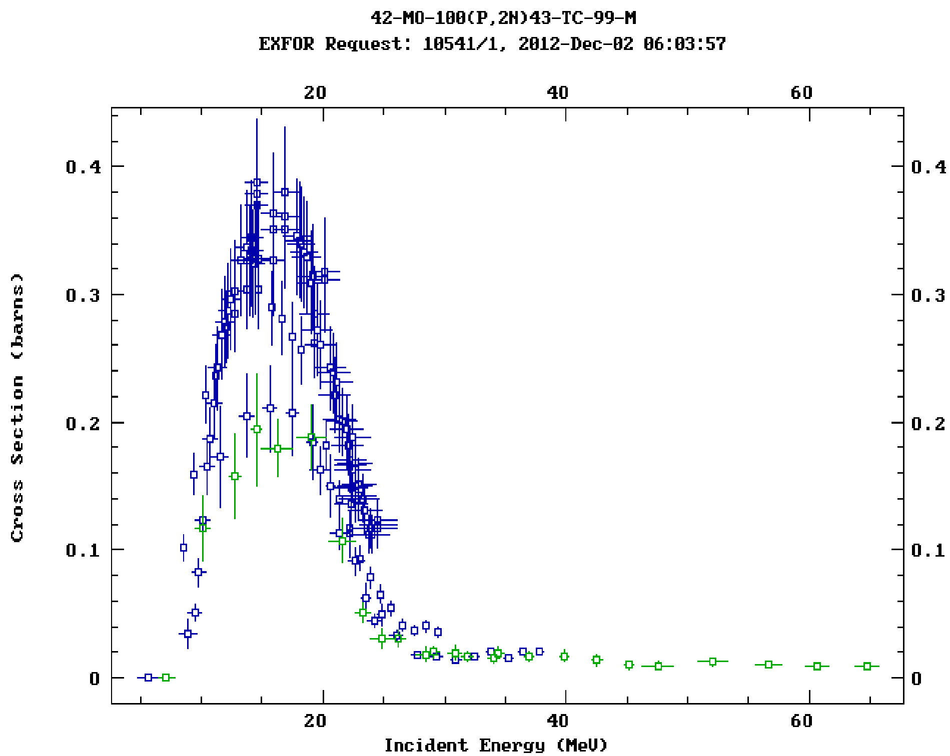


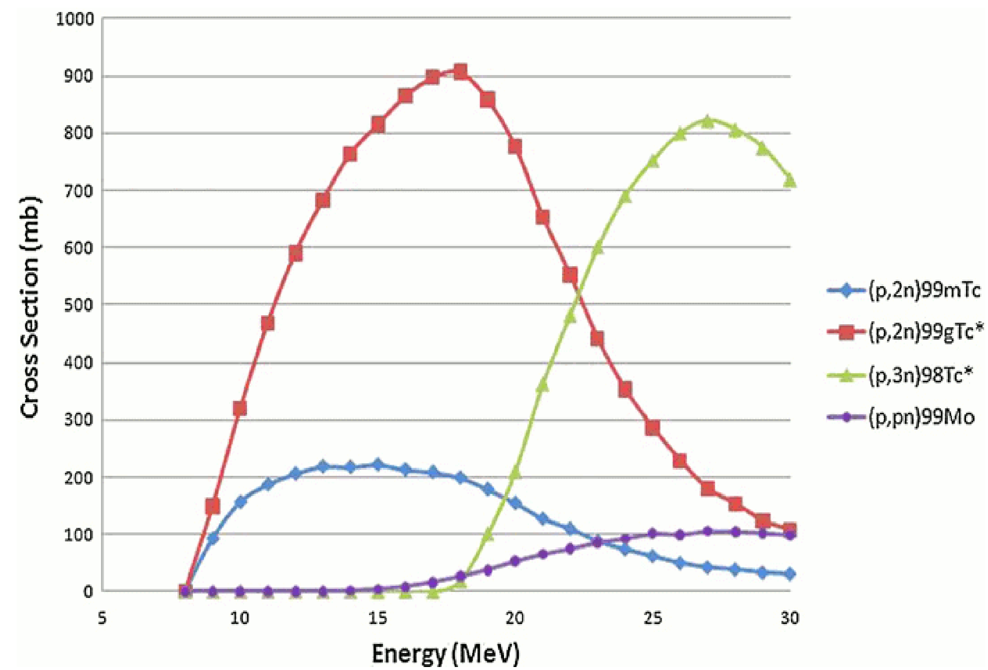
The (p,2n) cross section for production of ^{99m}Tc is larger than than the (p, np+d) cross section for production of ^{99}Mo. This direct production path seems to be the most indicated.

The cross sections in both cases show large differences, which due the the similar forms, might involve questions of normalization. An evaluation is needed.

Evaluation of p + ^{100}Mo → ^{99}Mo, ^{99m}Tc

The experimental data on the left can be compared with the calculation performed by Celler et al. using EMPIRE-3.1.



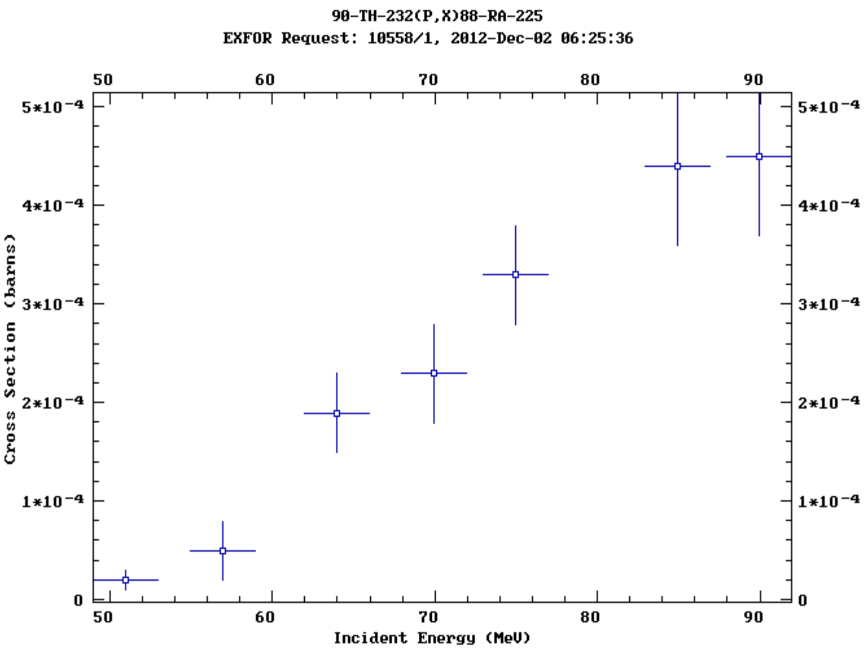


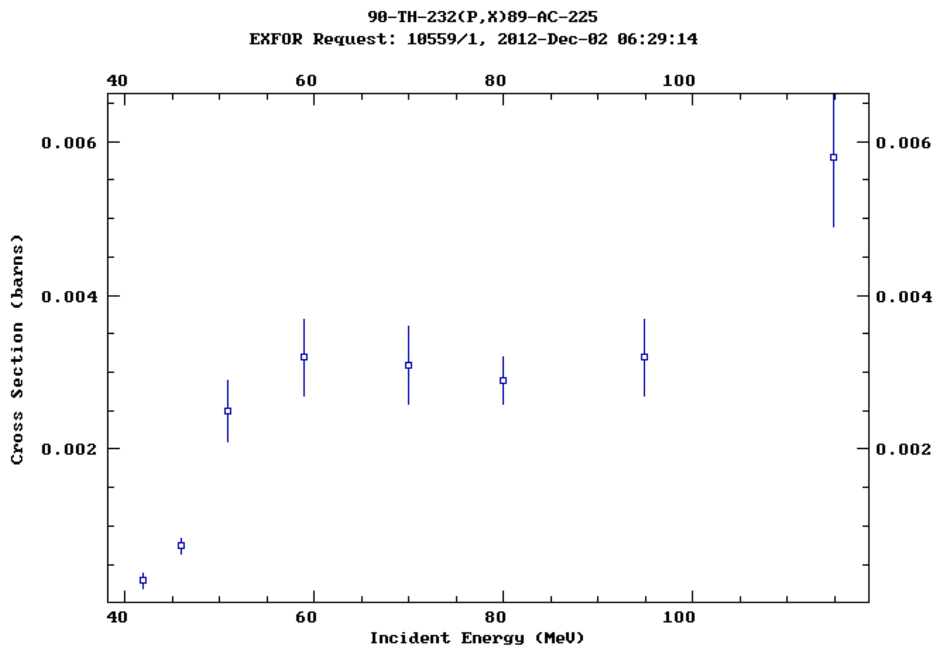
Celler et al. conclude that the optimal window for ^{99m}Tc production is between 16 and 19 MeV, where the production cross section is high (taking into account production of ^{100}Mo as well) but contaminant production is low.

Data evaluation will probably not change this conclusion but might change the overall production rate.

Evaluation of p + ^{232}Th production of alpha emitters

The isotopes ^{225}Ra and ^{225}Ac are produced by multiple emissions in proton induced reactions on ^{232}Th.





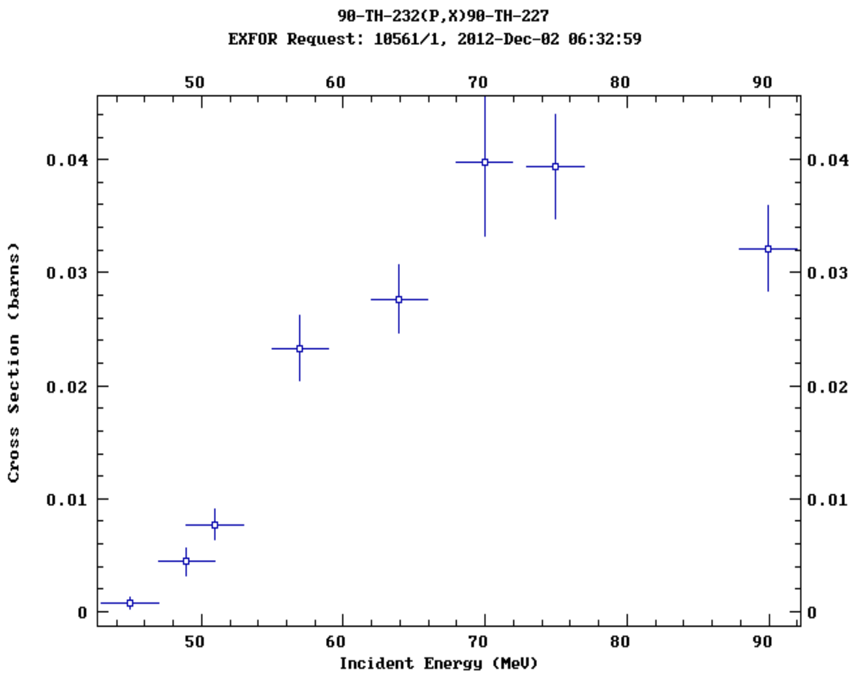
The simplest reaction producing ^{225}Ra would be ^{232}Th(p, α + 3n + p)^{225}Ra.

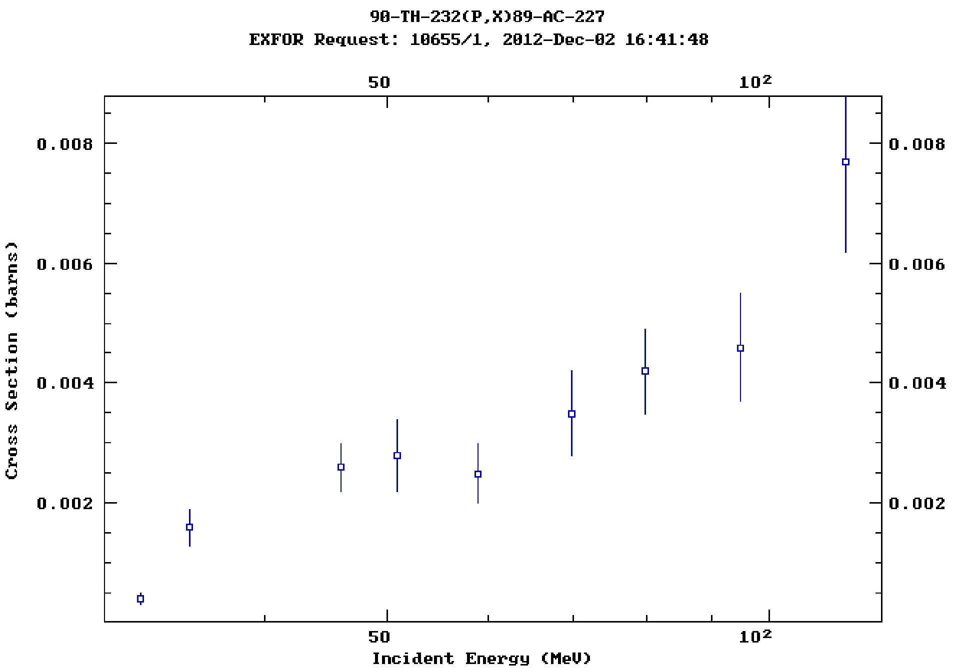
The simplest reaction producing ^{225}Ac would be ^{232}Th(p, α +4n)^{225}Ac, consistent with its larger cross section.

Evaluation of p + ^{232}Th production of alpha emitters

Other alpha emitters with relatively complicated production paths are

^{232}Th(p, p + 5n)^{227}Th and ^{232}Th(p, α + 2n)^{227}Ac (β-) $\rightarrow$ ^{227}Th (98.6%)





Other more complicated processes, (p, 4n + 2p), for example, also contribute to the production of ^{227}Ac.

Development and testing of the DDHMS module of the EMPIRE-3.1 system in photon and nucleon-induced reactions, including both double differential and residual production cross sections.

In some of the cases above, the experimental data available for the channels of interest is probably sufficient to permit a cubic spline fit.

A complete evaluation would require model calculations. The challenges in the cases discussed are:

1) the large number of open channels and the wide variety of particles that can be emitted;

For the $^{27}Al(p,x)^{22}Na$ reaction, we have seen that

$x = 3n + 3p,\ d + 2n + 2p,\ 2d + n + p,\ 3d,\ \alpha + n + p,\ \alpha + d,\ ^{6}Li$

For $^{232}Th(p,x)^{225}Ac$, it is

$x = 6n + 2p,\ d + 5n + p,\ 2d + 4n,\ \alpha + 4n,\ ^{6}He + 2n,\ ^{8}He$

2) the large range in energy and angular momentum of residual occupations.

At 200 MeV, the angular momentum of proton incident on ^{27}Al can reach 55*hbar.

For a proton incident on ^{232}Th, the maximum angular momentum (for a nuclear reaction) will be about 110*hbar.

The range of excitation energies involved is also extremely broad.

EMPIRE-3.1

- Spherical Optical Model
- Coupled Channels
- Distorted Wave Born Approximation
- Simplified CC for HI fusion
- Multi-step Direct reactions (up to two steps)
- Multi-step Compound with gamma emission
- Exciton Model with cluster emission in terms of Iwamoto-Harada model
- *Monte Carlo simulation of multiple pre-equilibrium emission - DDHMS*
- Widths fluctuations
- Hauser-Feshbach model with full gamma-cascade and dynamical deformation effects
- Multi-modal fission through multi-humped barrier in terms of optical model for fission

Double Differential Hybrid Monte Carlo Simulation - Refs:

- M. Blann, Phys. Rev. C 54 (1996) 1341.

- M. Blann and M. B. Chadwick, Phys. Rev. C 57 (1998) 233.

Double Differential Hybrid Monte Carlo Simulation

We have
1) refined the Monte Carlo sampling to
   a) reproduce the exact distributions of scattering from a Fermi sea;
   b) furnish exclusive cross sections;
and
2) integrated the results of the pre-equilibrium cascade in EMPIRE.

The DDHMS furnishes a semiclassical approximation to multistep direct processes, in which many particles can be in the continuum simultaneously.

Although approximate, it extends the range of application of EMPIRE up to about 200 to 250 MeV, when other implicit conditions, such as the existence of discrete levels and/or mass defects or angular momentum limitations, permit.

This is still work in progress.