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^{99m}Tc DIRECT PRODUCTION USING PROTON BEAM FROM C18 CYCLOTRON

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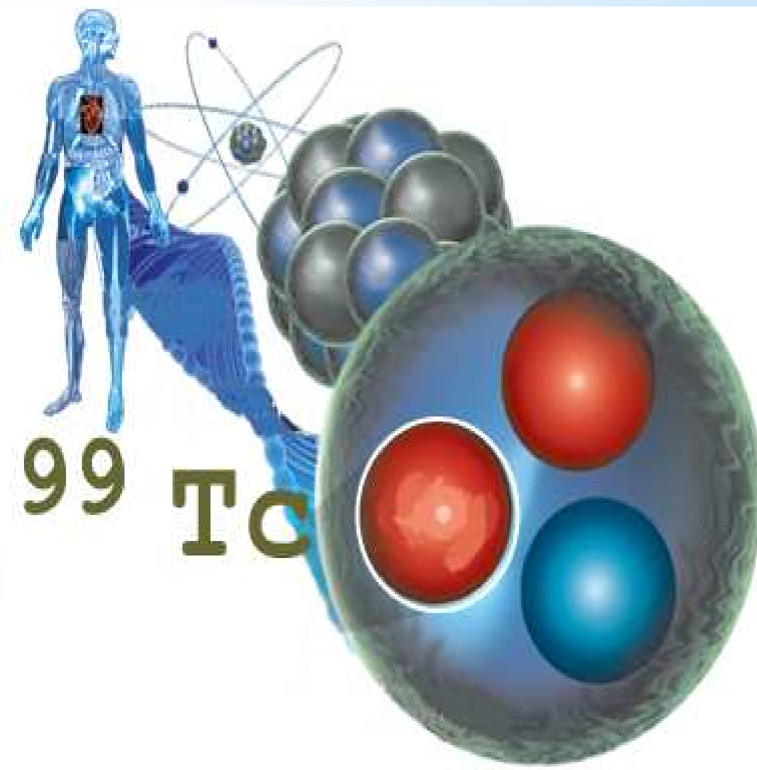


Introduction

- The present world demand for ^{99}Mo is about 450000 GBq/week, and the annual demand for ^{99}Mo is considered to have an 8 – 12% growth over the next decade. Currently, most ^{99}Mo is produced by using five nuclear research reactors in Canada, Belgium, France, Netherlands, and South Africa.

The situation is hazardous: first, routine shipments of ^{99}Mo could be stopped for any reasons, such as planned maintenance of or an unscheduled shutdown of a reactor, or due to any problems related to the transportation of ^{99}Mo, etc

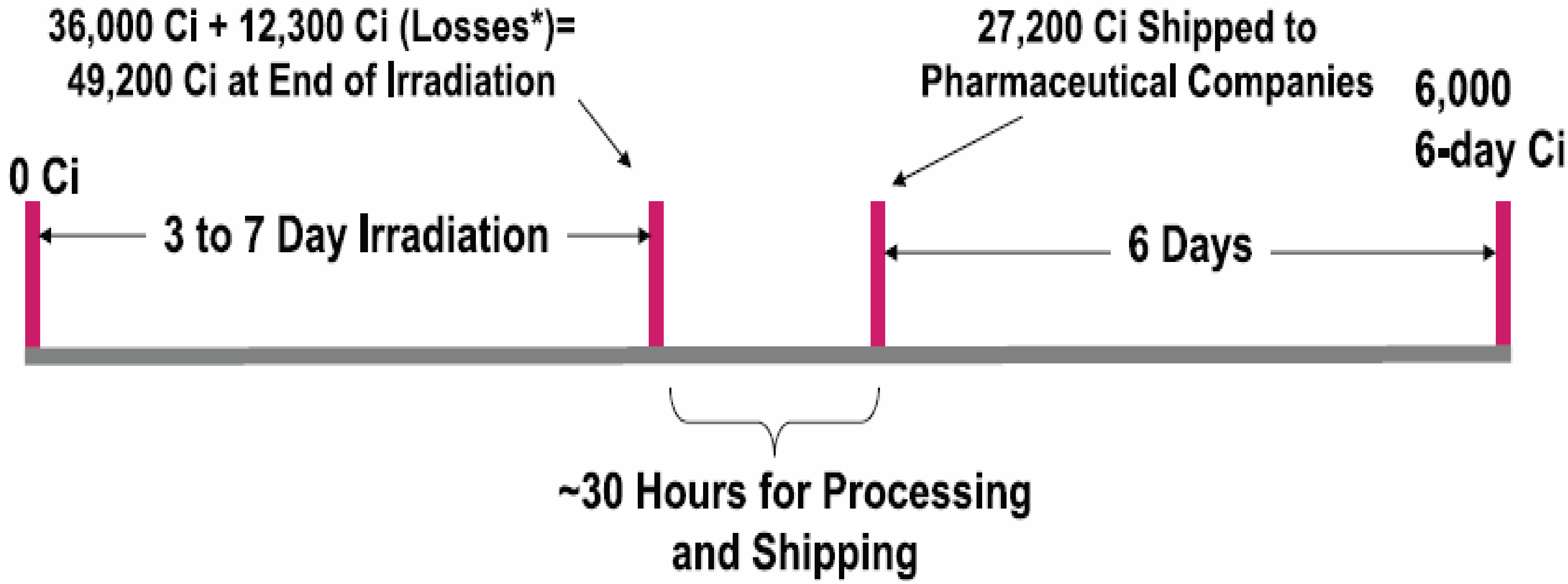
Second, these reactors use highly enriched ^{235}U (HEU), which is a direct use material for nuclear weapons.

In 2007 about 50 kg of HEU was used by the reactors mentioned above, and the quantity is considered to be sufficient for the construction of the two nuclear bombs.

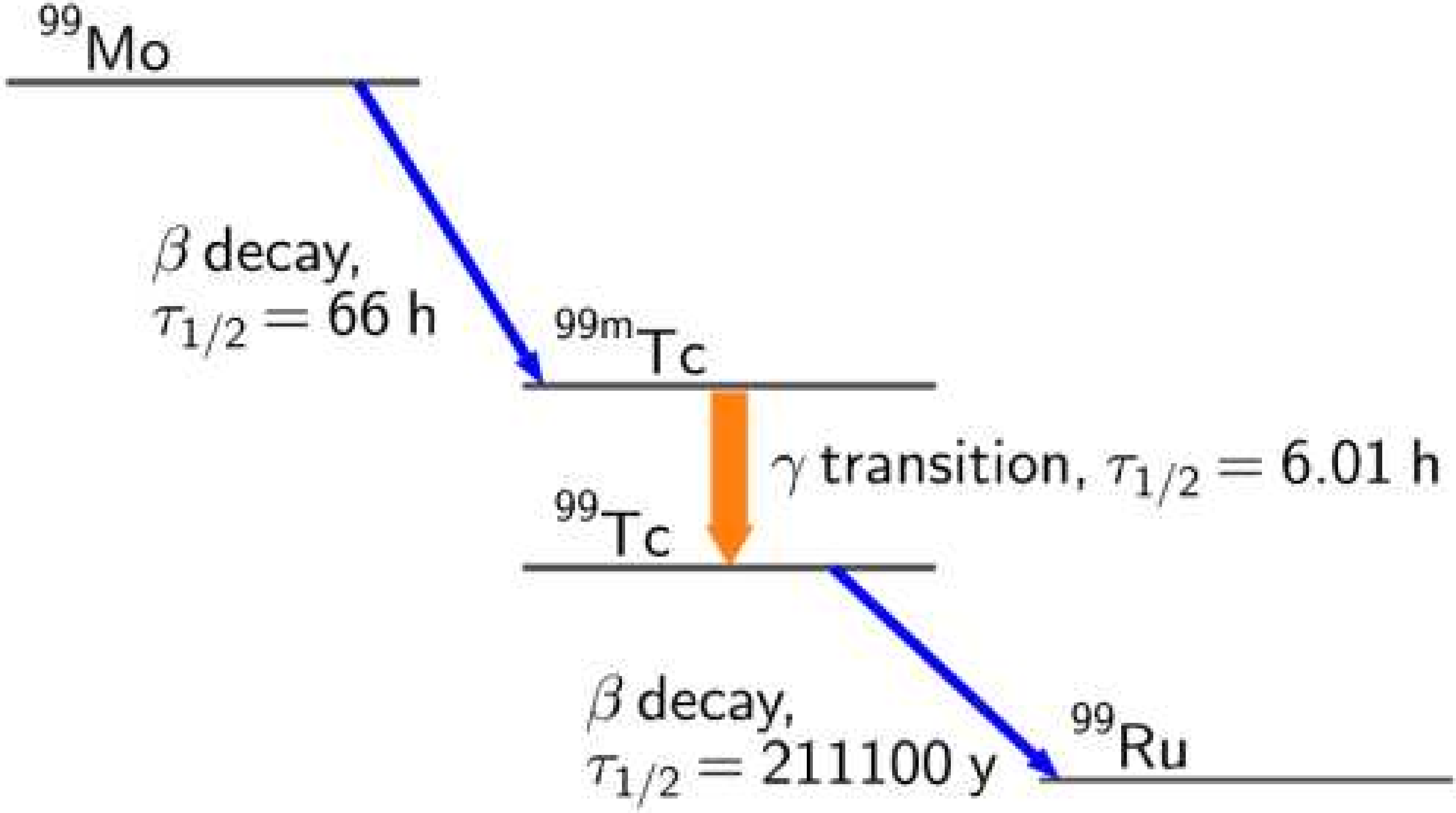
In fact, about a five week unscheduled shutdown of a reactor in Canada, which happened in 2007, reinforced concerns about a reliable long-term supply of ^{99}Mo.

Note that the reactors mentioned above range in age from 42 and 51 year, and it is considered to be quite difficult nowadays to get approval to build a new reactor.

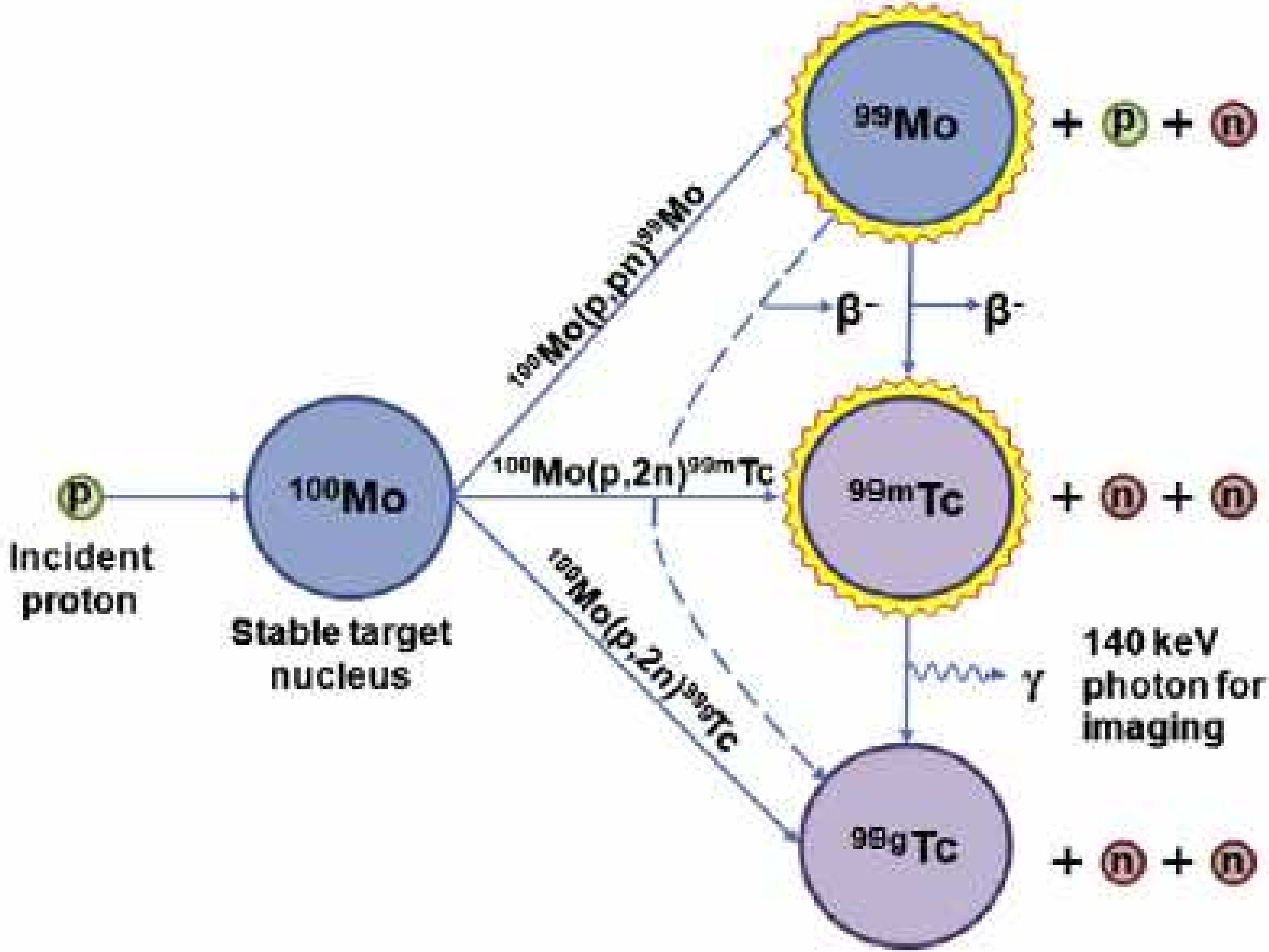
Estimated 2009 U.S. Mo-99 Demand
at least 6,000 6-day Curies/Week



*Assumes 75% Mo-99 Recovery During Processing


^{99}Mo
β decay,
$\tau_{1/2} = 66$ h
^{99m}Tc
γ transition, $\tau_{1/2} = 6.01$ h
^{99}Tc
β decay,
$\tau_{1/2} = 211100$ y
^{99}Ru

REPORT on the 1st Research Coordination Meeting on “Accelerator-based Alternatives to Non-HEU Production of $^{99}Mo/^{99m}Tc$”, 16 - 20 April 2012, Vancouver, Canada



Incident proton
100Mo
Stable target nucleus
100Mo(p,pn)99Mo
100Mo(p,2n)99mTc
100Mo(p,2n)99gTc
99Mo
+ p + n
β-
β-
99mTc
+ n + n
γ
140 keV photon for imaging
99gTc
+ n + n

Participant	Production	Target	Chemistry	Labelling	Dosimetry	Recycling
Brazil		X	X	X	X	X
Canada TRIUMF	X	X	X	X	X	X
Canada U. of Alberta	X	X			X	X
Hungary	X		X		X	
India		X	X	X	X	X
Japan		X	X	X		X
Kingdom of Saudi Arabia		X	X	X	X	X
Republic of Korea	X	X	X	X		X
Italy INFN	X	X	X	X	X	X
Italy U. of Pavia			X	X	X	X
Malaysia		X	X	X		X
Poland	X	X	X	X	X	X
Syria		X	X	X		X
Turkey		X		X		X
USA, WU	X	X	X	X	X	X

During the proton irradiation of ^{nat}Mo (of which secondary neutrons are also considered), ^{99m}Tc may arise from the following reactions:

(i) $^{100}Mo(p,2n)^{99m}Tc$,

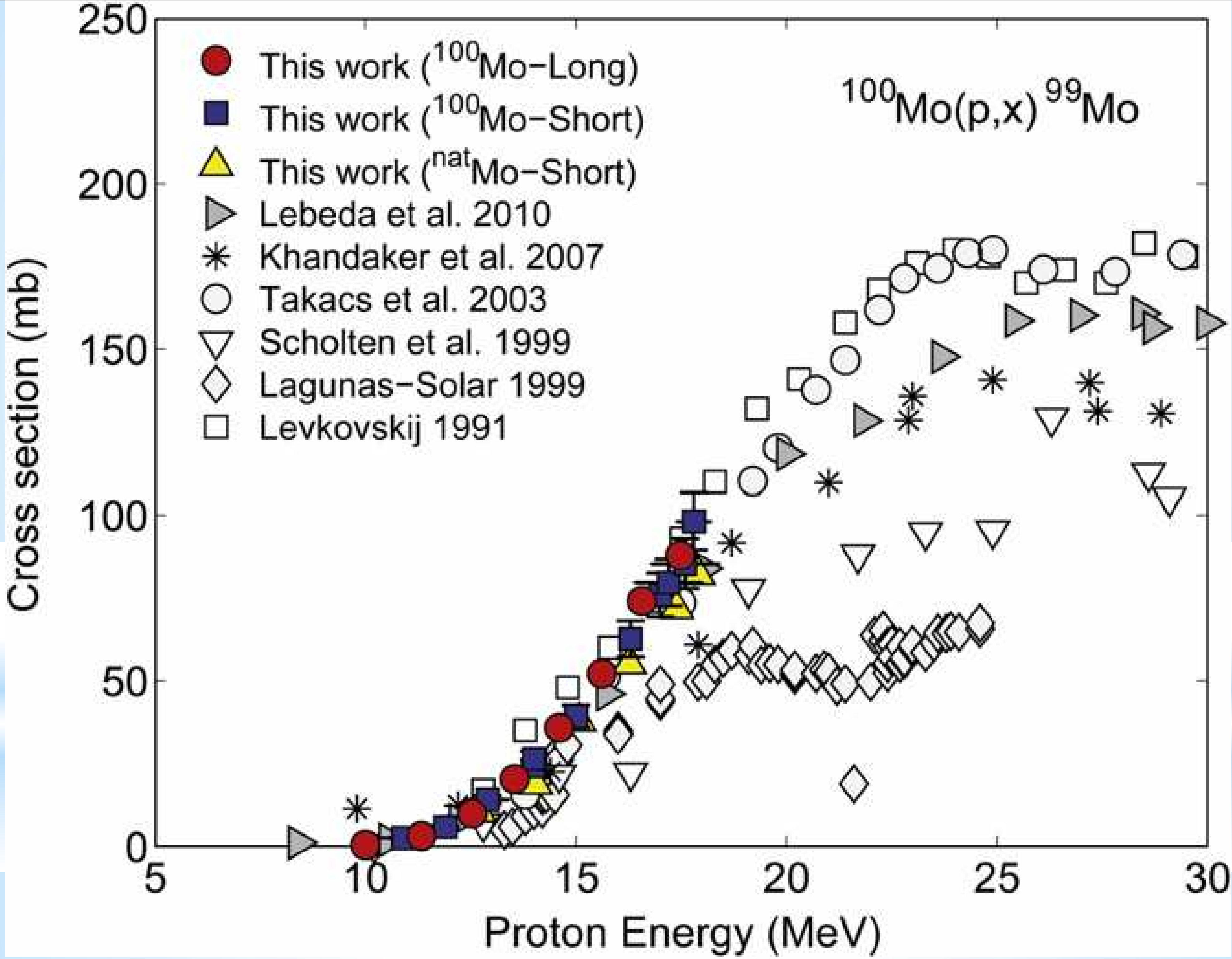
(ii) $^{100}Mo(p,pn)^{99}Mo \rightarrow ^{99m}Tc$,

(iii) $^{100}Mo(n,2n)^{99}Mo \rightarrow ^{99m}Tc$,

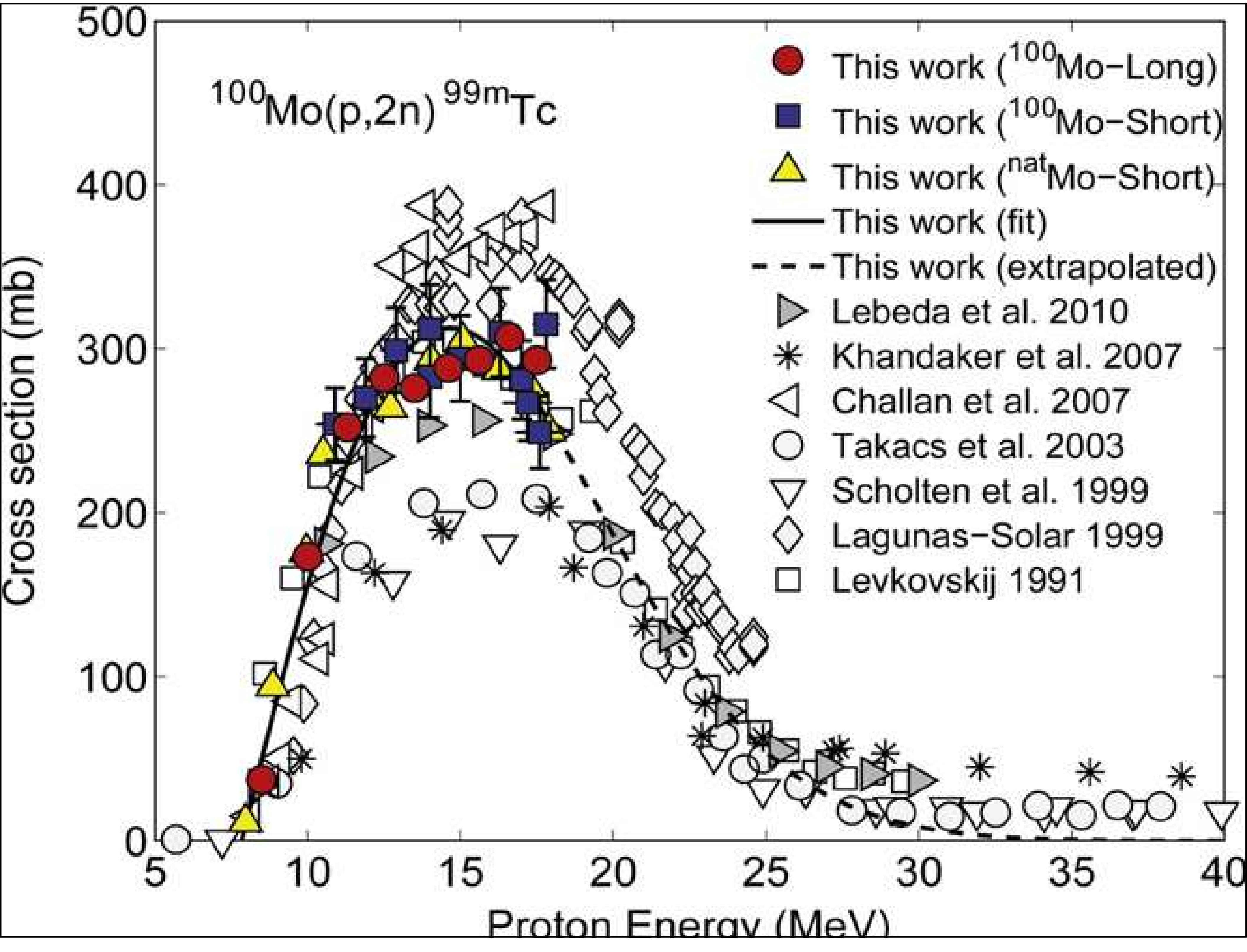
(iv) $^{100}Mo(p,2p)^{99}Nb \rightarrow ^{99}Mo \rightarrow ^{99m}Tc$,

(v) $^{98}Mo(n,\gamma)^{99}Mo \rightarrow ^{99m}Tc$ and

(vi) $^{98}Mo(p,\gamma)^{99m}Tc$.

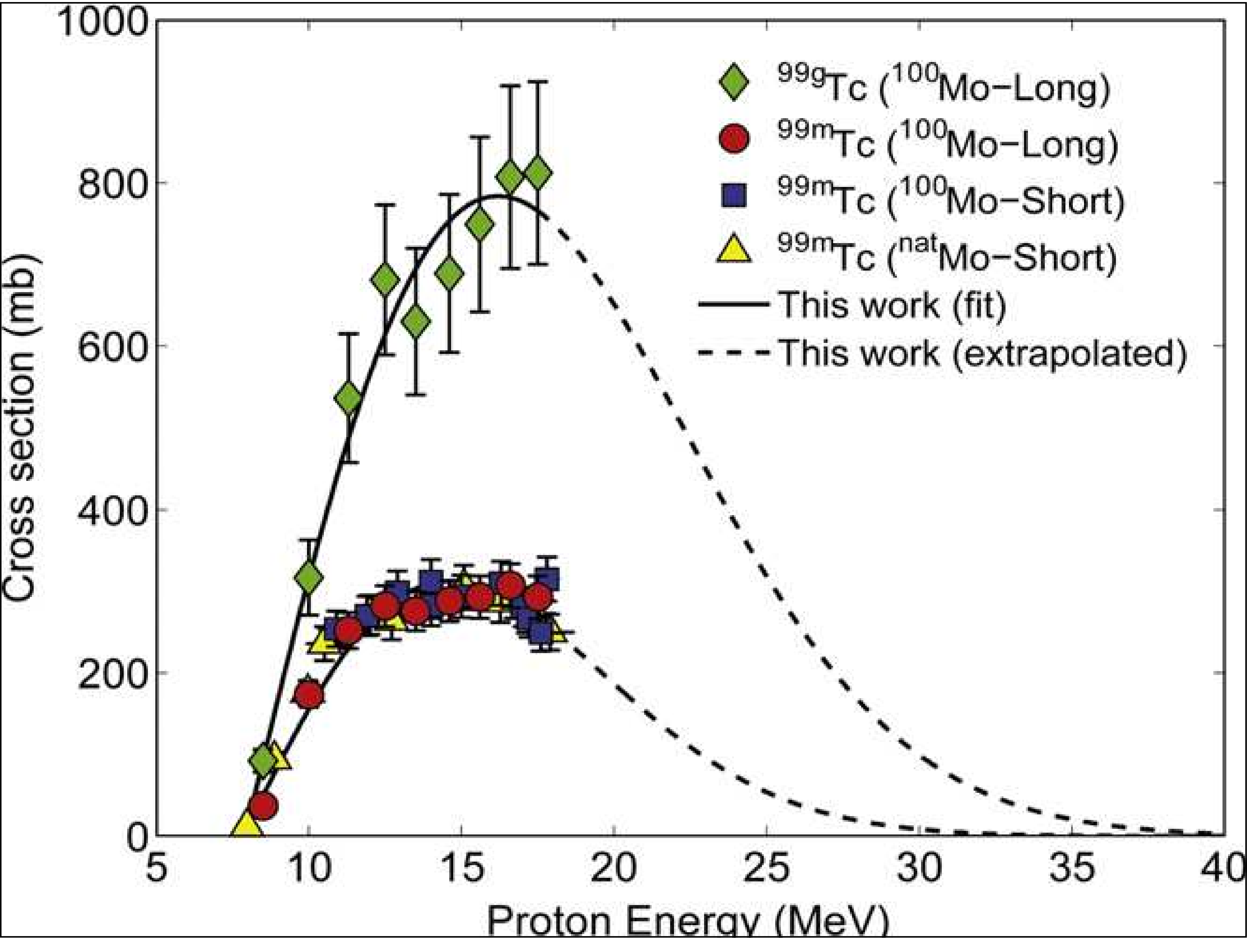


This work (^{100}Mo−Long)
This work (^{100}Mo−Short)
This work (^{nat}Mo−Short)
Lebeda et al. 2010
Khandaker et al. 2007
Takacs et al. 2003
Scholten et al. 1999
Lagunas−Solar 1999
Levkovskij 1991
^{100}Mo(p,x) ^{99}Mo
Cross section (mb)
Proton Energy (MeV)
250
200
150
100
50
0
5
10
15
20
25
30



$^{100}Mo(p,2n)^{99m}Tc$
This work (^{100}Mo−Long)
This work (^{100}Mo−Short)
This work (^{nat}Mo−Short)
This work (fit)
This work (extrapolated)
Lebeda et al. 2010
Khandaker et al. 2007
Challan et al. 2007
Takacs et al. 2003
Scholten et al. 1999
Lagunas−Solar 1999
Levkovskij 1991
Cross section (mb)
Proton Energy (MeV)
0
100
200
300
400
500
5
10
15
20
25
30
35
40

The same time during such a irradiation a so called ^{99g}Tc also is producing. ^{99g}Tc is stable isotope of Tc with T1/2~20000 years and is absolutely unusable for medicine so that its contamination in the final product should be minimized.



^{99g}Tc (^{100}Mo−Long)
^{99m}Tc (^{100}Mo−Long)
^{99m}Tc (^{100}Mo−Short)
^{99m}Tc (^{nat}Mo−Short)
This work (fit)
This work (extrapolated)
Cross section (mb)
Proton Energy (MeV)
0
200
400
600
800
1000
5
10
15
20
25
30
35
40

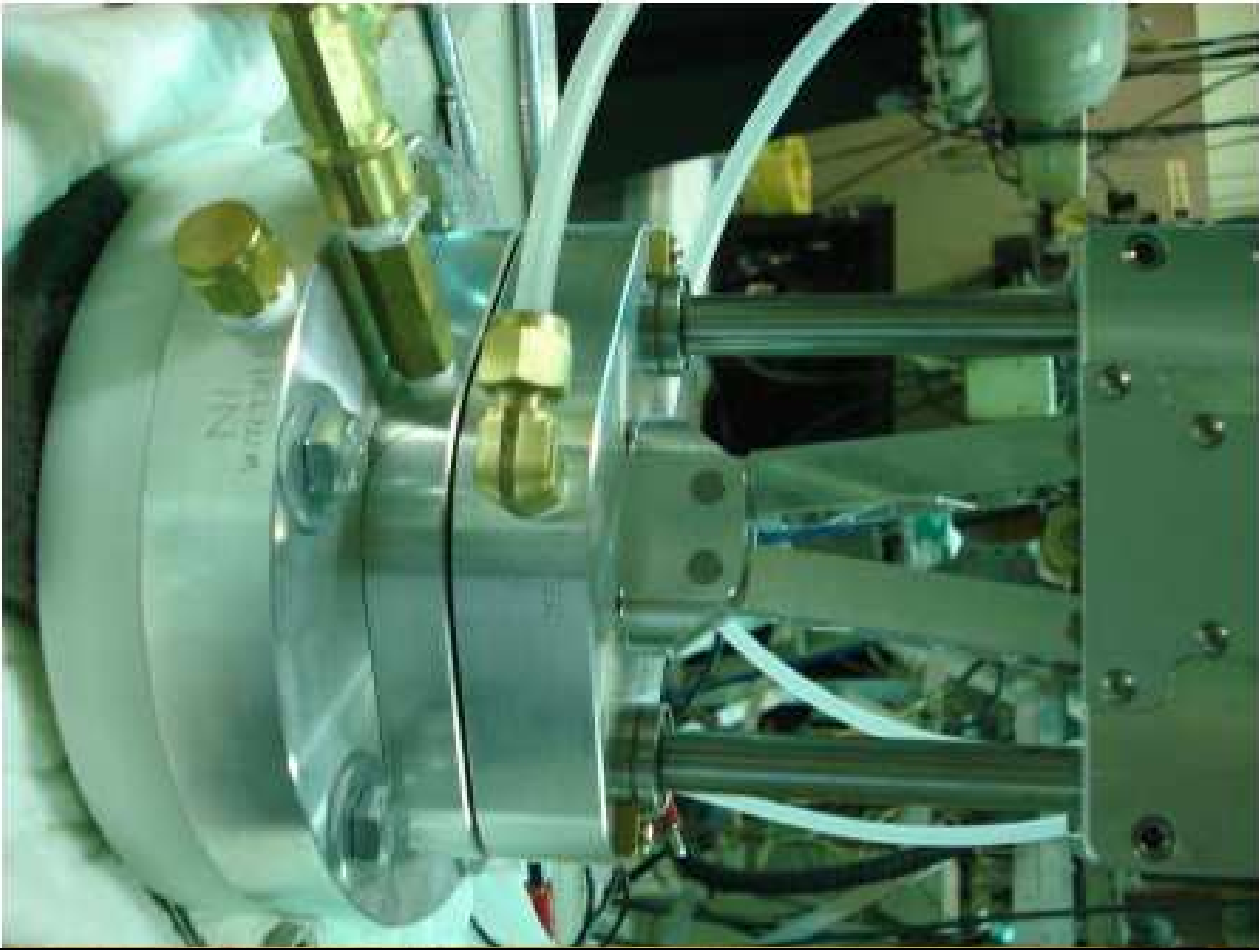
That factor creates sometimes some limitation to look the direct production of 99mTc as a real alternative way.

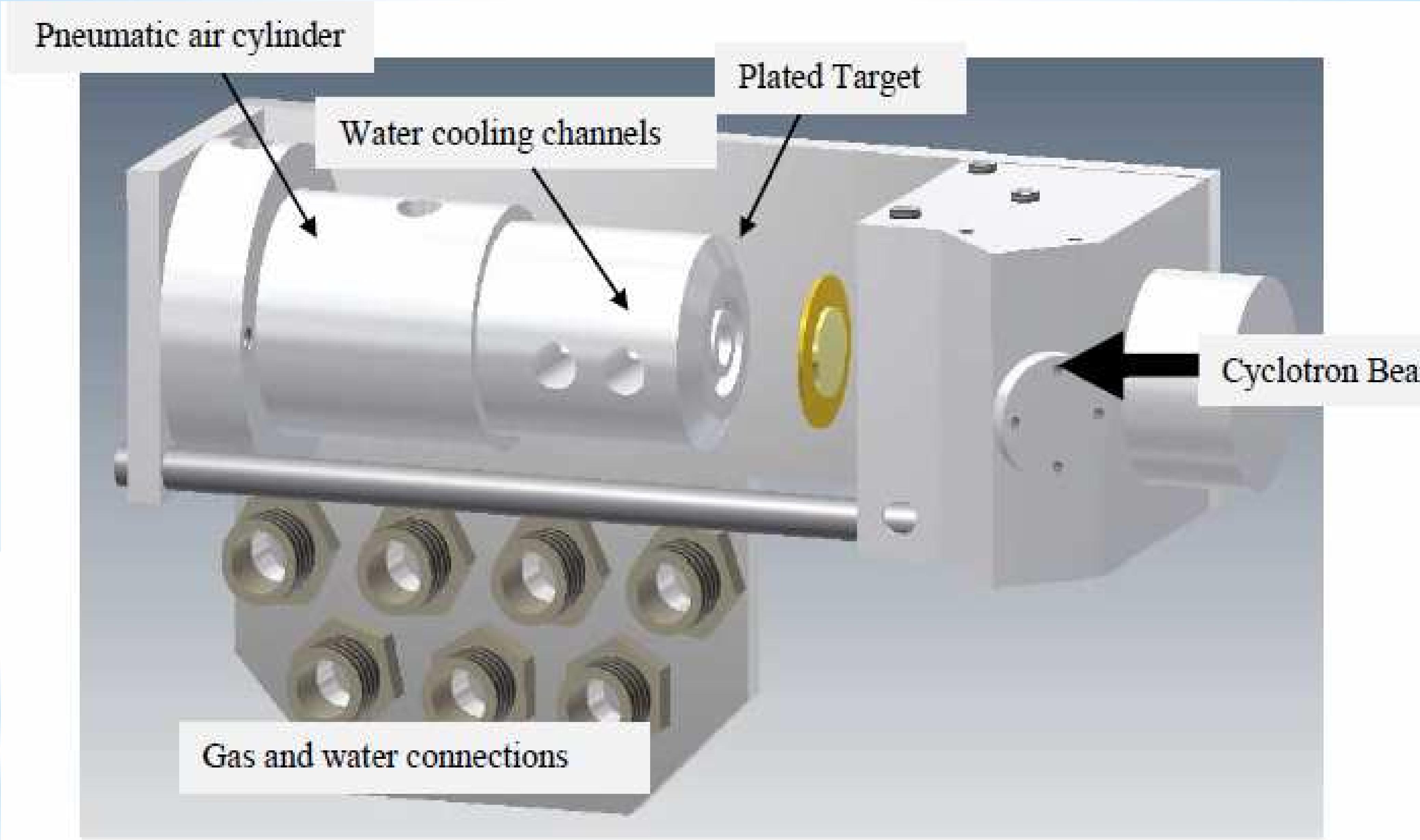
$$C = N_{mTc}/N_{m+gTc}$$

C= 26% for 99mTc eluated from Mo/Tc generator

C=19-31% for 99mTc directly produced under proton beam

With 150 μA on target using 19 MeV protons for 6 hours, up to 9 Ci (333 GBq) of ^{99m}Tc can be produced 2 to 3 times per day, which is enough to supply a large metropolitan area.




Pneumatic air cylinder
Water cooling channels
Plated Target
Cyclotron Bea
Gas and water connections

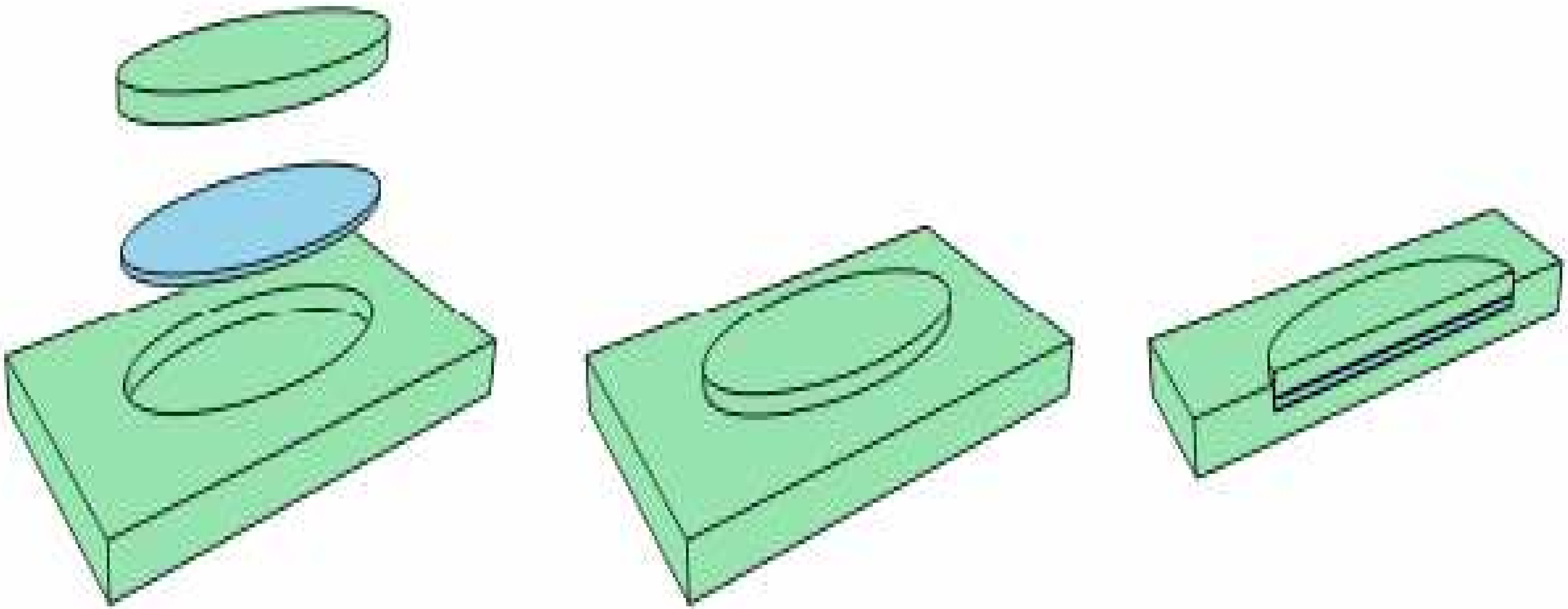
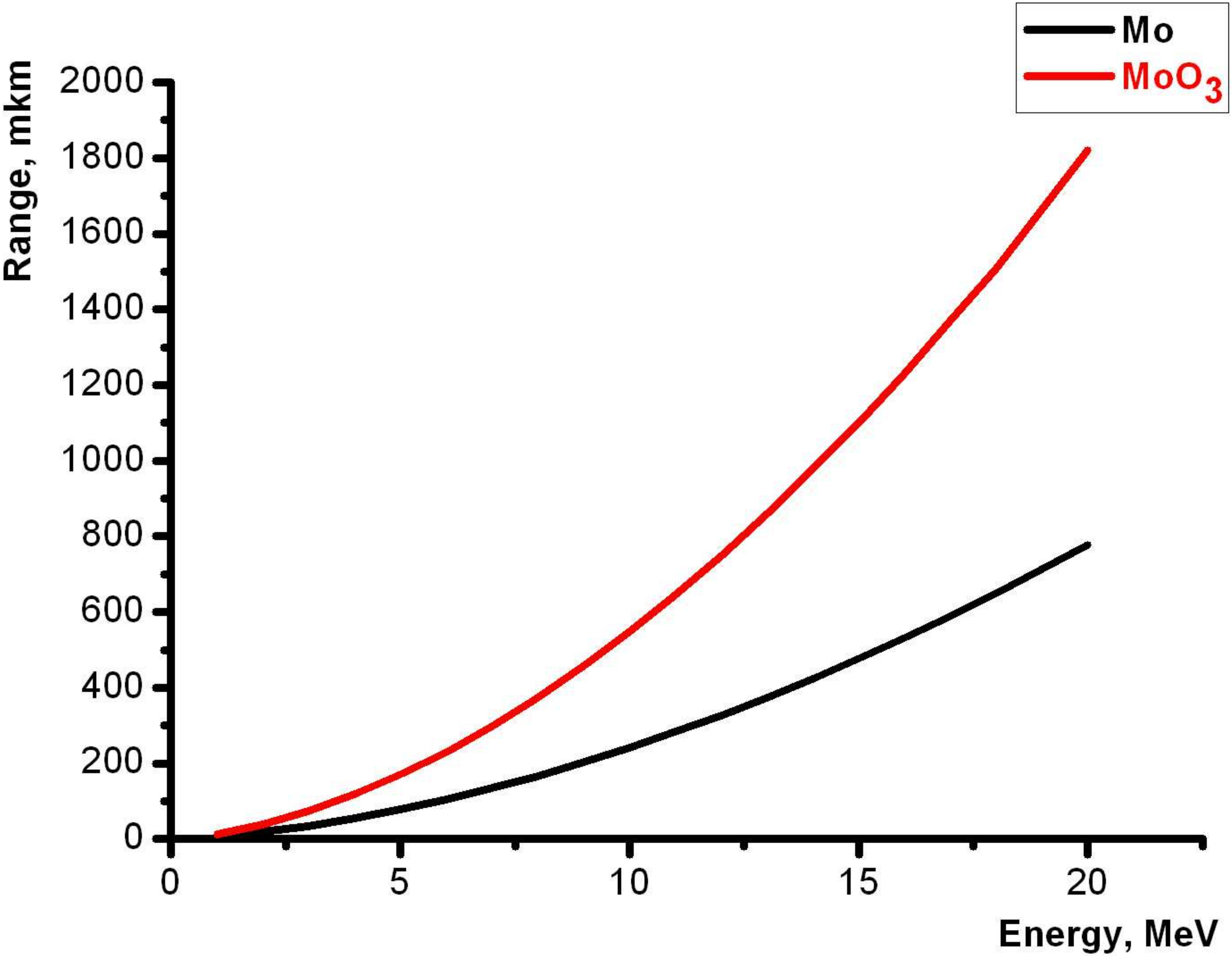


FIG. 1. Chemical Processing unit for separation of ^{99m}Tc from molybdenum using solvent extraction methodology.

Характеристики MoO_3

Тип таблетки	Спеченный тип
Плотность таблетки	3,25-3,4 г/см^3
Темп-ра сублимации	~750℃
Обилие изотопа ^{98}Mo	24%
Молекулярный вес MoO_3	143,95 (Mo:66,6% O:33,3%)
Плотность MoO_3	4,692 г/см^3
Темп-ра плавления MoO_3	780℃


Mo
MoO_3
Range, mkm
2000
1800
1600
1400
1200
1000
800
600
400
200
0
0
5
10
15
20
Energy, MeV

OUR CASE

Beam energy	**18 MeV**
Beam current	75 mkA
Irradiation time	2(3) h
Target	$^{nat}MoO_3$($^{100}MoO_3$)
Activity EOB (Ci) nat	0.1(0.14)
Activity EOB (Ci) enriched	1.05(1.54)

COMMERCIAL ASPECT

for case of enriched $^{100}MoO_3$

Target mass ~ 0.5 gram

Cost ~ 1000US$

Recovery ~95%

Loss ~5% (50US$)

Cost of final product 1000-1500 US$

Thus, the micro-factory concept and our development goals are summarized as:

*A scale of production at less than10 GBq (several hundred milli-curies) for "in-house" uses or local supplies by pre-existing medical cyclotron

*A shipping distance from each production site of nearly 1 h or 30 km

- **Operator-friendly production by fully automated equipment with lower costs**
- This method is an alternative to using HEU and thus solves the proliferation risk associated with current methods of production

Plan of activity

- Theoretical calculations and Monte-Carlo simulation of nuclear processes during different target materials irradiation under proton beam from C18 cyclotron;
- Theoretical calculation of excitation function for metallic natural $^{nat}MoO_3$ and enriched $^{100}MoO_3$ as a function of proton energy for different reactions such as

i) $^{100}Mo(p,2n)^{99m}Tc$,

(ii) $^{100}Mo(p,pn)^{99}Mo \rightarrow {}^{99m}Tc$,

(iii) $^{100}Mo(n,2n)^{99}Mo \rightarrow {}^{99m}Tc$,

(iv) $^{100}Mo(p,2p)^{99}Nb \rightarrow {}^{99}Mo \rightarrow {}^{99m}Tc$,

(v) $^{98}Mo(n,\gamma)^{99}Mo \rightarrow {}^{99m}Tc$ and

(vi) $^{98}Mo(p,\gamma)^{99m}Tc$.

- Collect the reported cross-section data. Analyze and evaluate the data.
- Calculation tool for having estimations of Tc radioisotopes production vs energy range, irradiation time and beam currents.

- **R&D of Mo or MoO_3 target for irradiation under cyclotron proton beam**
- **Experimental measurement of ^{99m}Tc production yield for different energies of protons, irradiation time and intensity, as well as for other isotopes**

- Comparison of experimental measurements with literature data (theoretical and measured) as well as with own results
- Development of the methods of ^{99m}Tc extraction from irradiated material
- Development of target material recovery for multiple use

- **Examination of radionuclide impurities by gamma spectroscopy**
- **Investigation of the radiological impurity of final ^{99m}Tc**
- **Preparation of full technology documentation for a ^{99m}Tc direct production under C18 proton beam.**

CONCLUSION

- The world activity in the area of 99mTc direct production area is under extremely high attention (see Report from…..
- The bases of awaiting success in our department activity are

❖ Accumulated experience during ISTC projects execution and theoretical and experimental investigations of photonuclear reactions

- **Enough reach world data reported on many conferences**
- **Awaiting C18/18 cyclotron will be commissioned till end of 2013**
- **Created trial production on separate building**
- **Enough good instrument base such as HP Ge detector with digital analyzer, centrifuge extractor, exhaust hoods etc.**
- **Experienced team and students.**

Thanks for attention!