Production of Medical Isotopes From a Thorium Target Irradiated by Light Charged Particles up to 70 MeV

Duchemin C., Guertin, A., Haddad, F., Michel, N. and Métivier, V. Phys. Med. Biol. **60 (2015) 931–946**

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SUBATECH Laboratory

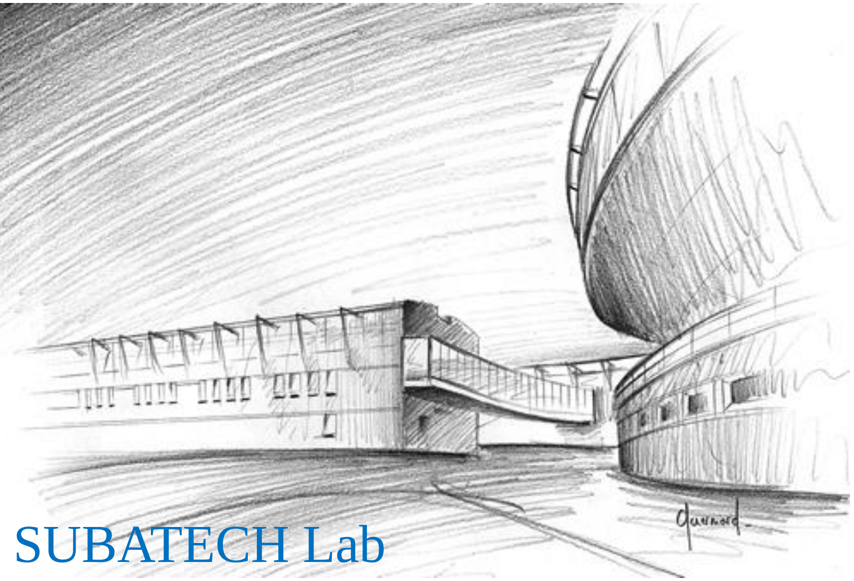
Nantes, France

Workshop on Compact Accelerators for Isotope Production

The Cockcroft Institute, Warrington (UK)

26th March 2015

Context



SUBATECH Lab

Artistic view

SUBATECH Lab

PRISMA group

- Sensors development for robotics
- Non destructive analysis of materials
- **Radio-isotopes production for medical applications**

In close collaboration with the

ARRONAX cyclotron

Accelerator for the
Research in
Radiochemistry and
Oconlogy at
Nantes
Atlantique
X

ARRONAX cyclotron
(Saint-Herblain, 44)

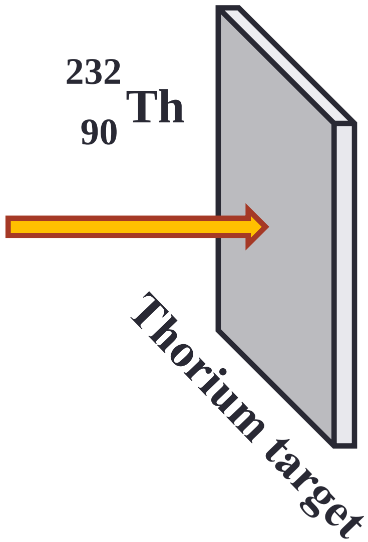
ARRONAX

Particles	Energy (MeV)	Max. current (µA)	Dual beam
Protons	30-70	2 x 350	Yes
Deuterons	15-34	2 x 50	Yes
Alphas	68	70	No

Multi-particles
High energy
High intensity

- 4 vaults (P2, P3, A1, A2) devoted to isotope production and connected to hot cells through a pneumatic system
- 1 vault (P1) devoted to a neutron activator system
- 1 vault (AX) devoted to physics, radiolysis and radiobiology experiments

Motivations



- Irradiation using light charged particles: deuterons and protons
- Production of radioisotopes of medical interest

ACTIVATION PRODUCTS

- Pa-230 → U-230 → Th-226
- Ac-225 → Fr-221 → At-217 → Bi-213
- Th-227 → Ra-223

Alpha emitters for α targeted therapy

FISSION PRODUCTS

- Mo-99 → Tc-99, I-131
- Cd-115g → In-115m

Isotopes for diagnosis and therapy

⇨ **Measure production cross sections for both projectiles and compare production yields**

Cross section measurements @ ARRONAX

First, measurements using deuterons as projectiles. Motivations:

→ In some cases, they give higher production yields (i.e. Cu-64)

→ Available with ARRONAX up to 35 MeV

→ Improvements of data base and referenced cross sections

→ Obtain large set of data to constrain theoritical codes like TALYS

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- Nuclear reaction program → light particles on nuclei heavier than carbon.

- Many theoretical models to predict observables as a function of the incident particle energy (from 1 keV to 1 GeV).

- Default models in the code, defined by the authors
- Code can work with few informations: target type, incident particle and energy.

- Other models included in the code can be better to describe the exp. σ.

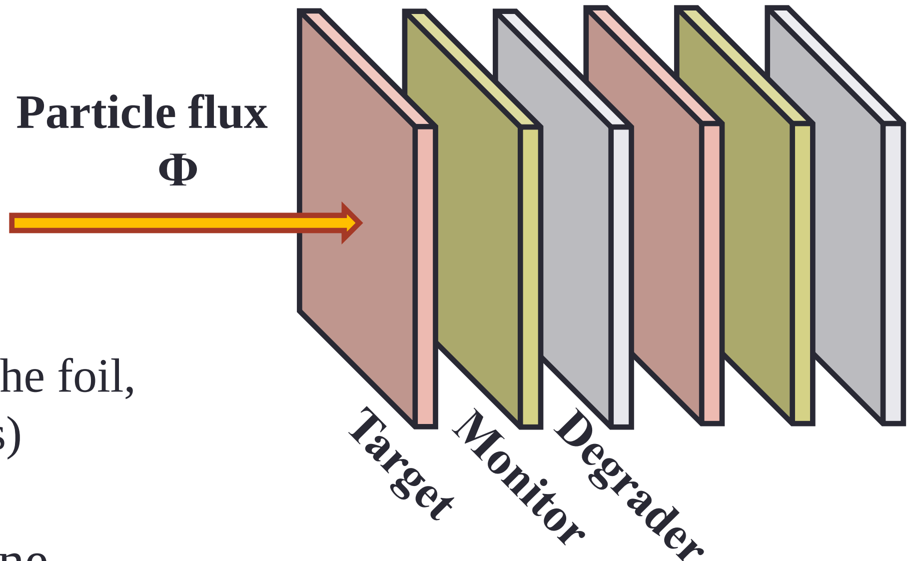
* Koning A J and Rochman D 2012 Modern nuclear data evaluation with the TALYS code system *Nucl.Data Sheets* ***113 2841***

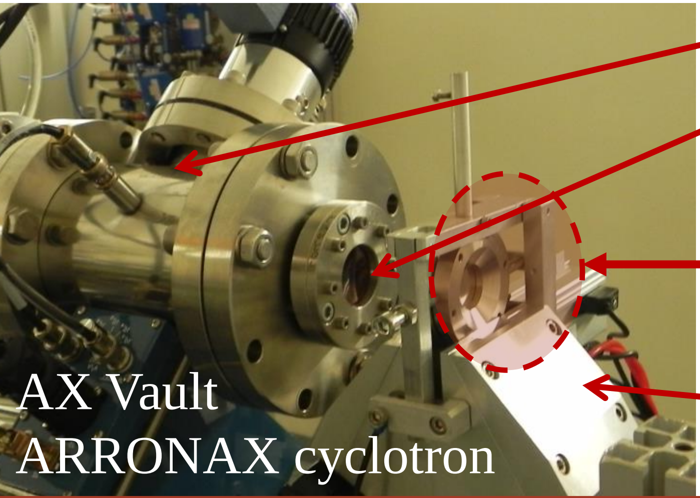
Cross section measurements

1 → **AX Vault**
Cross section measurements

STACKED-FOILS TECHNIQUE

- Thin foils (few µm)
- Energy loss constant and small
- One thin foil
  - → one energy (taken in the middle of the foil, after SRIM simulations)
  - → one cross section value

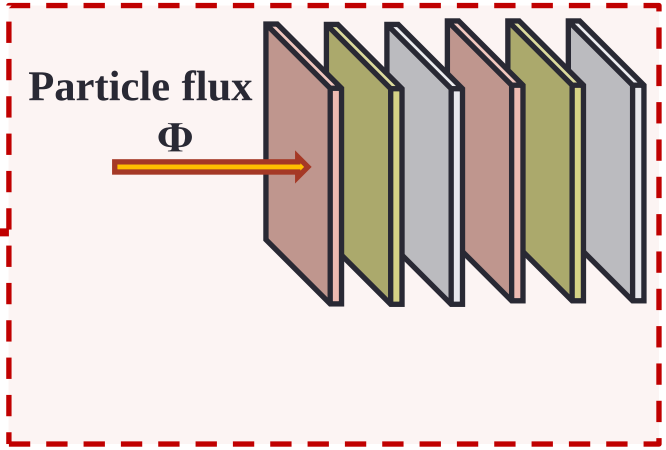




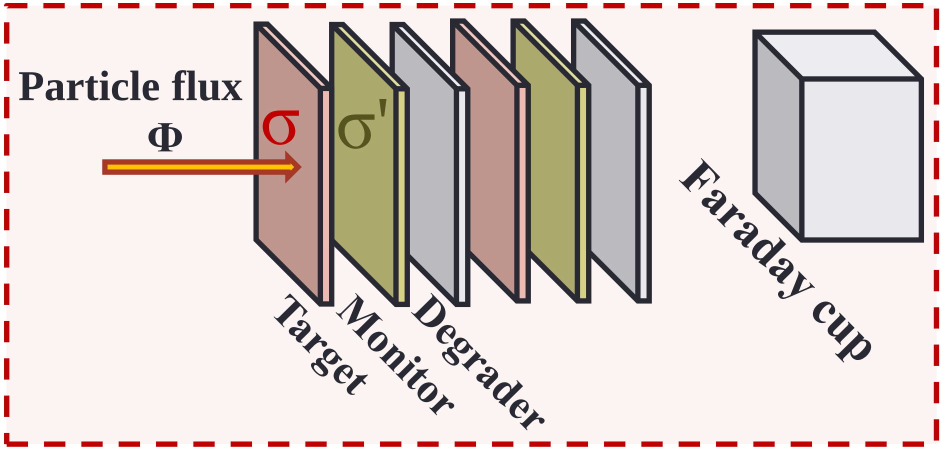
Beam line

Kapton foil
(to close the beam line)

Irradiation station



Data Measurements

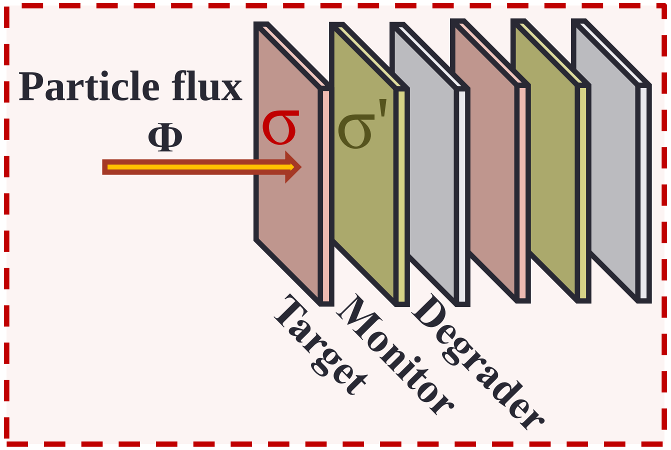


$$\sigma = \frac{\mathrm{Act} \cdot A}{\Phi \cdot \chi \cdot \rho \cdot e_f \left(1 - e^{-\lambda t}\right)}$$

- A Faraday cup is placed at the end of the stack to measure the intensity during the irradiation

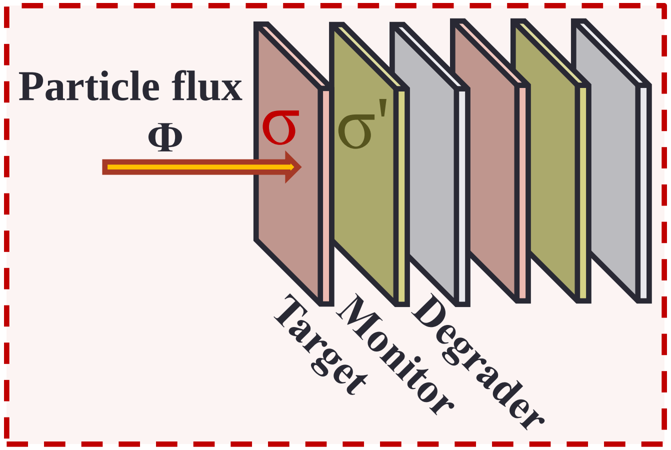
- Flux attenuation all along the stack?

Data Measurements



$$\sigma = \sigma' \cdot \frac{\chi' \cdot \mathrm{Act} \cdot A \cdot \rho' \cdot e_f' \left(1 - e^{-\lambda' t}\right)}{\chi \cdot \mathrm{Act}' \cdot A' \cdot \rho \cdot e_f \left(1 - e^{-\lambda t}\right)}$$

Data Measurements



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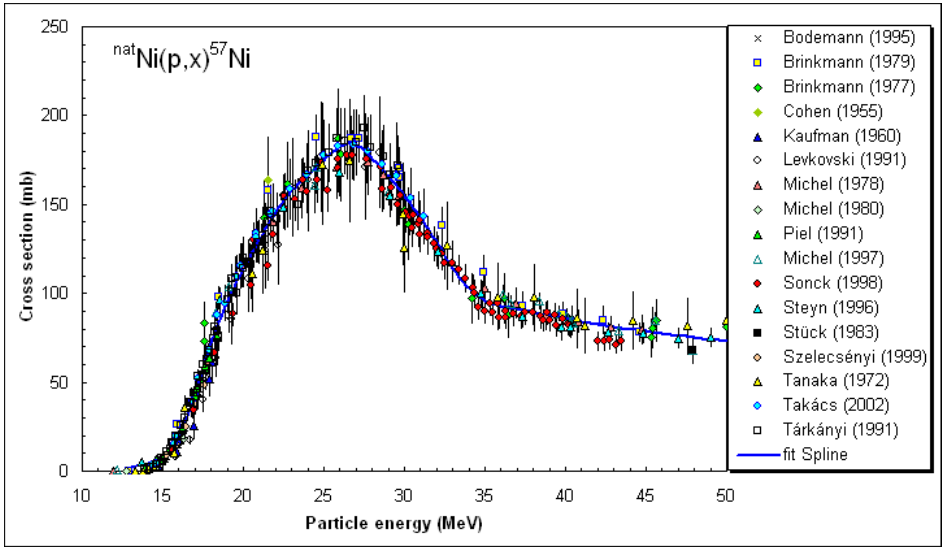
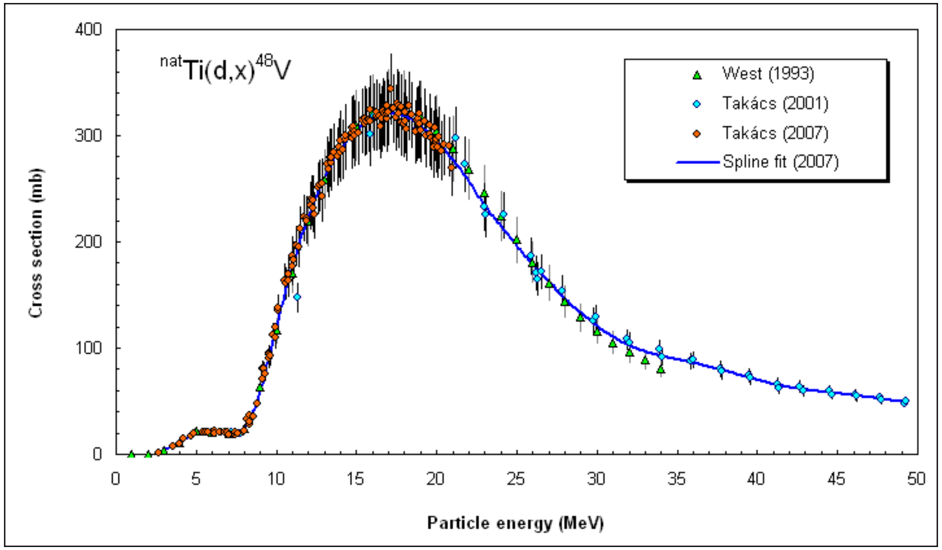
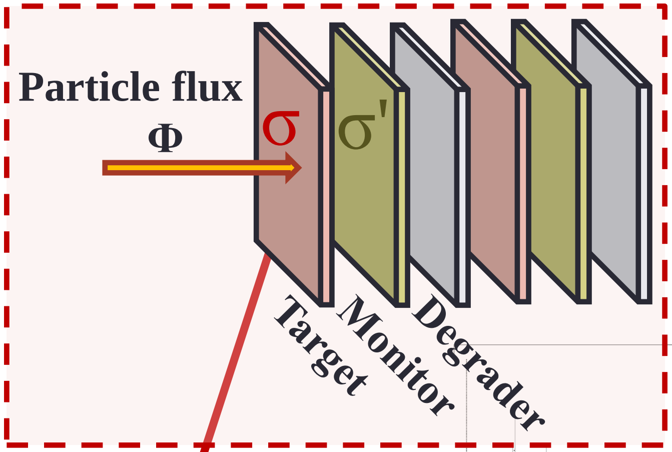
Monitor reactions used :



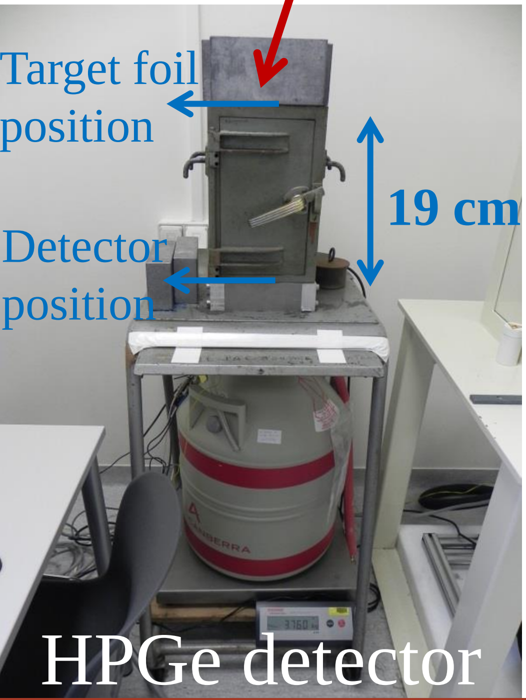


International Atomic Energy Agency (IAEA) - Medical Applications @ Nuclear Data Section

Data Measurements

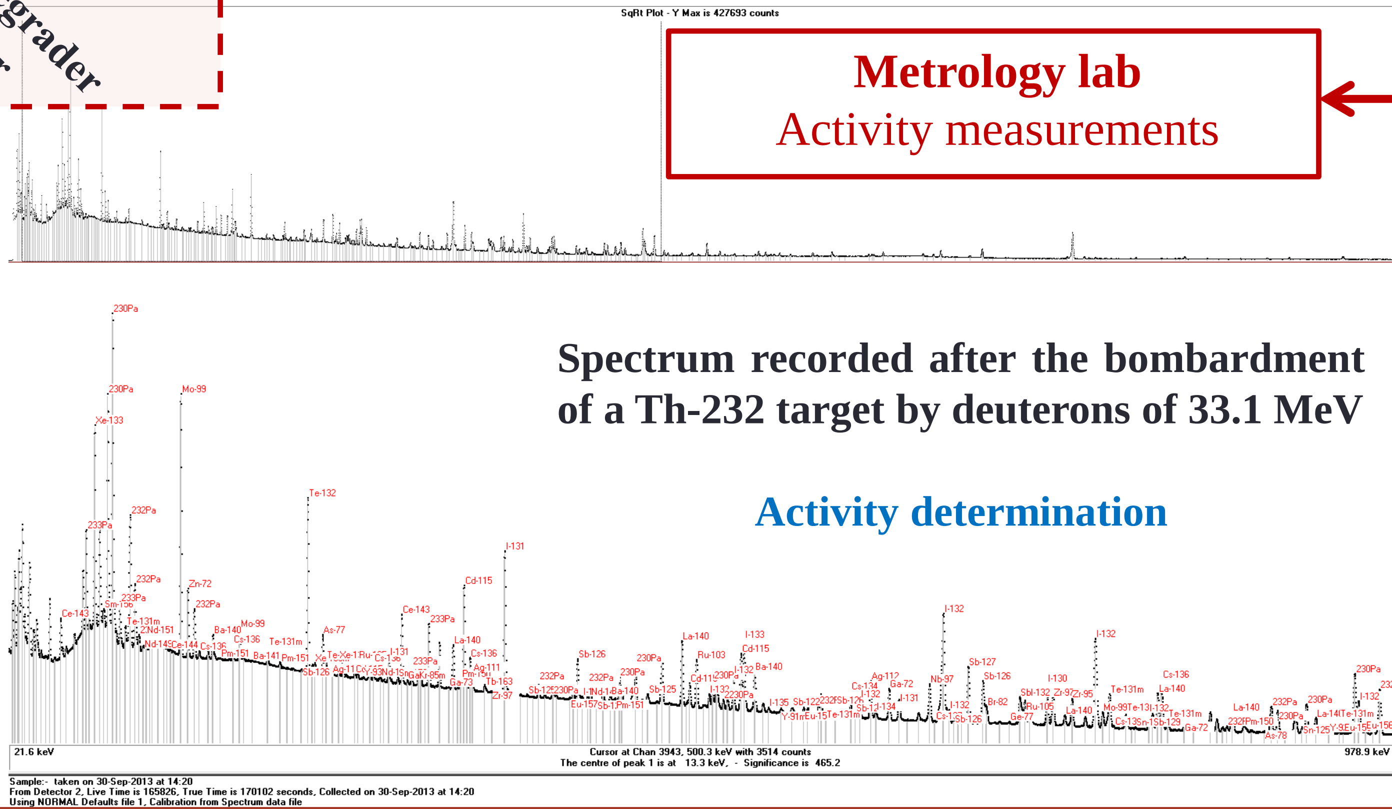


$$\sigma = \sigma' \cdot \frac{\chi' \cdot Act \cdot A \cdot \rho' \cdot e'_f \left(1 - e^{-\lambda' t}\right)}{\chi \cdot Act' \cdot A' \cdot \rho \cdot e_f \left(1 - e^{-\lambda t}\right)}$$



Metrology lab

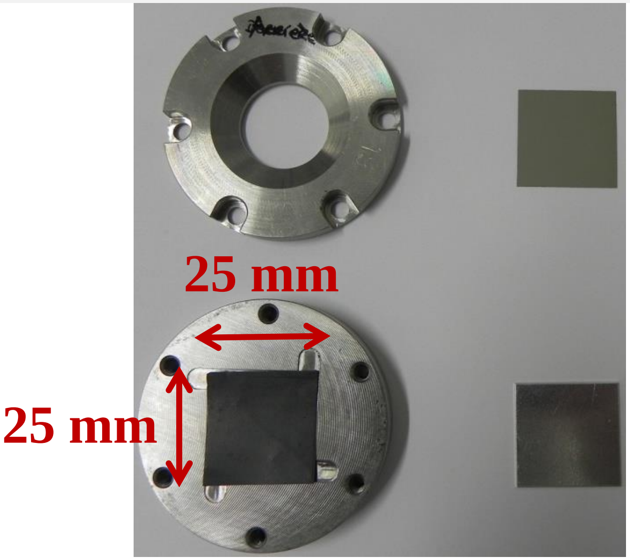
Activity measurements



Spectrum recorded after the bombardment of a Th-232 target by deuterons of 33.1 MeV

Activity determination

Experimental conditions



Typical irradiations :

→ Time of irradiation : 30 minutes

→ Beam intensity : 100 nA

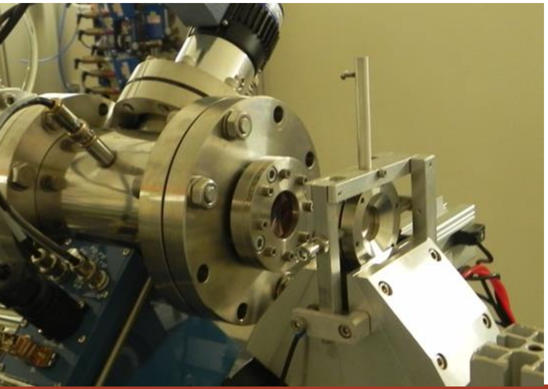
→ Cooling time : around 14 hours

To limit errors :

→ Several activity measurements, depending on the half-lives

→ Each thickness of the foils have been precisely measured:

Weighted to obtain its mass and scanned to determined its area

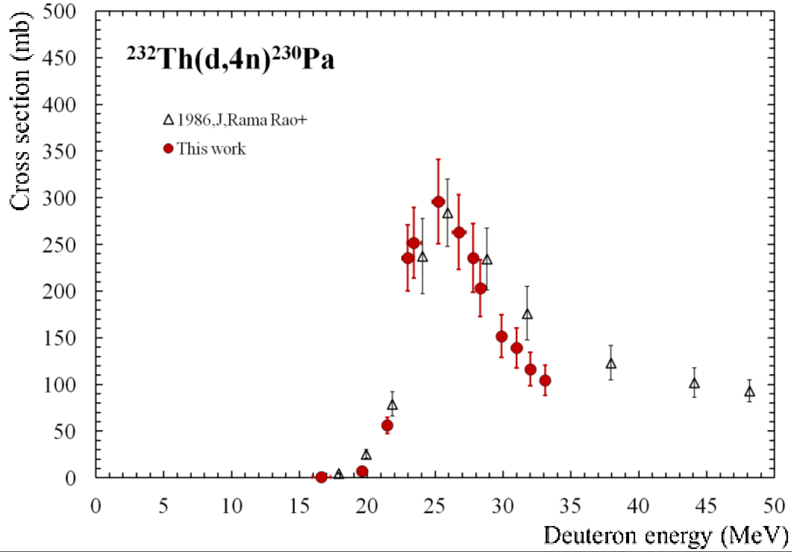


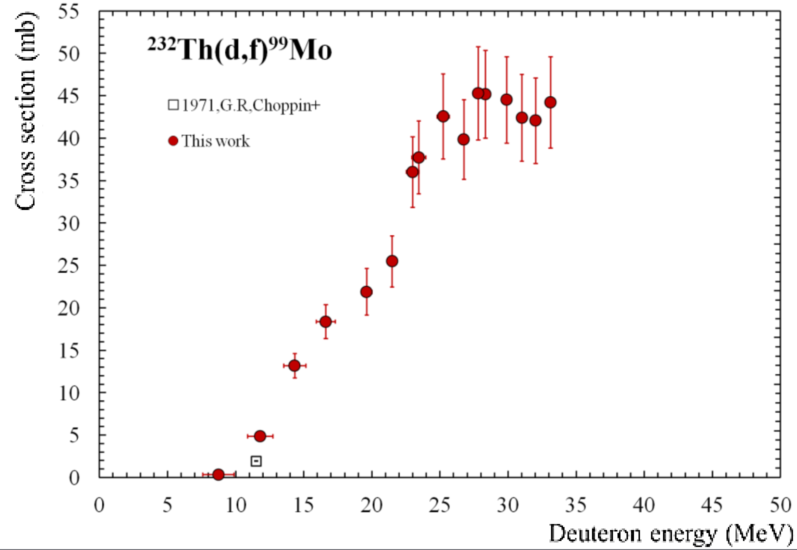
Production Cross Section

Activation product: Pa-230 → U-230 → Th-226 (α)

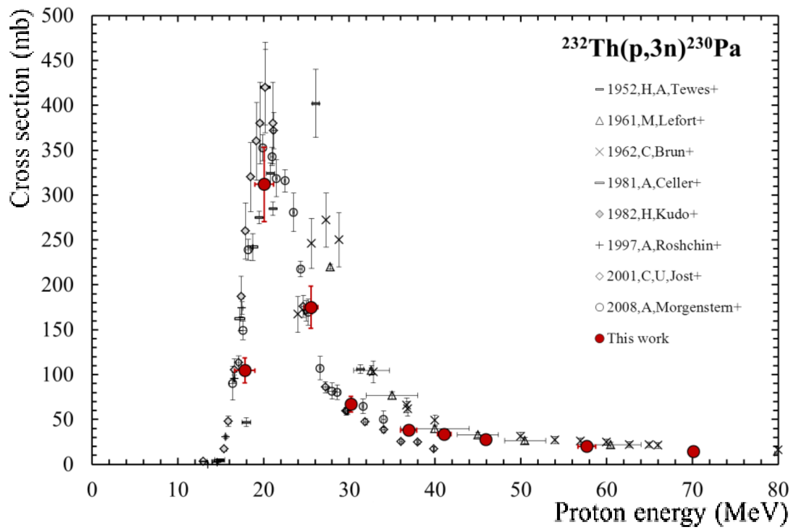
Fission product: Mo-99 → Tc-99m

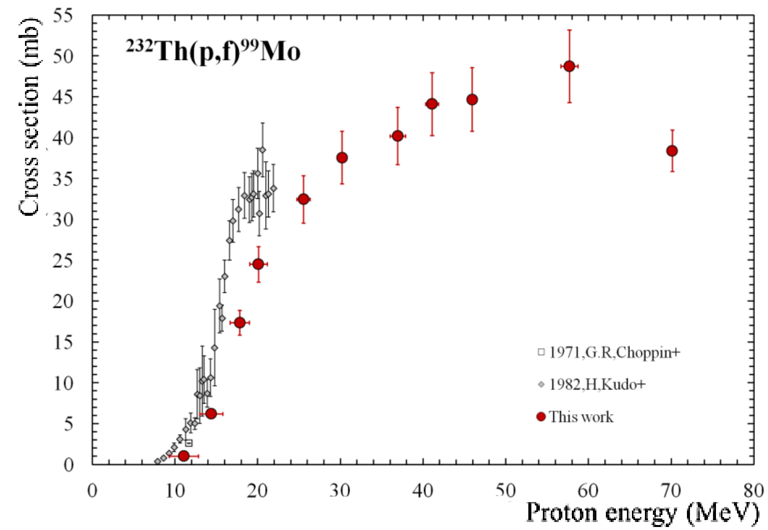
DEUTERONS





PROTONS

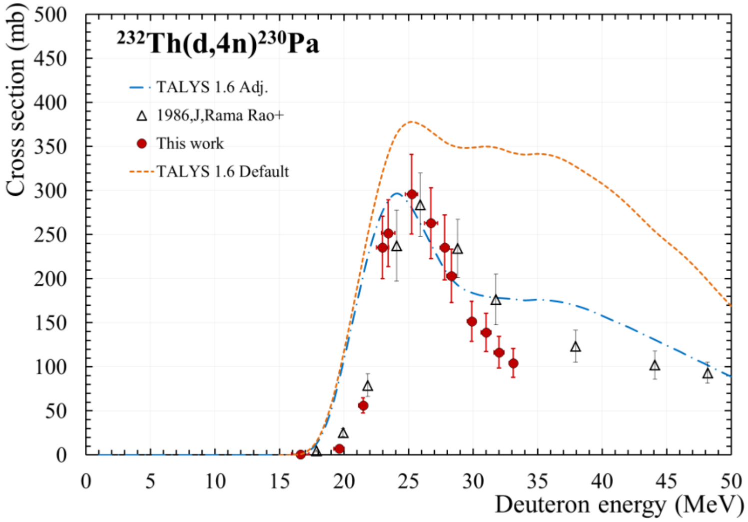


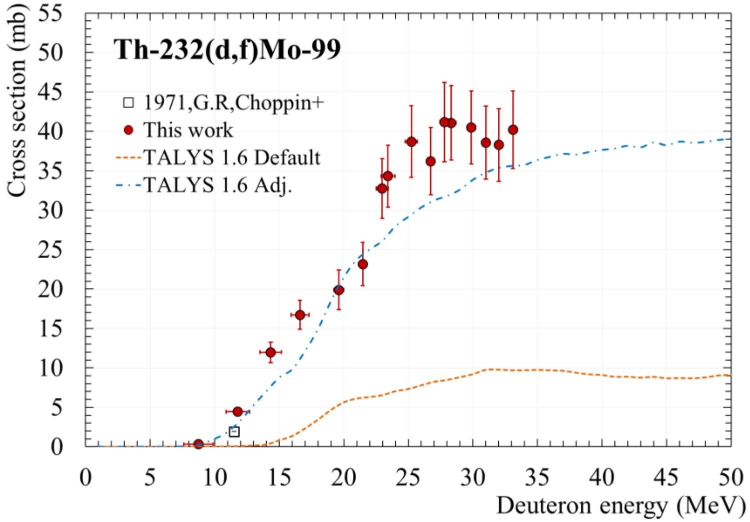


Comparisons with TALYS

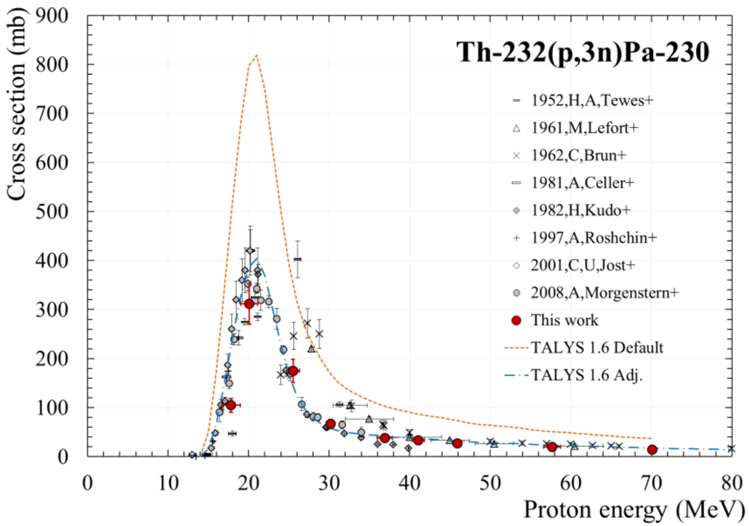
Talys 1.6: Comparison between default and « adjusted »

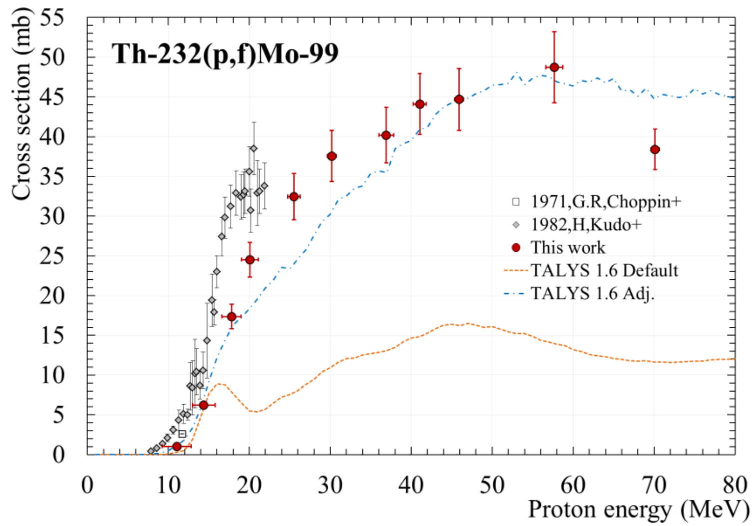
DEUTERONS



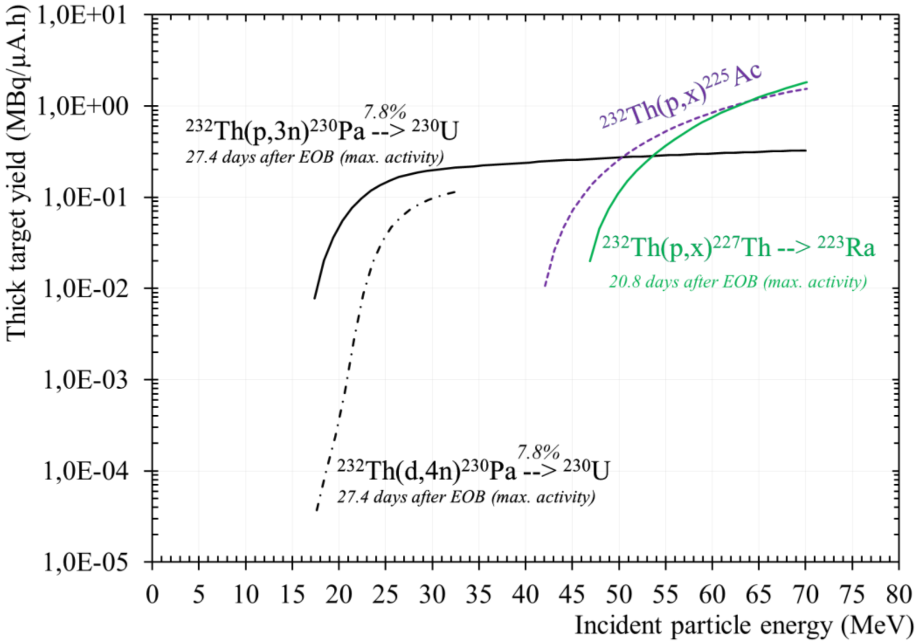


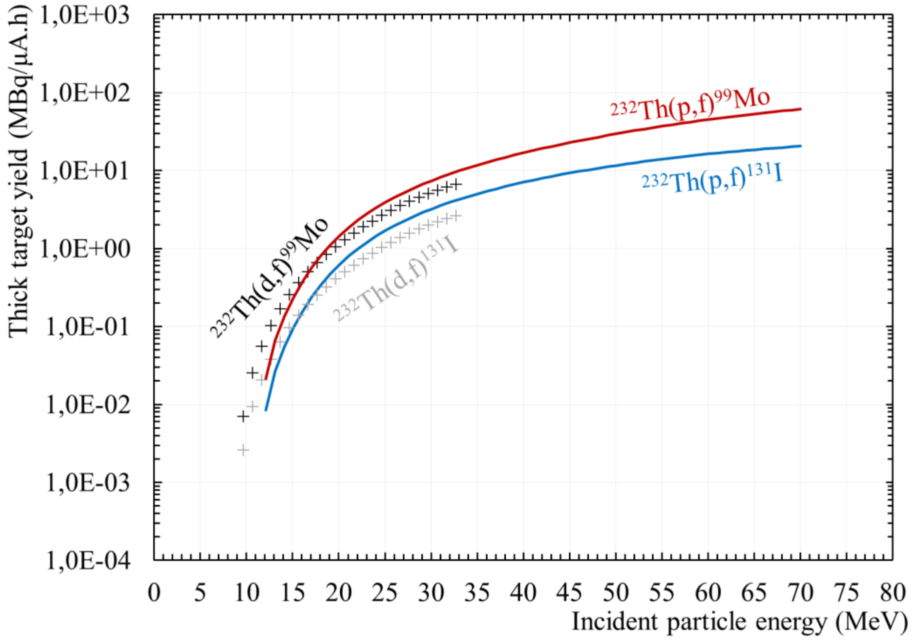
PROTONS





Thick Target Production Yield (TTY)





The use of protons as projectiles allows to:

- Produce ^{225}Ac and ^{223}Ra
- Obtain higher ^{230}U production rate than with deuterons
- Obtain higher ^{99}Mo and ^{131}I production rate

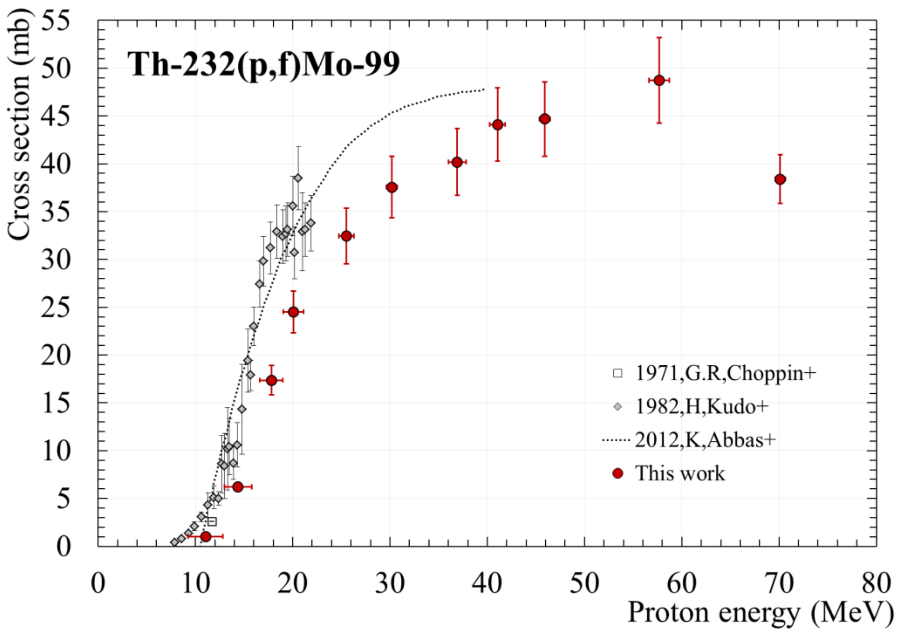
Medical isotopes production in a Th target

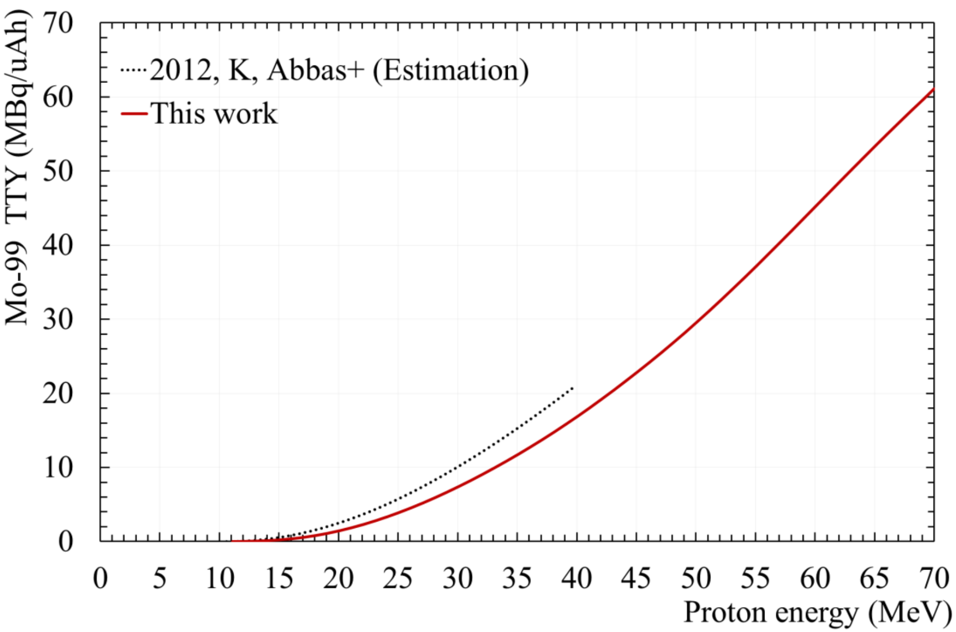
- Review of the medical radioisotopes products in a thorium target irradiated by protons of 70 MeV

- Production rate using **70 MeV proton beam @ 350 µA** during 3 days (based on Mo-99 half-life) → 90 runs / year

	Radio-isotope	Half-life $T_{1/2}$	TTY at 70 MeV (MBq/µA.h)	Prod. 350 µA during 3 days (Ci)	Prod. on a dedicated machine at 70 MeV 90 runs/year (Ci)
Activation products	U-230	20.8 d	0.3	0.2	18
	Ac-225	10.0 d	1.6	1.1	99
	Ra-223	11.435 d	1.8	1.2	108
Fission products	I-131	8.0207 d	20.6	14.0	1 260
	Cd-115g	53.46 h	83.1	56.6	5 094
	Mo-99	65.94 h	61.1	41.6	3 744

Focus on Mo-99 and K. Abbas estimation





- In 2012, estimation of the Mo-99 cross section up to 40 MeV (K. Abbas et al.) from the experimental values of H. Kudo et al., 1982.

- This work is not in agreement with H. Kudo et al., 1982
→ disagreement with the K. Abbas et al., 2012, estimation (≠ up to 50%)

- Mo-99 TTY obtain in this work is up to 25% lower (at 40 MeV) than the one estimate by K. Abbas et al., 2012.

→ **And with other production routes ?**

Focus on Mo-99 TTY

- **Comparison between different production routes with data available in the literature**
- Production rate using the parameters available at **ARRONAX** with an irradiation duration of **3 days** (based on Mo-99 half-life)

- Production rate using **protons as projectiles**
- **350 µA**

- Production rate using **deuterons as projectiles**
- **50 µA**

- Production rate using **alphas as projectiles**
- **70 µA**

Focus on Mo-99 TTY

Group	Reaction	Author	Energy (MeV)	TTY (MBq/µA.h)	Production **Protons: 350 µA** **Deuterons: 50 µA** **Alphas: 70 µA** during 3 days (Ci)	Production on a dedicated machine with 90 runs/year (Ci)	Specific activity
Protons	**^{100}Mo(p,p+n)^{99}Mo**	**F.Tarkanyi (2012)**	**38**	**98.8**	**67.3**	**6 057**	
Protons	**^{nat}U(p,f)^{99}Mo**	**M.A.M. Uosif (2005)**	**70**	**80.4**	**54.8**	**4 932**	
Protons	**^{232}Th(p,f)^{99}Mo**	**This work (2014)**	**70**	**61.1**	**41.6**	**3 744**	
Protons	**^{nat}Pb(p,f)^{99}Mo**	**J.Kuhnhenn (2001)**	**70**	**1.3**	**0.9**	**81**	
Deuterons	**^{100}Mo(d,p+2n)^{99}Mo**	**F.Tarkanyi (2011)**	**35**	**76.0**	**7.4**	**666**	
Deuterons	**^{232}Th(d,f)^{99}Mo**	**This work (2014)**	**33**	**6.7**	**0.7**	**63**	
Alphas	**^{100}Mo(α,α+n)^{99}Mo**	**V.N.Levkovskij (1991)**	**46**	**2.0**	**0.3**	**25**	
Alphas	**^{96}Zr(α,n)^{99}Mo**	**G.Pupillo (2014)**	**30**	**1.6**	**0.2**	**18**	

Conclusion

- New cross section measurements have been made for ^{232}Th(p,x) and ^{232}Th(d,x) reactions, using the stacked-foils technique at ARRONAX (St Herblain, France).

- **Activation:** new data have been obtained for Pa-230, Ac-225, Ra-223

- **Fission:** new data up to 70 MeV using protons
  first data on the energy range 8-33 MeV using deuterons as projectiles

26 fission products have been detected including Mo-99, Cd-115g and I-131 in both case.

Mo-99 TTY obtained in this work is up to 25% lower than the one estimate by K.Abbas et al., 2012 up to 40 MeV.

3.7 kCi could be annually obtained on a dedicated 70 MeV 350 µA machine using the reaction Th-232(p,f)Mo-99 and considering 90 runs/year.

but other production routes lead to less contaminants

Some comparisons with the TALYS 1.6 code: default models don't well reproduce the experimental data

other models include in the code are better

checked with protons and deuterons on different targets

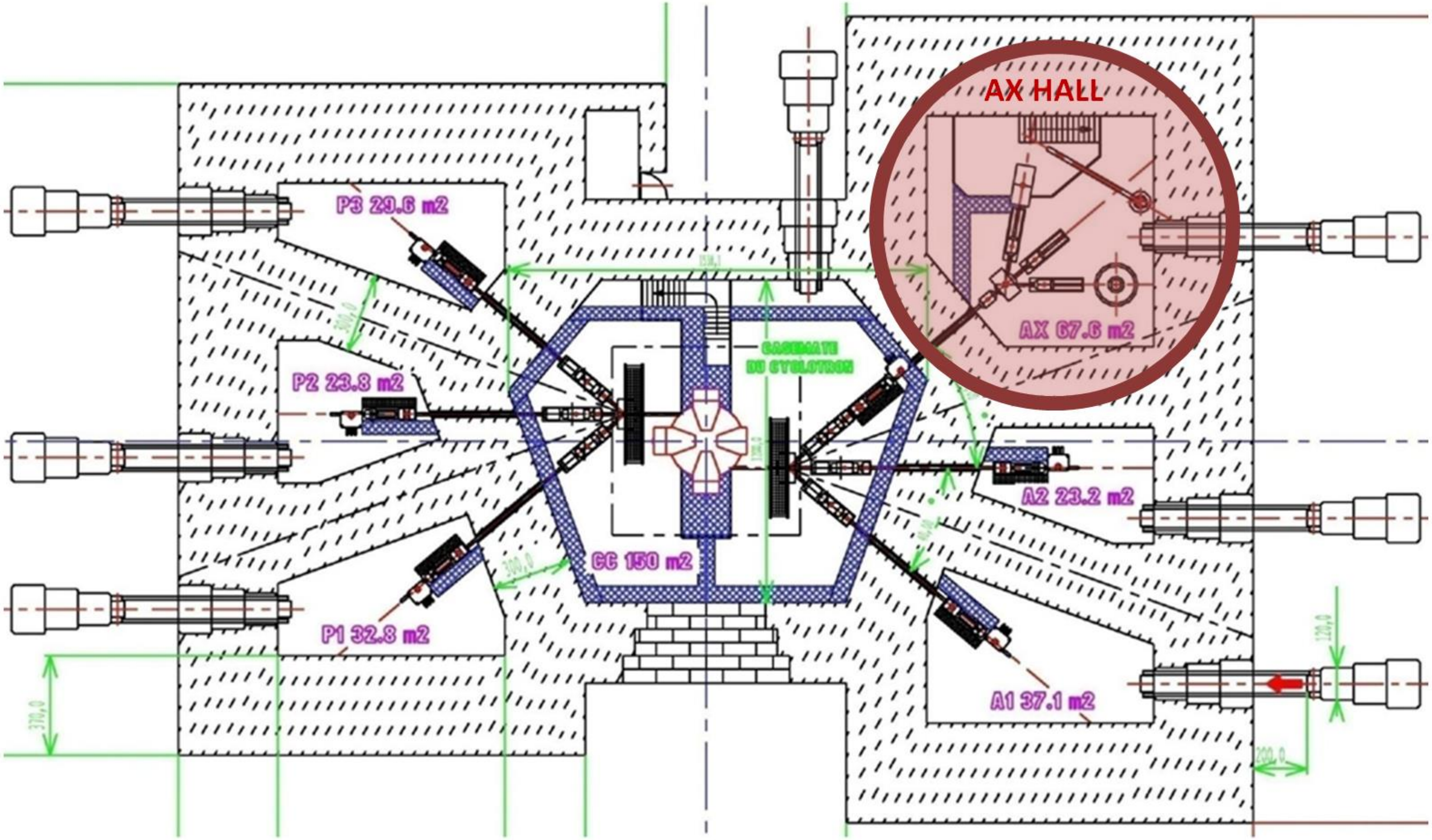
THANK YOU FOR YOUR ATTENTION

Production of Medical Isotopes From a Thorium Target Irradiated by Light Charged Particles up to 70 MeV

Duchemin C., Guertin, A., Haddad, F., Michel, N. and Métivier, V. Phys. Med. Biol. **60 (2015) 931–946**

The ARRONAX cyclotron is a project promoted by the Regional Council of Pays de la Loire financed by local authorities, the French government and the European Union. This work has been, in part, supported by a grant from the French National Agency for Research called "Investissements d'Avenir", Equipex Arronax-Plus nºANR-11-EQPX-0004 and Labex nºANR-11-LABX-0018-01.

ARRONAX cyclotron

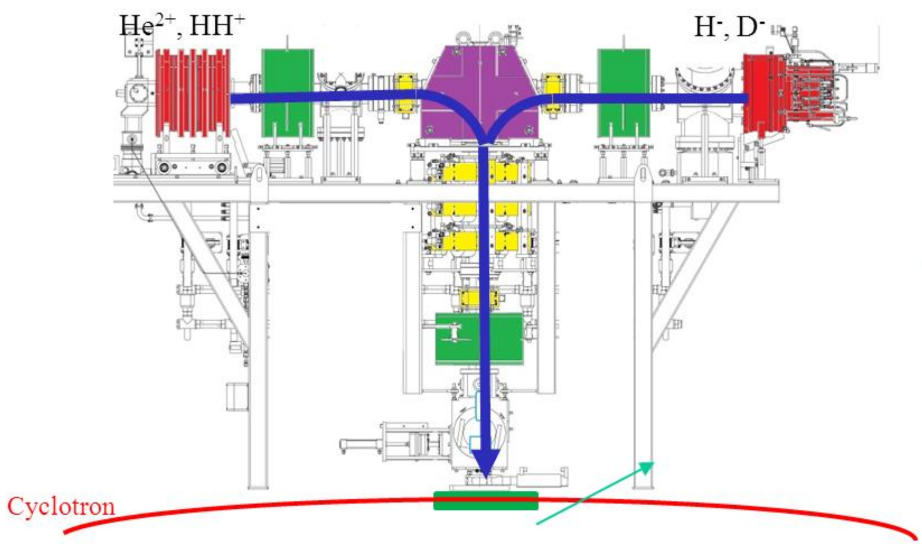


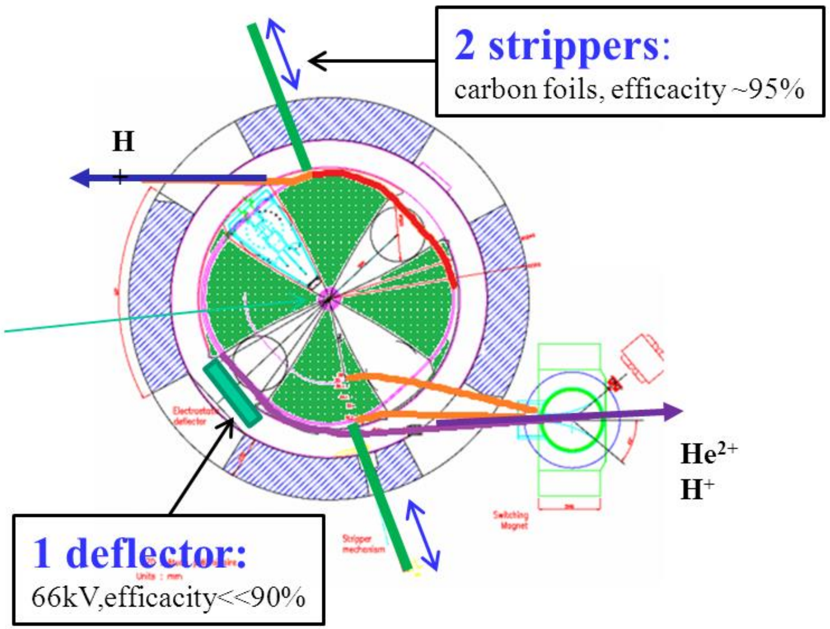
ARRONAX cyclotron

C70 Cyclotron build by IBA:

- **4 sectors** isochron **cyclotron** (~ 4m of diameter)
  - RF: 30.45 MHz
  - Acceleration Voltage: 65 kV
  - Max magn. field : 1.6 T
  - Max kin. energy/n: 30-70 MeV
- **2 multi-particle sources**:
  - H^-,D^-: multicusp, 5 mA max.
  - He^{2+},HH^+: supernanogan ECR
- Extraction: stripper ($^-$) or electrostatic deflector ($^+$)

Extracted	Energy (MeV)	Max. current (μA)
H^+	30 – 68.7	2 x 350
D^+	15 – 35	2 x 50
He^{2+}	67.4	70
HH^+	17	50

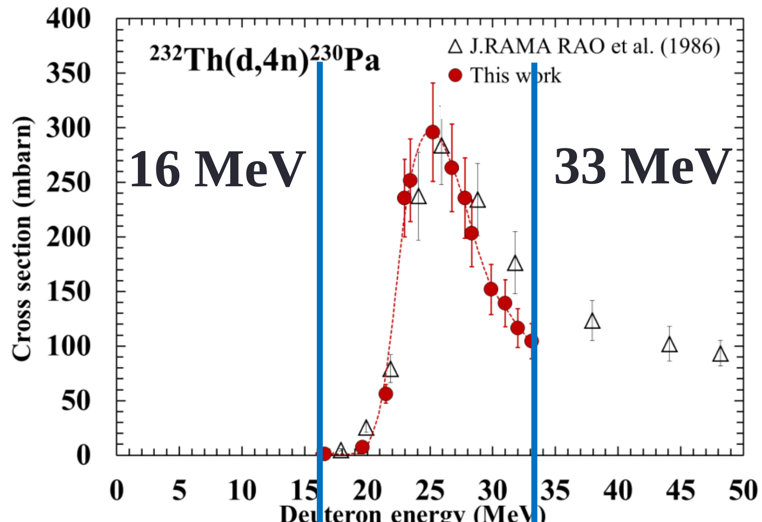


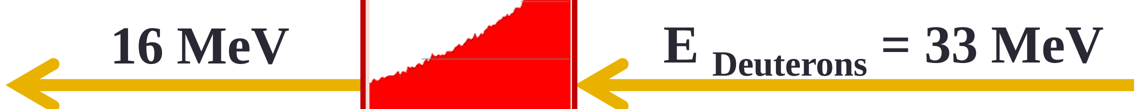


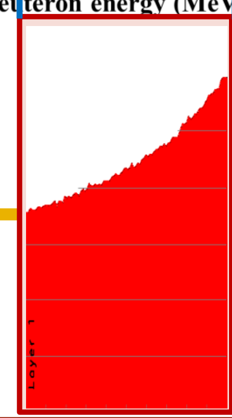
Thick Target Yield (MBq/µAh)

- Determination of the production rate of a radionuclide from its cross section

Pa-230 → **U-230** → **Th-226**





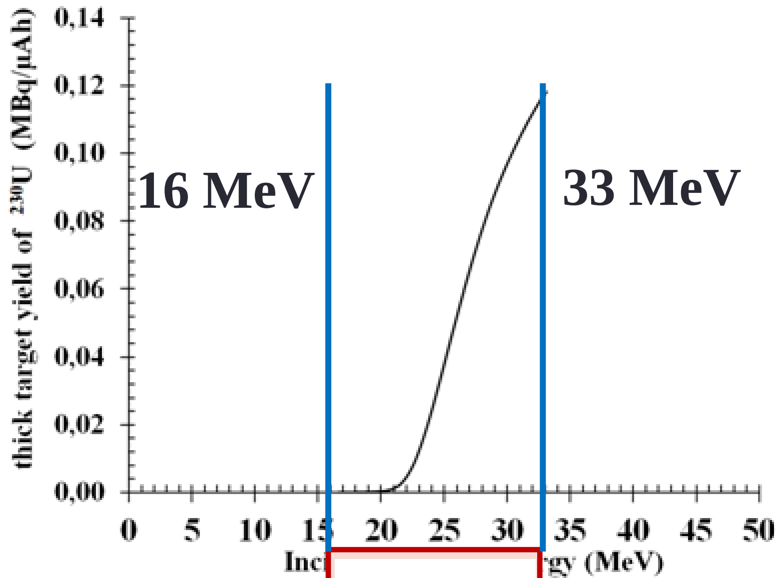


$$TTY = \Phi.\chi.\frac{N_a.\rho}{A}(1 - e^{-\lambda.t}) \int_{Emin}^{Emax} \frac{\sigma(E)}{\frac{dE}{dx}} dE$$

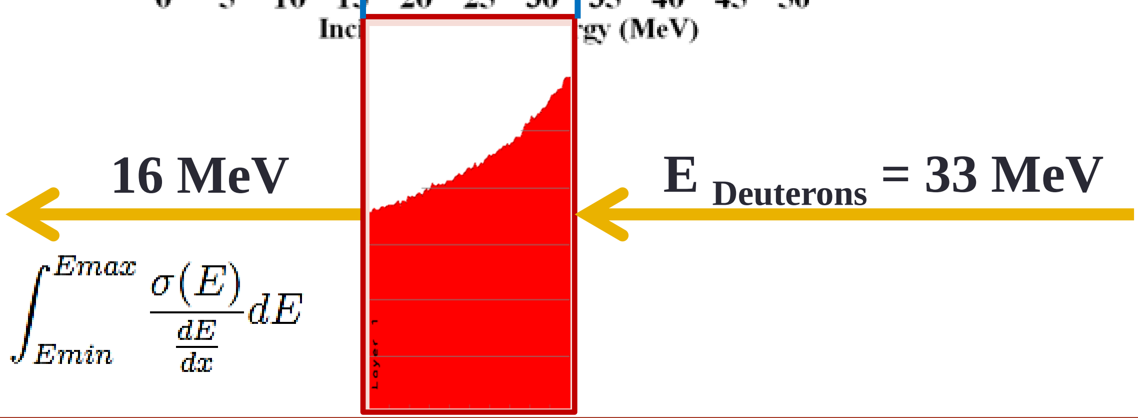
Thick Target Yield (MBq/µAh)

- Determination of the production rate of a radionuclide from its cross section

Pa-230 → U-230 → Th-226

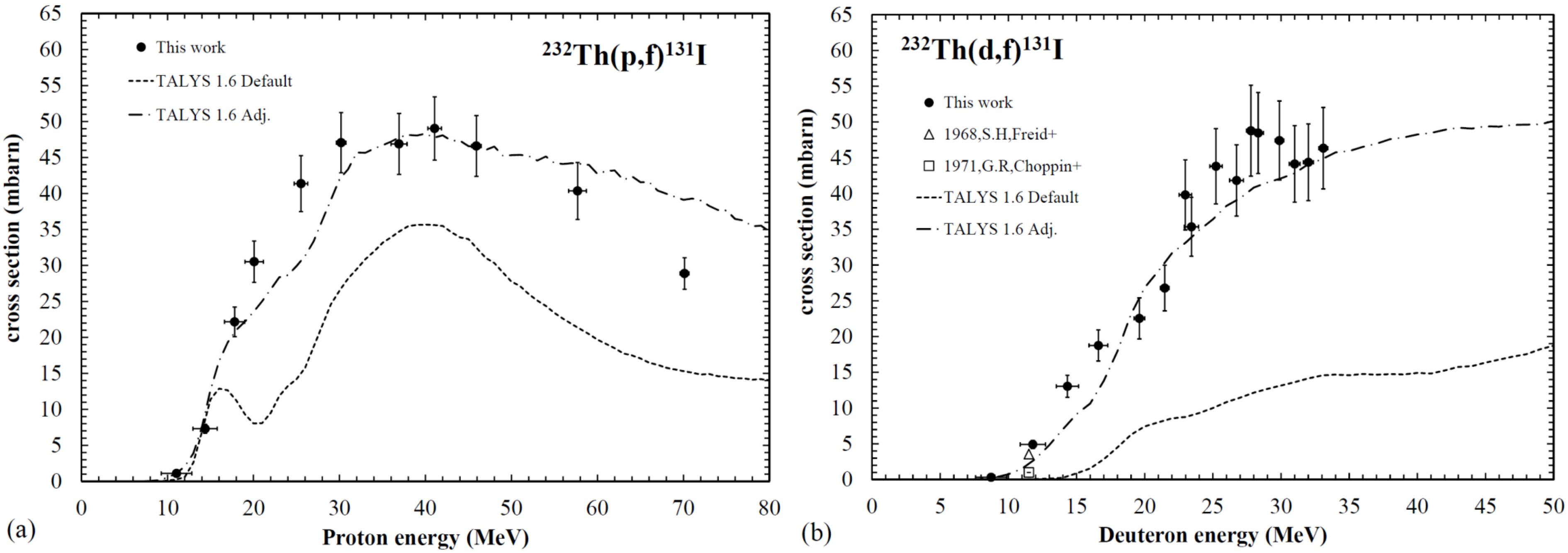


0.12 MBq/µA.h

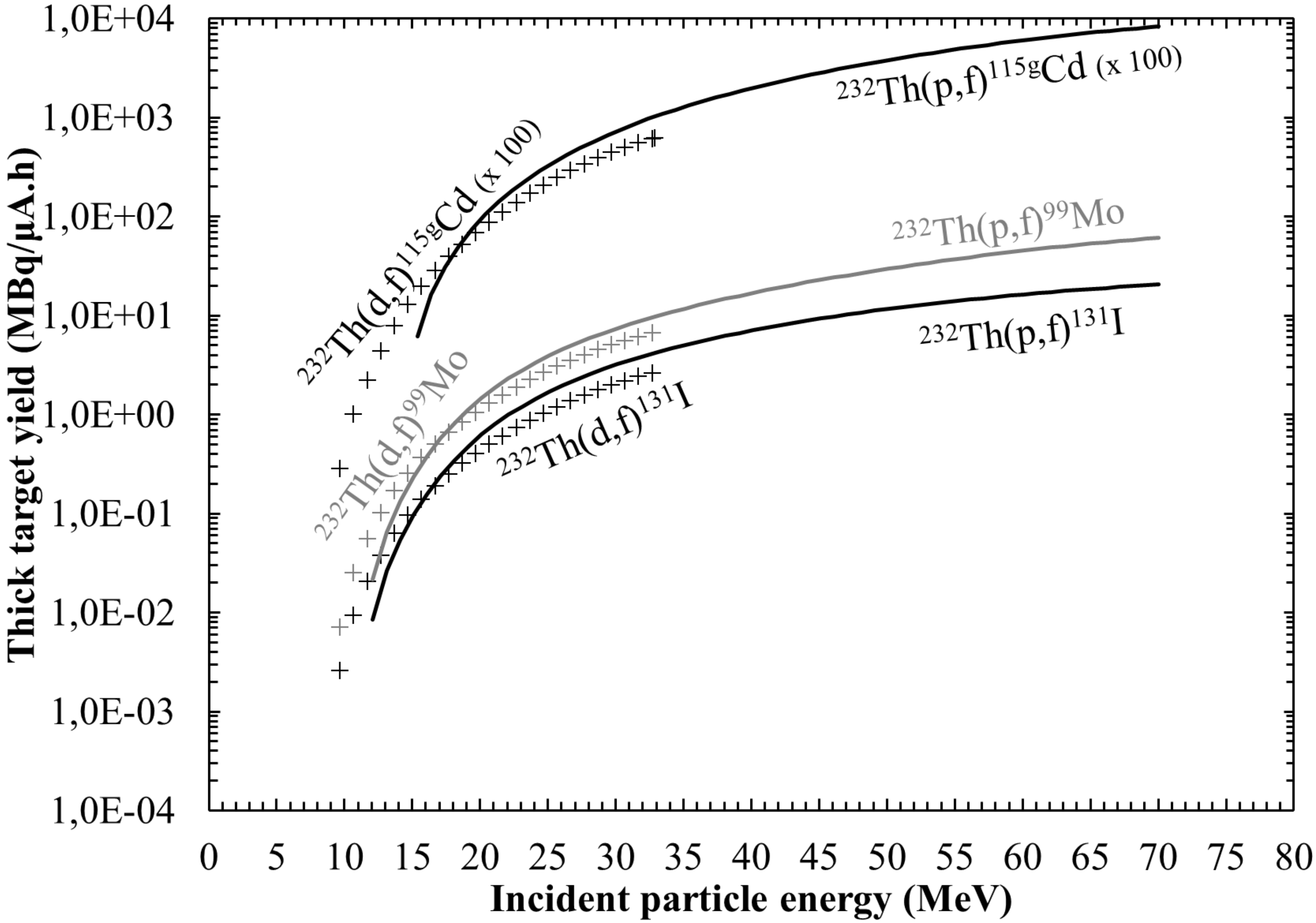


$$TTY = \Phi.\chi.\frac{N_a.\rho}{A}(1-e^{-\lambda.t})\int_{Emin}^{Emax}\frac{\sigma(E)}{\frac{dE}{dx}}dE$$

Iodine-131 production cross section



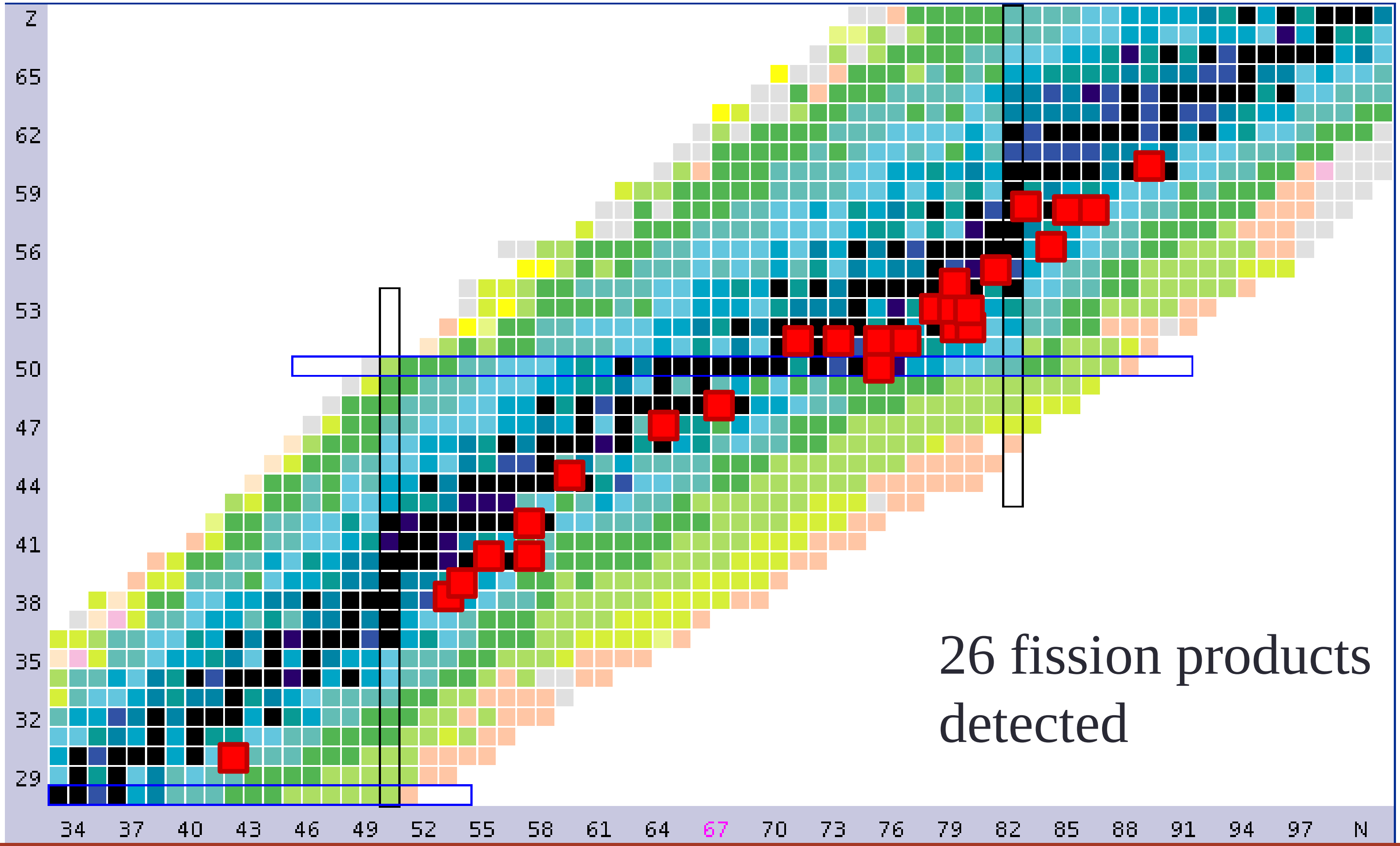
TTY Cd-115g



Thick target yield (MBq/µA.h)
1,0E+04
1,0E+03
1,0E+02
1,0E+01
1,0E+00
1,0E-01
1,0E-02
1,0E-03
1,0E-04
0 5 10 15 20 25 30 35 40 45 50 55 60 65 70 75 80
Incident particle energy (MeV)
232Th(p,f)115gCd (x 100)
232Th(d,f)115gCd (x 100)
232Th(p,f)99Mo
232Th(d,f)99Mo
232Th(p,f)131I
232Th(d,f)131I

Fission products

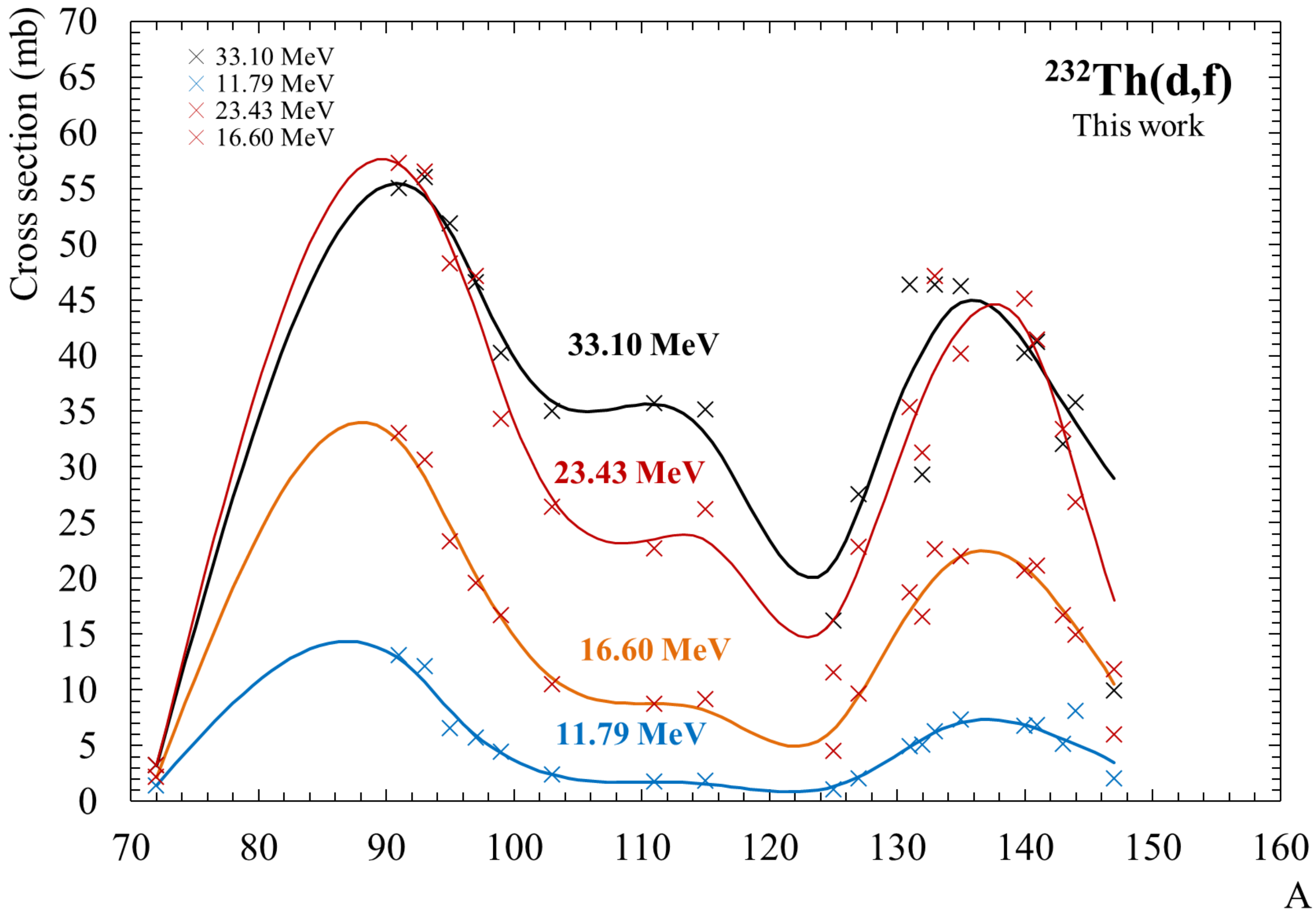
- The irradiation of a thorium target by light charged particles induced **fission products**



26 fission products detected

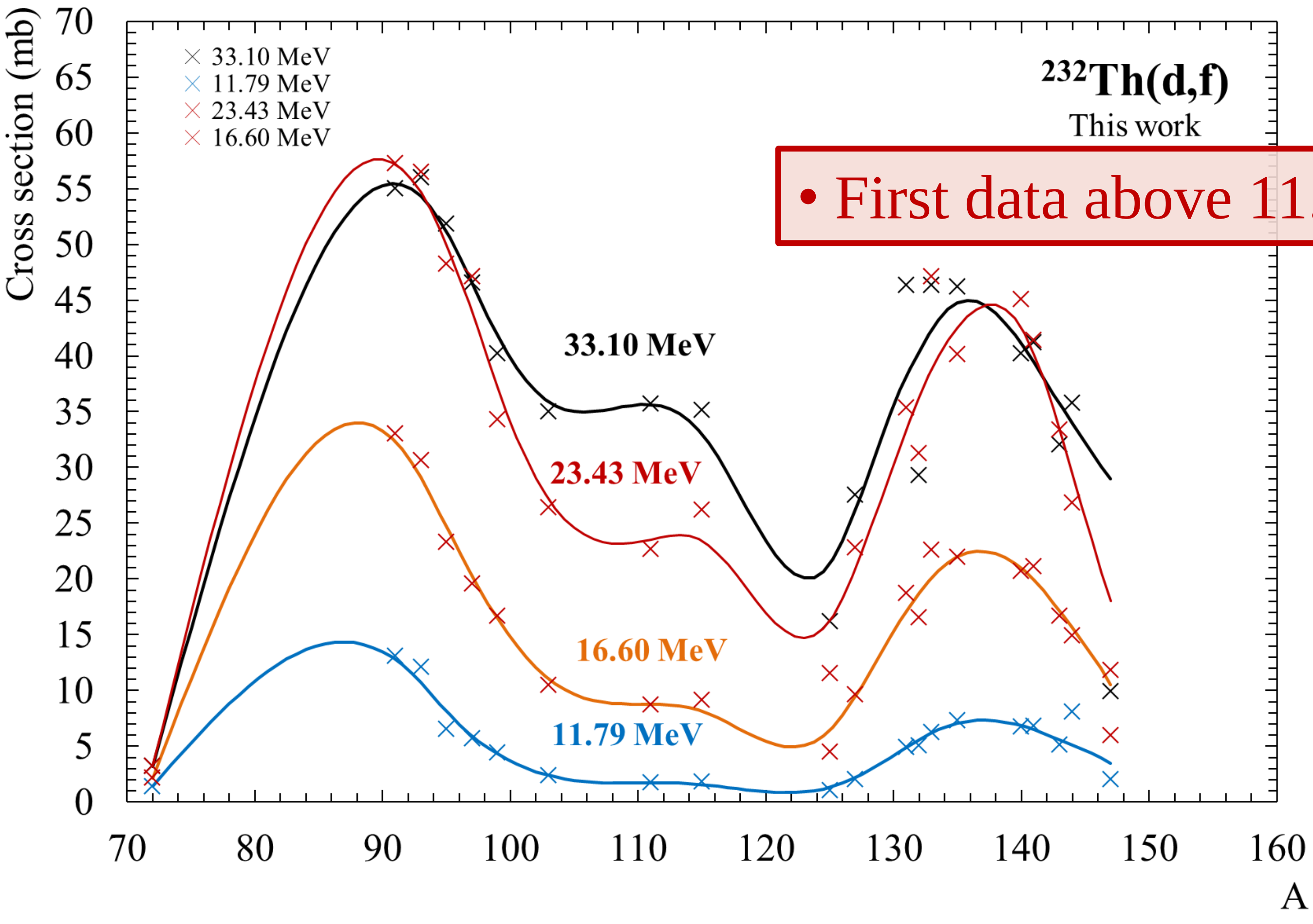
Fission products

- Exp. production cross sections determined as a function of the energy and their mass A



Fission products

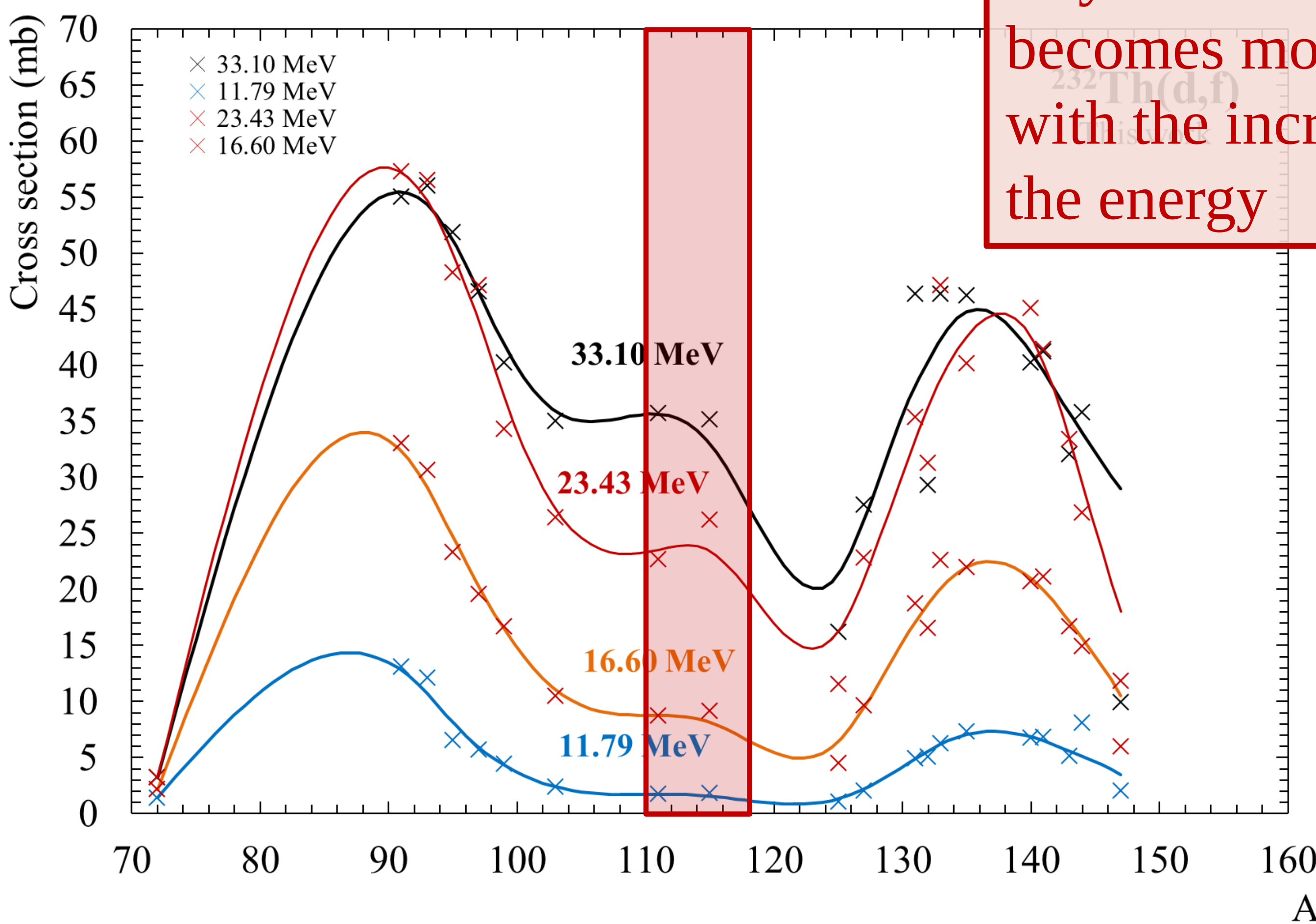
- Exp. production cross sections determined as a function of the energy and their mass A



- First data above 11.8 MeV

Fission products

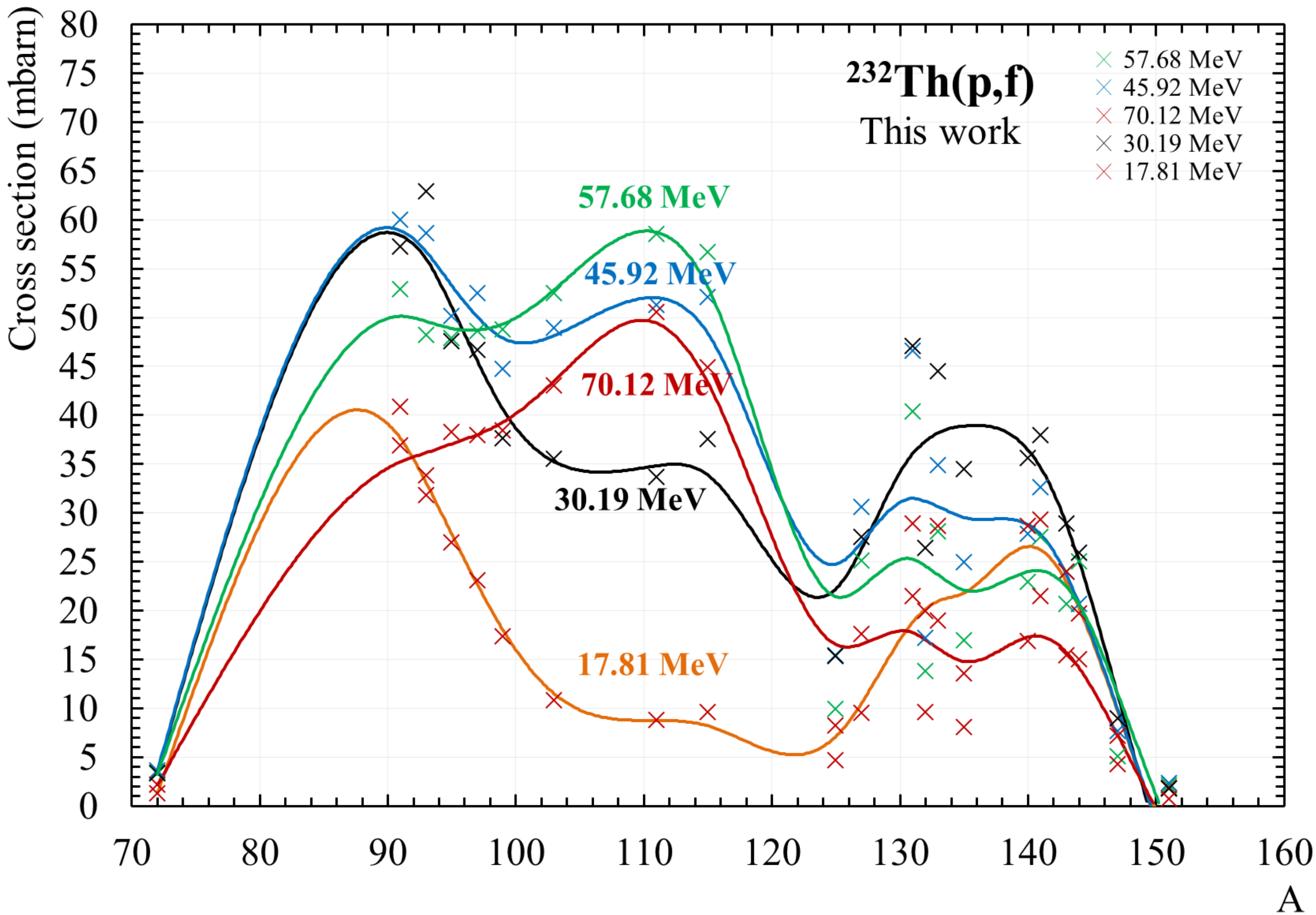
- Exp. production cross sections determined as a function of the energy and their mass A



- Symetric fission becomes more likely with the increase of the energy

Fission products

- Using **protons** : same fission products



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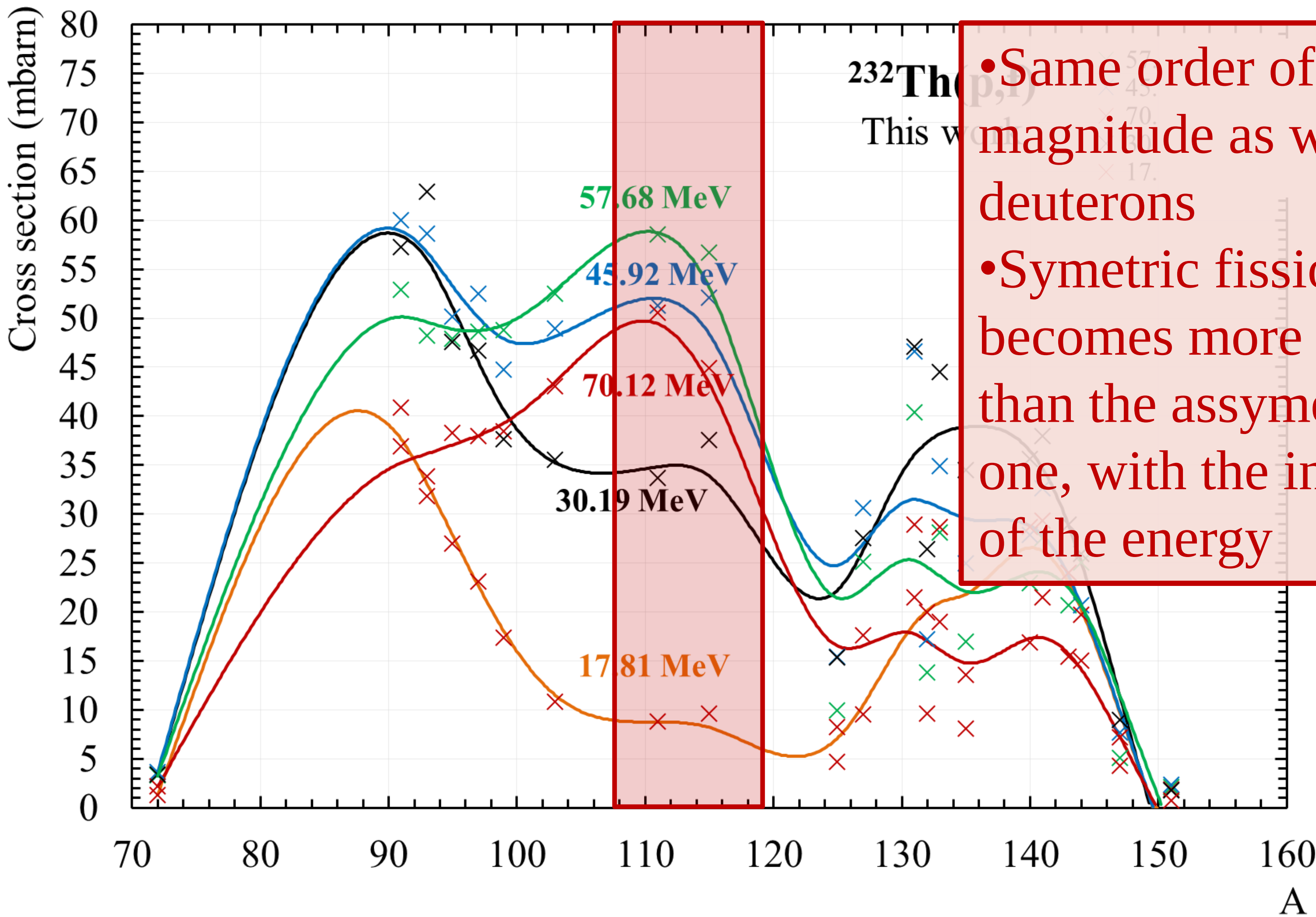

- Same order of magnitude as with deuterons
- Symetric fission becomes more likely than the assymetric one, with the increase of the energy

Fission products

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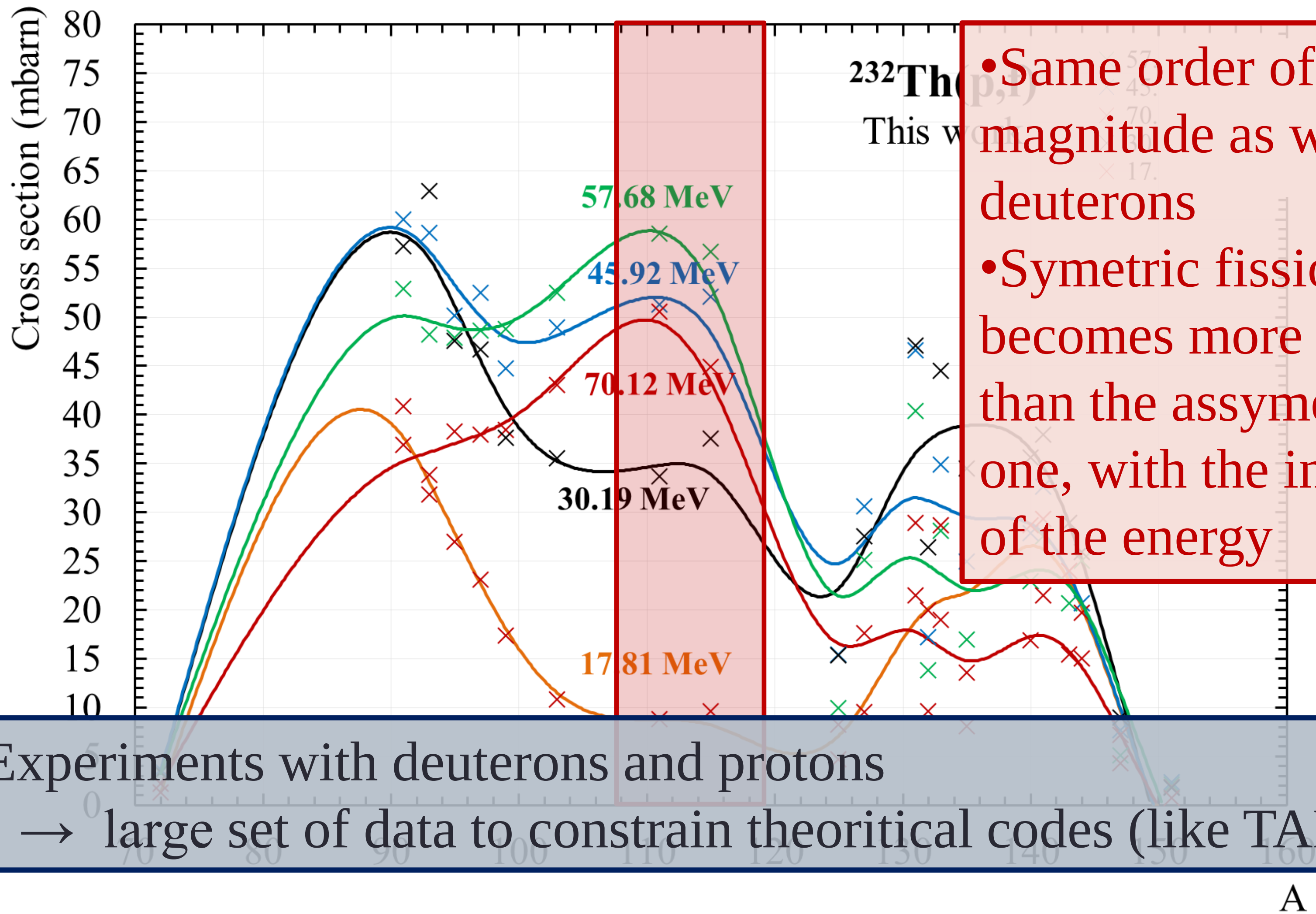

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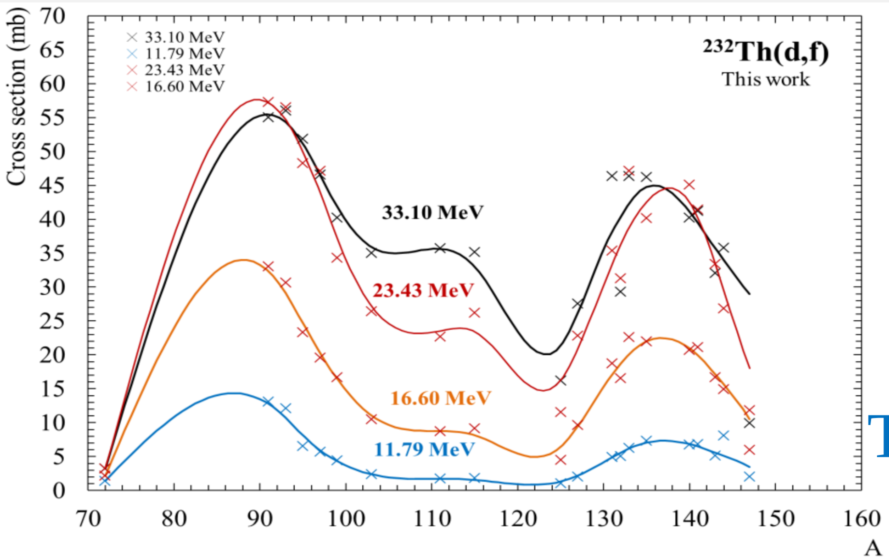
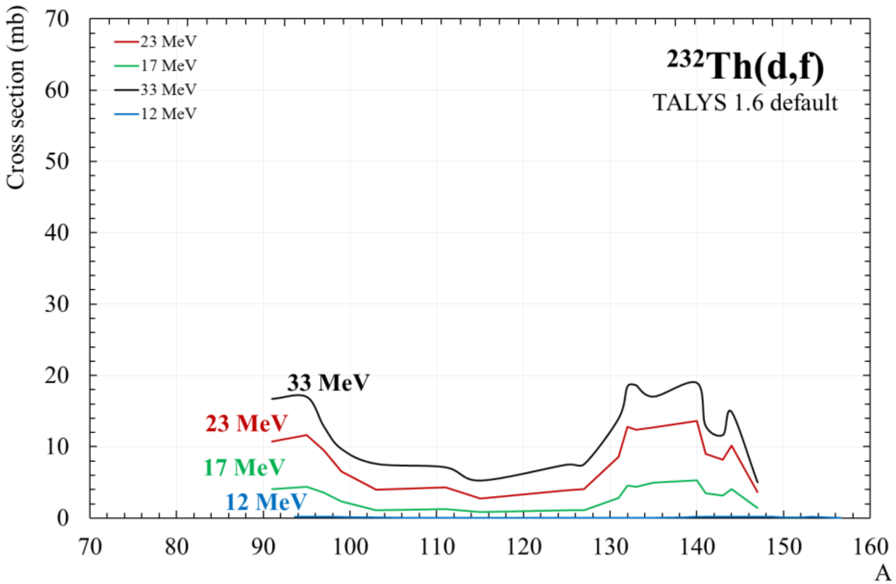
- Experiments with deuterons and protons
  → large set of data to constrain theoritical codes (like TALYS)

Fission products: the TALYS code

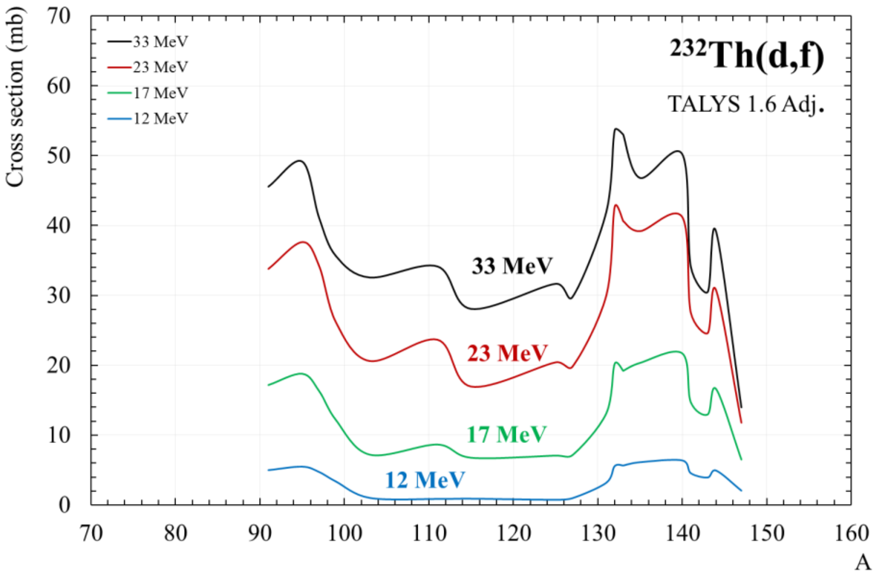
Exp. Values →



TALYS 1.6 Default ↓



TALYS 1.6 Adj. ↓



Fission products: the TALYS code