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## **How nuclear data collected for medical radionuclides production could constrain nuclear codes**

Arnaud Guertin, Charlotte Duchemin, Ferid Haddad, Etienne Nigrón, Vincent Métivier

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HAL Authorization

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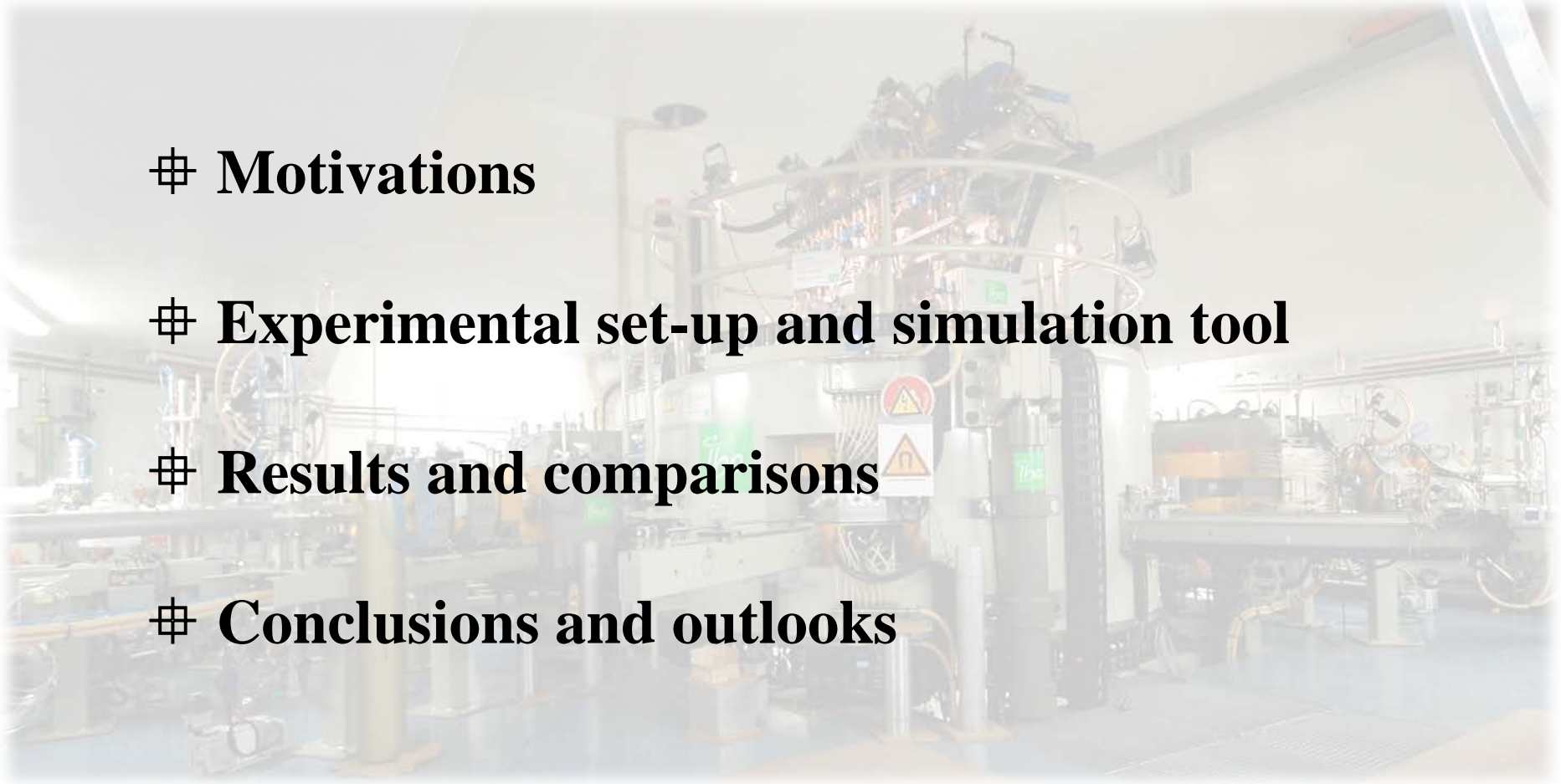
Chargé de Recherche *CNRS*



**IN2P3**

Institut national de **physique nucléaire**  
et de **physique des particules**

**How nuclear data collected for medical  
radionuclides production could constrain  
nuclear codes**

- 
- # Motivations
  - # Experimental set-up and simulation tool
  - # Results and comparisons
  - # Conclusions and outlooks

## Nuclear medicine

Many useful / potentially useful isotopes identified for applications in **nuclear medicine**

Cyclotrons and accelerators being used **in an increasing number** of countries along with reactors

- Diagnosis ( $\gamma$ ,  $\beta^+$ )
- Therapy ( $\beta^-$ ,  $\alpha$ ,  $e_{\text{Auger}}$ )

## Nuclear data and IAEA

- **Accurate and reliable** sets of data
- Well defined **production routes and decay properties**
- **Optimum production** of specific radionuclides, minimization / elimination of **impurities**, **realistic dose calculations**

**Nuclear data needs** addressed by successive:

- Experimental physicist generations
- Coordinated Research Projects initiated in the 90's

## Nuclear codes

Provide a **large set of nuclear data** in terms of targets, projectiles and energy range **to constrain and develop** predictive simulation tools of nuclear reactions

- 
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# Experimental facility

Since 2010 Subatech & ARRONAX launched a program on production of innovative radionuclides



## C70 Cyclotron build by IBA:

- 4 sectors isochron cyclotron
- 2 multi-particle sources:
  - H<sup>-</sup>, D<sup>-</sup>: multicusp
  - He<sup>2+</sup>, HH<sup>+</sup>: supernanogan ECR
- 2 extraction lines:
  - stripper or electrostatic deflector

| Extracted        | Energy (MeV) | Max. current (μA) |
|------------------|--------------|-------------------|
| H <sup>+</sup>   | 30 – 70      | 2 x 375           |
| D <sup>+</sup>   | 15 – 35      | 2 x 50            |
| He <sup>2+</sup> | 68           | 70                |
| HH <sup>+</sup>  | 17           | 50                |

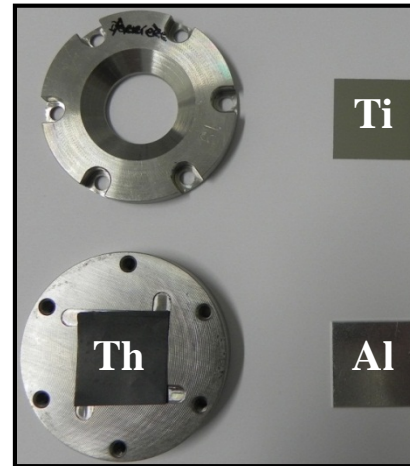
# Experimental set-up

## Stacked-foil technique:

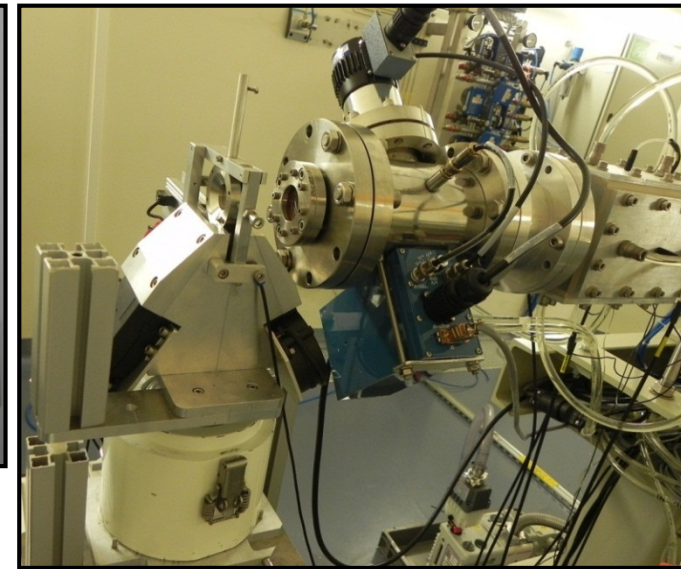
- Target/monitor/degrader **pattern**
- **Thin** foils:  
E loss small and constant
- One cross section value per foil

## Activity and cross section:

$$\sigma = \frac{\text{Act} \cdot A}{\chi \cdot \Phi \cdot \mathcal{N}_A \cdot \rho \cdot e \cdot (1 - e^{-\lambda \cdot t})}$$



Capsule and foils



Irradiation station and beam line

## Use of a Faraday cup:

- Beam dump placed at the end of the stack to control the intensity during the irradiation

## Use of a monitor foil:

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

- error on e, e':  $\leq 1\%$
- error on t: negligible

## IAEA recommended cross sections:

- 8 reactions available for protons  
 $^{27}\text{Al}$  (2),  $^{\text{nat}}\text{Ni}$ ,  $^{\text{nat}}\text{Ti}$  and  $^{\text{nat}}\text{Cu}$  (4)
- 5 reactions available for deuterons  
 $^{27}\text{Al}$  (2),  $^{\text{nat}}\text{Fe}$ ,  $^{\text{nat}}\text{Ni}$  and  $^{\text{nat}}\text{Ti}$
- 6 reactions available for alpha-particles  
 $^{27}\text{Al}$  (2),  $^{\text{nat}}\text{Ti}$  and  $^{\text{nat}}\text{Cu}$  (3)



## Proton induced reactions:

Ac-225 from Th-232(p,x)

Ra-223 from Th-232(p,x)

Fission fragment distribution from Th-232(p,x)\*

Monitor reactions on Ti, Ni and Cu

C. Duchemin et al, Phys Med Biol **60** (2015) **931-946**

C. Duchemin et al, Phys Med Biol **60** (2015) **931-946**

To be published

E. Garrido et al., Nucl Instr Meth Phys Res B **383** (2016) **191-212**

## Deuteron induced reactions:

Sc-44 New data set for Ca-44(d,x)

Tb-155 New data set for Gd-nat(d,x)

Re-186g New data set for W-186(d,x)

Th-226 New data set for Th-232(d,x)

Fission fragment distribution from Th-232(d,x)\*

Monitor reactions on Ti

C. Duchemin et al, Phys Med Biol **60** (2015) **6847-6864**

Submitted to Appl Radiat Isot

C. Duchemin et al, Appl Radiat Isot **97** (2015) **52-58**

C. Duchemin et al, Appl Radiat Isot **103** (2015) **160-165**

## Alpha induced reactions:

Sn-117m from Cd-116( $\alpha$ ,x)

Monitor reactions on Cu, Ti, Ni

C. Duchemin et al, Appl Radiat Isot **115** (2016) **113-124**

\* S415, Ambassadeur, 15/09/16, 15:45 and P120, Witte Roos 8:30-18:10

V. Métyvier et al., Thorium-232 fission induced by light charged particles up to 70 MeV

Code for the simulation of nuclear reactions

Projectiles : n, p, d, t, He-3,  $\alpha$  particles

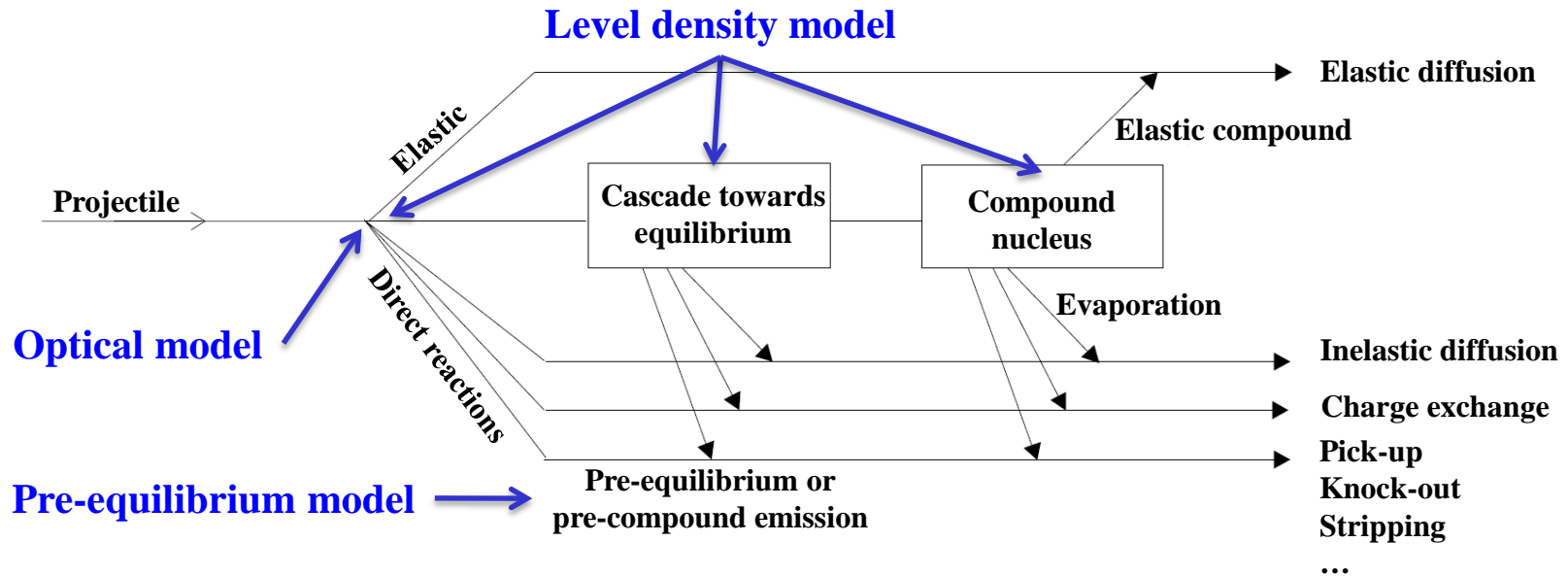
Many state-of-the-art nuclear models

Energy : 1 keV to 1 GeV

Provide a complete description of all reactions channels and observables

Targets :  $Z = 3$  to 110

## Nuclear reactions



Adapted from S. Benck, PhD thesis, 1999

⇒ Influence on the calculated production cross section values

## TENDL2015

Nuclear data library based on both default and adjusted TALYS calculations and data from other sources



By A.J. Koning<sup>1</sup>, D. Rochman<sup>2</sup>, J. Kopecky<sup>3</sup>, J.Ch. Sublet<sup>4</sup>, M. Fleming<sup>4</sup>, E. Bauge<sup>7</sup>, S. Hilaire<sup>7</sup>, P. Romain<sup>7</sup>, B. Morillon<sup>7</sup>, H. Duarte<sup>7</sup>, S.C van der Marck<sup>6</sup>, S. Pomp<sup>5</sup>, H. Sjostrand<sup>5</sup>, R. Forrest<sup>1</sup>, H. Henriksson<sup>8</sup>, O. Cabellos<sup>9</sup>, S. Goriely<sup>10</sup>, J. Leppanen<sup>11</sup>, H. Leeb<sup>12</sup>, A. Plompen<sup>13</sup>, and R. Mills<sup>14</sup>

<sup>1</sup> IAEA, <sup>2</sup> PSI, <sup>3</sup> JUKO Research, <sup>4</sup> CCFE, <sup>5</sup> Uppsala Univ., <sup>6</sup> NRG, <sup>7</sup> CEA, <sup>8</sup> Vattenfall, <sup>9</sup> NEA, <sup>10</sup> ULB, <sup>11</sup> VTT, <sup>12</sup> ATI, <sup>13</sup> IRMM, <sup>14</sup> NNL.

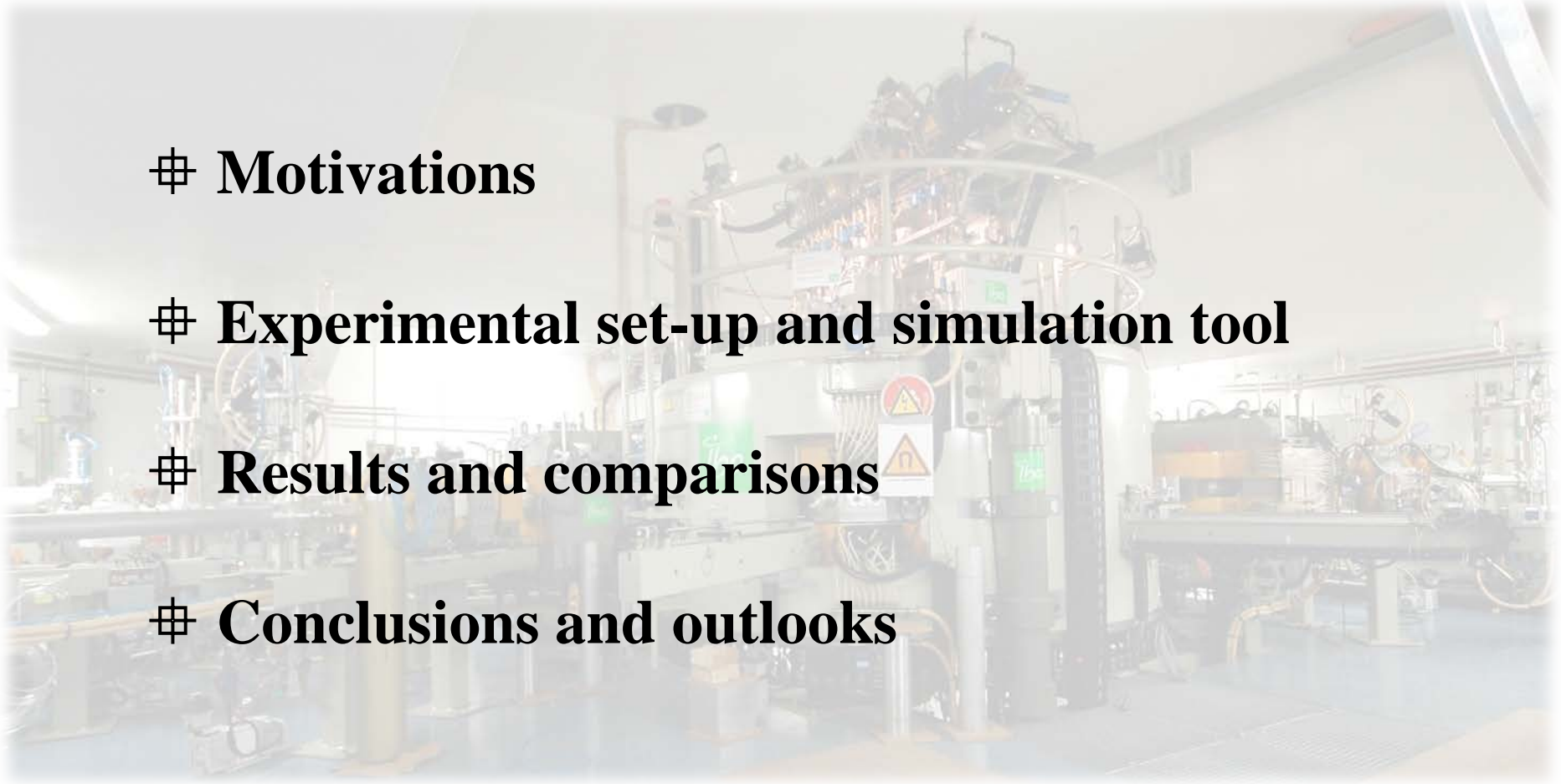
TENDL2015 contains evaluations for :

seven types of incident particles (n, p, d, t, He-3, alpha-particle, gamma ray)

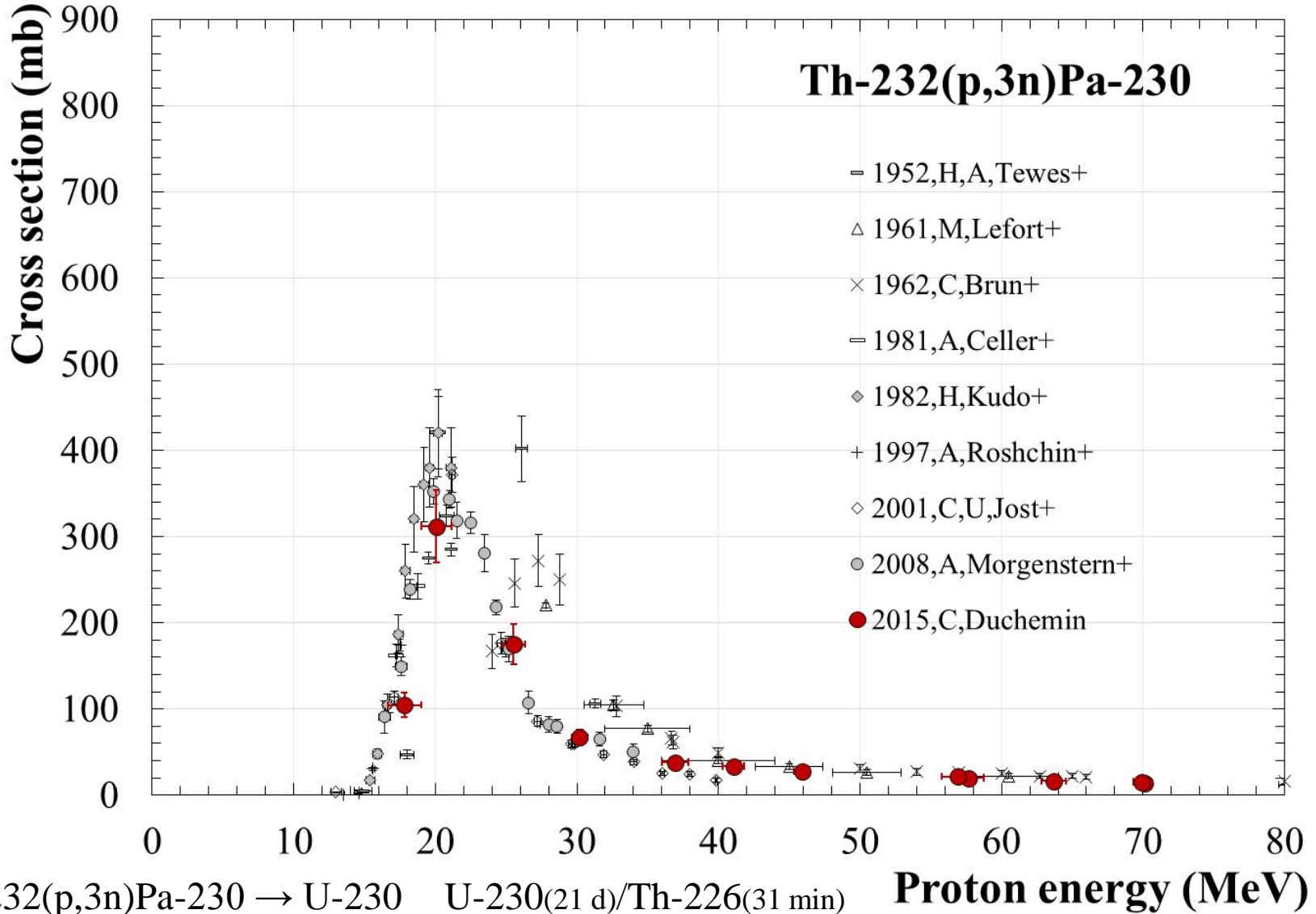
all isotopes living more than 1 second (~ 2800 isotopes)

all files are original except 15 (natural carbon from JENDL-4.0, <sup>1,2,3</sup>H, <sup>2,3</sup>He, <sup>6,7</sup>Li, <sup>10,11</sup>B, <sup>9</sup>Be, <sup>14,15</sup>N, <sup>16</sup>O and <sup>19</sup>F from ENDF/B-VII.1)

Koning A.J. et al., [https://tendl.web.psi.ch/tendl\\_2015/tendl2015.html](https://tendl.web.psi.ch/tendl_2015/tendl2015.html)  
 Koning A.J. and Rochman D., Nucl. Data Sheets, 113, 2012

- 
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# Novel therapeutic nuclide

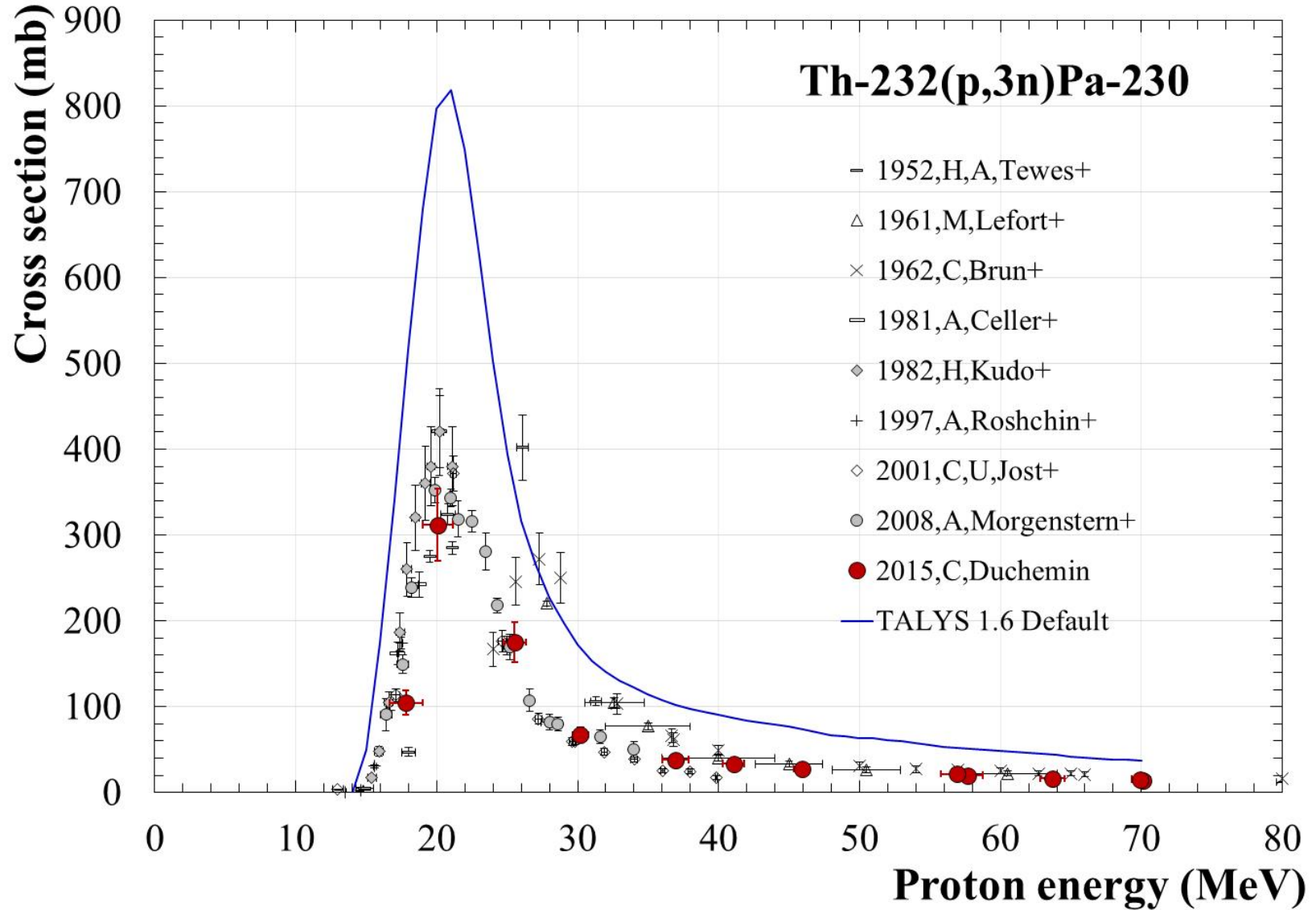


α RIT for leukaemia treatment

4 α cascade of 27.7 MeV

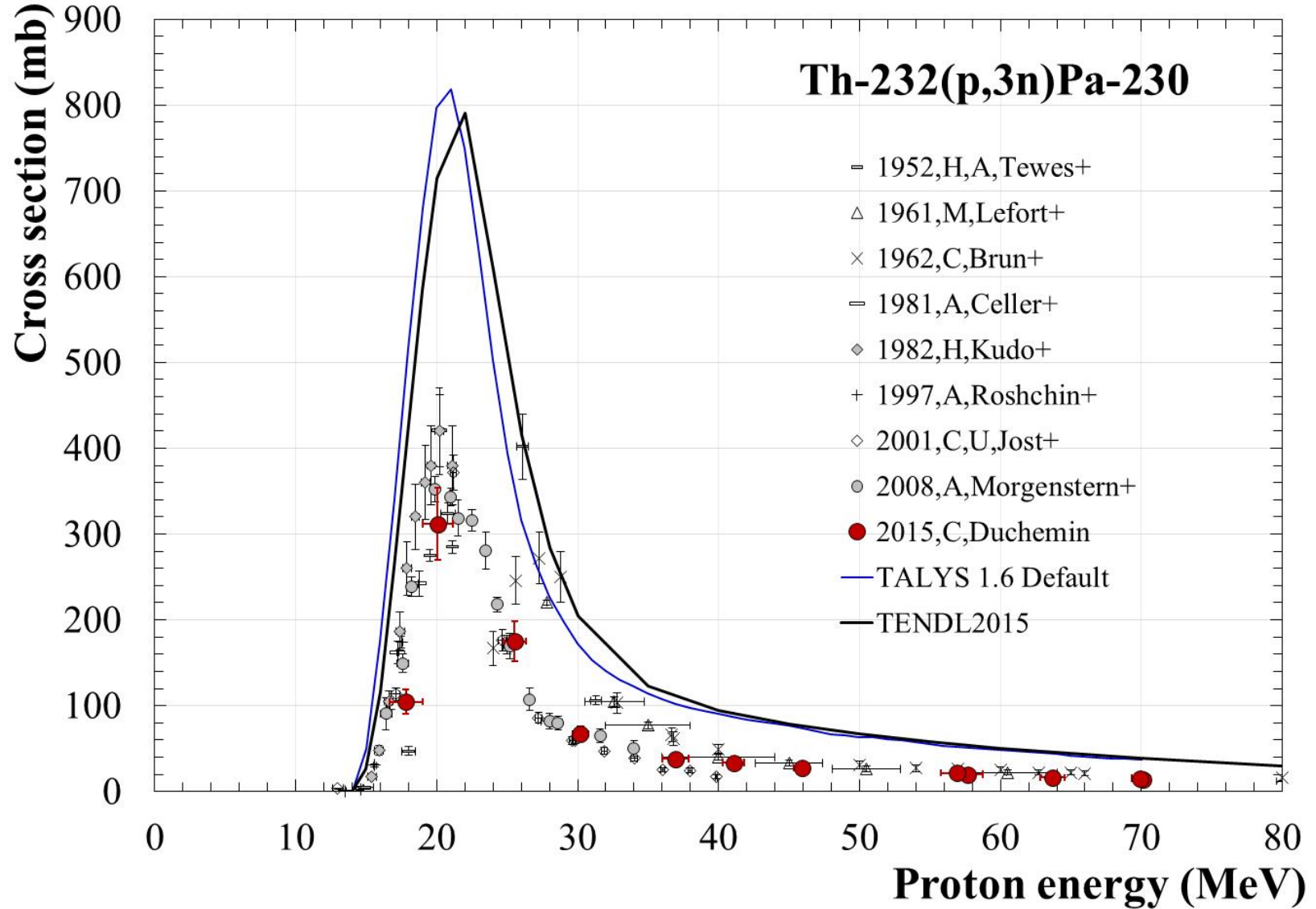
*C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946*

# Novel therapeutic nuclide



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# Novel therapeutic nuclide



*C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946*

## TALYS default and adjusted calculations

TALYS code version 1.6

the combination of models that best describes the whole set of available data for all projectiles, targets and incident energies defined by the TALYS authors

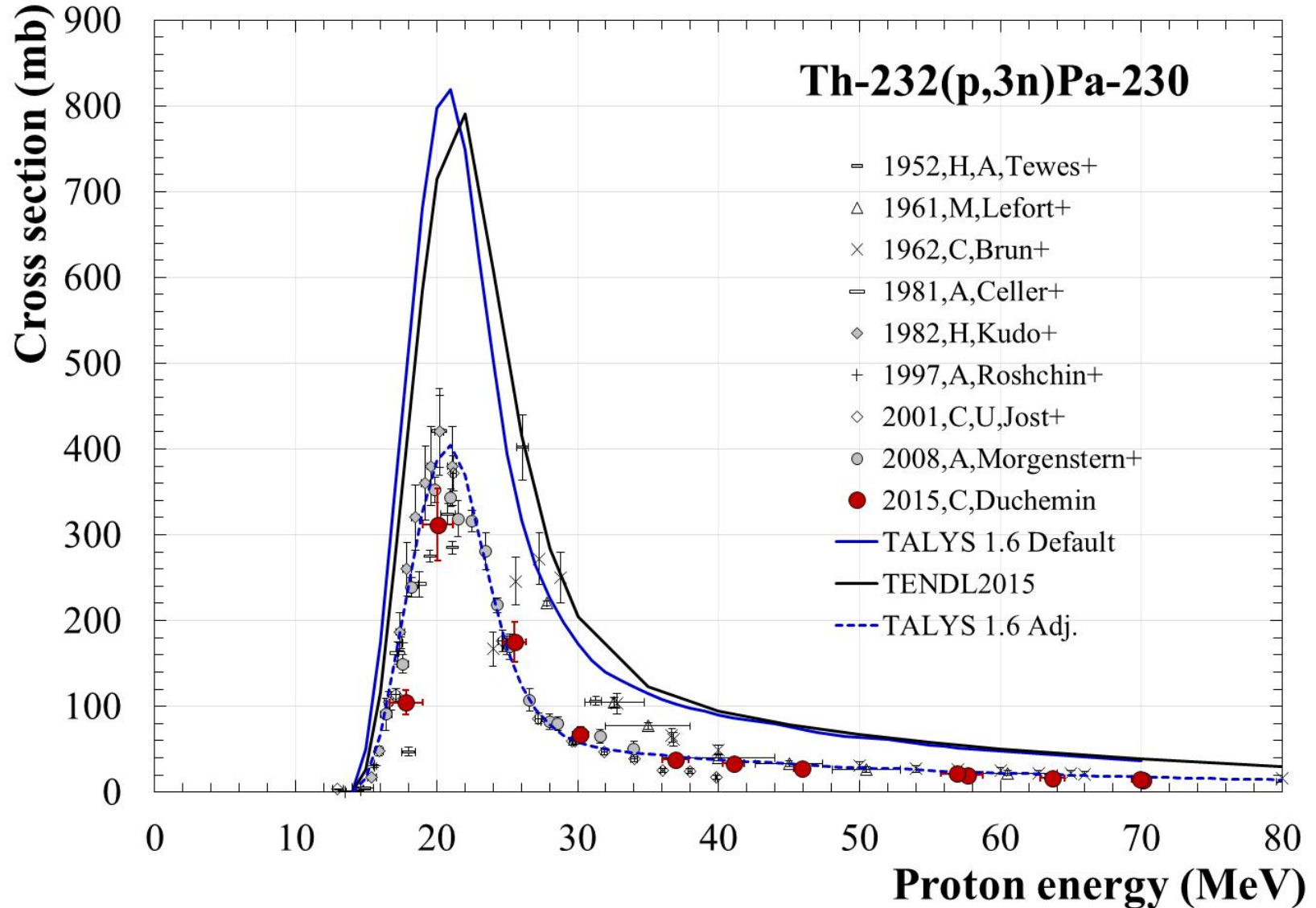
⇒ TALYS 1.6 Default

One combination of models that best describes our whole set of data for proton, deuteron, alpha particles as projectile (and literature data) has been defined by C. Duchemin et al.

⇒ TALYS 1.6 Adj.

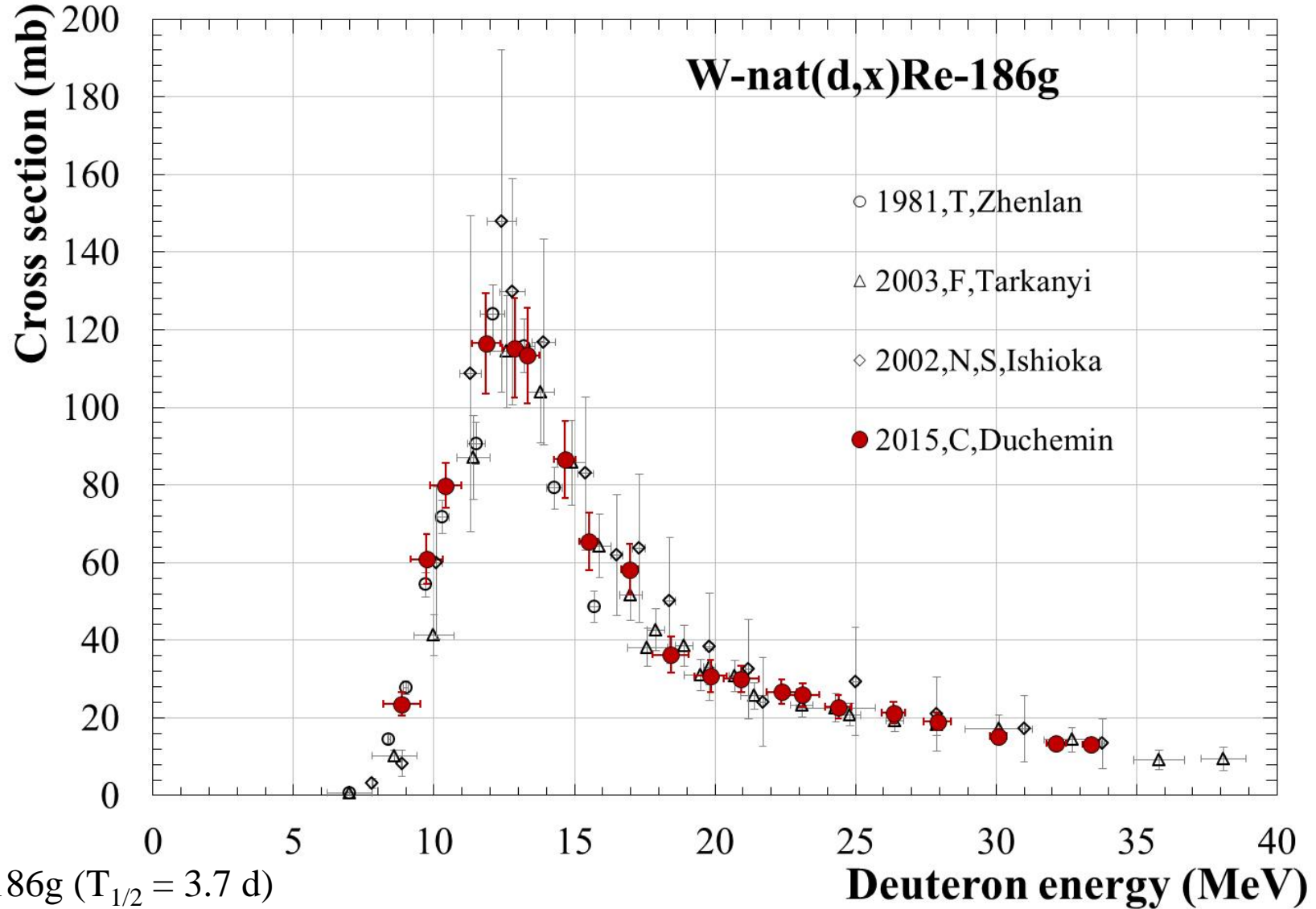
| Models          | Projectile   | Default  | Adj.   |
|-----------------|--------------|--|--|
| Optical         | p (1)        | A.J. Koning and J.P. Delaroche (2003)                                  | A.J. Koning and J.P. Delaroche (2003)  |
|                 | d (5)        | S. Watanabe (1958)<br>...  | Y. Han et al. (2006)<br>...  |
|                 | $\alpha$ (5) | L. McFadden and G.R. Satchler (1966)<br>...                            | Demetriou et al. (2002)<br>...   |
| Pre-equilibrium | All (4)      | 2 components exciton<br>A.J. Koning and M.C. Duijvestijn (2004)<br>... | Exciton model including numerical transition rates for collision probabilities<br>A.J. Koning and M.C. Duijvestijn (2004)<br>... |
| Level density   | All (6)      | Fermi gas<br>A.J. Koning et al. (2008)<br>...                          | Hilaire's combinatorial tables<br>Goriely et al. (2008)<br>...   |

# Novel therapeutic nuclide



C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946

# Novel therapeutic nuclide

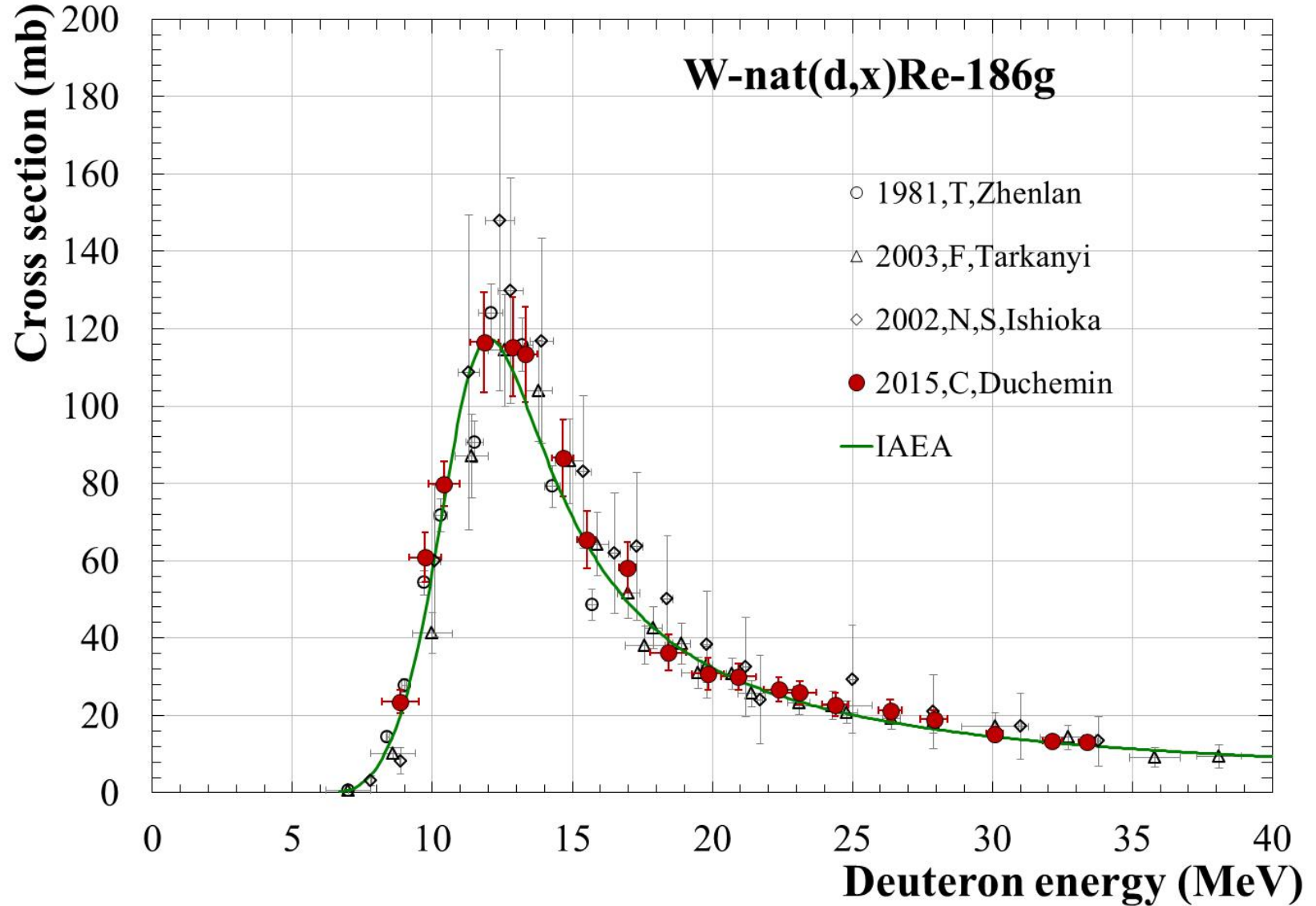


Re-186g ( $T_{1/2} = 3.7$  d)

$\beta^-$  emitter used in clinical trials for the palliation of painful  
 bone metastases resulting from prostate and breast cancer

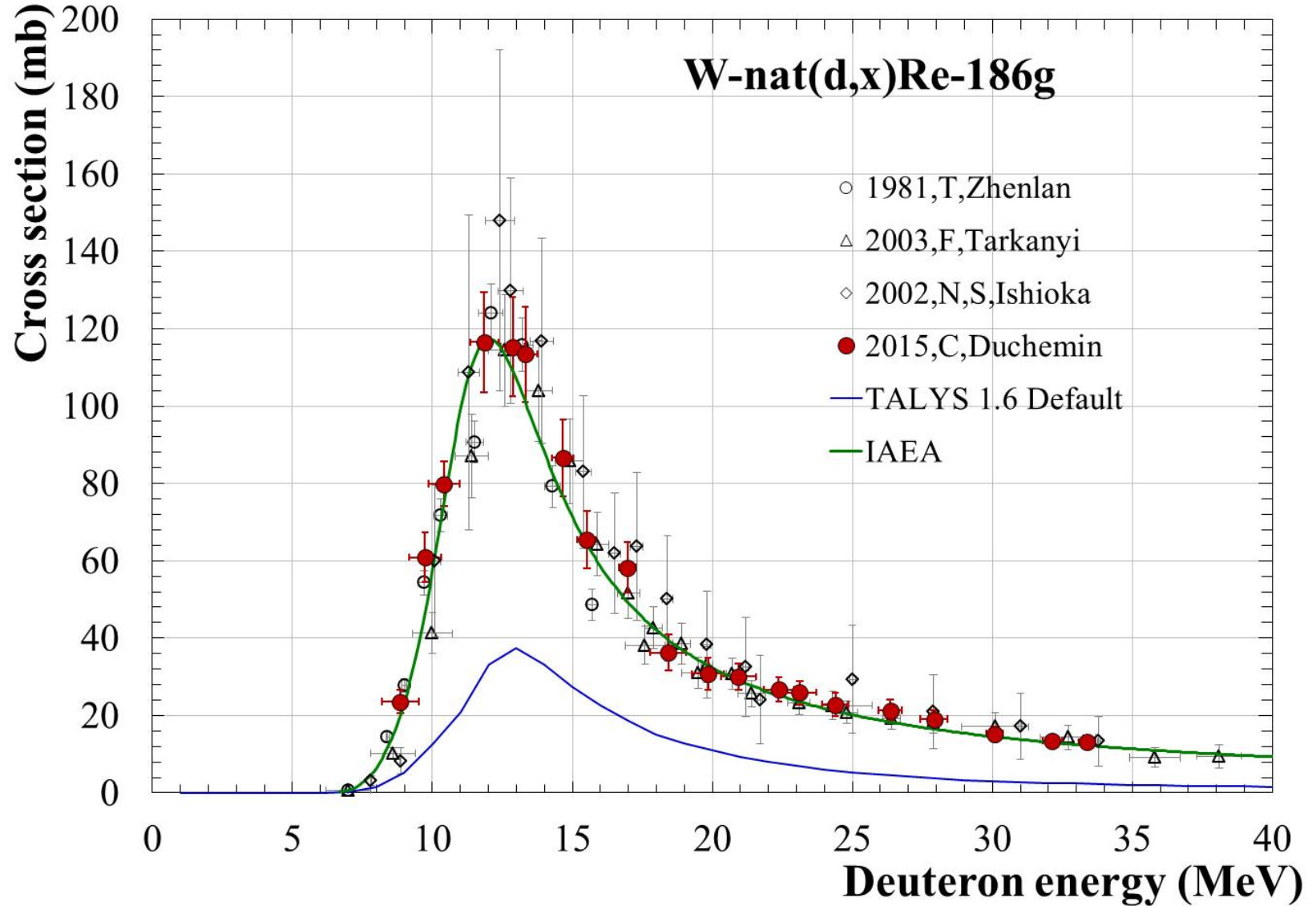
*C. Duchemin et al., Appl. Radiat. Isot. 97 (2015) 52-58*

# Novel therapeutic nuclide

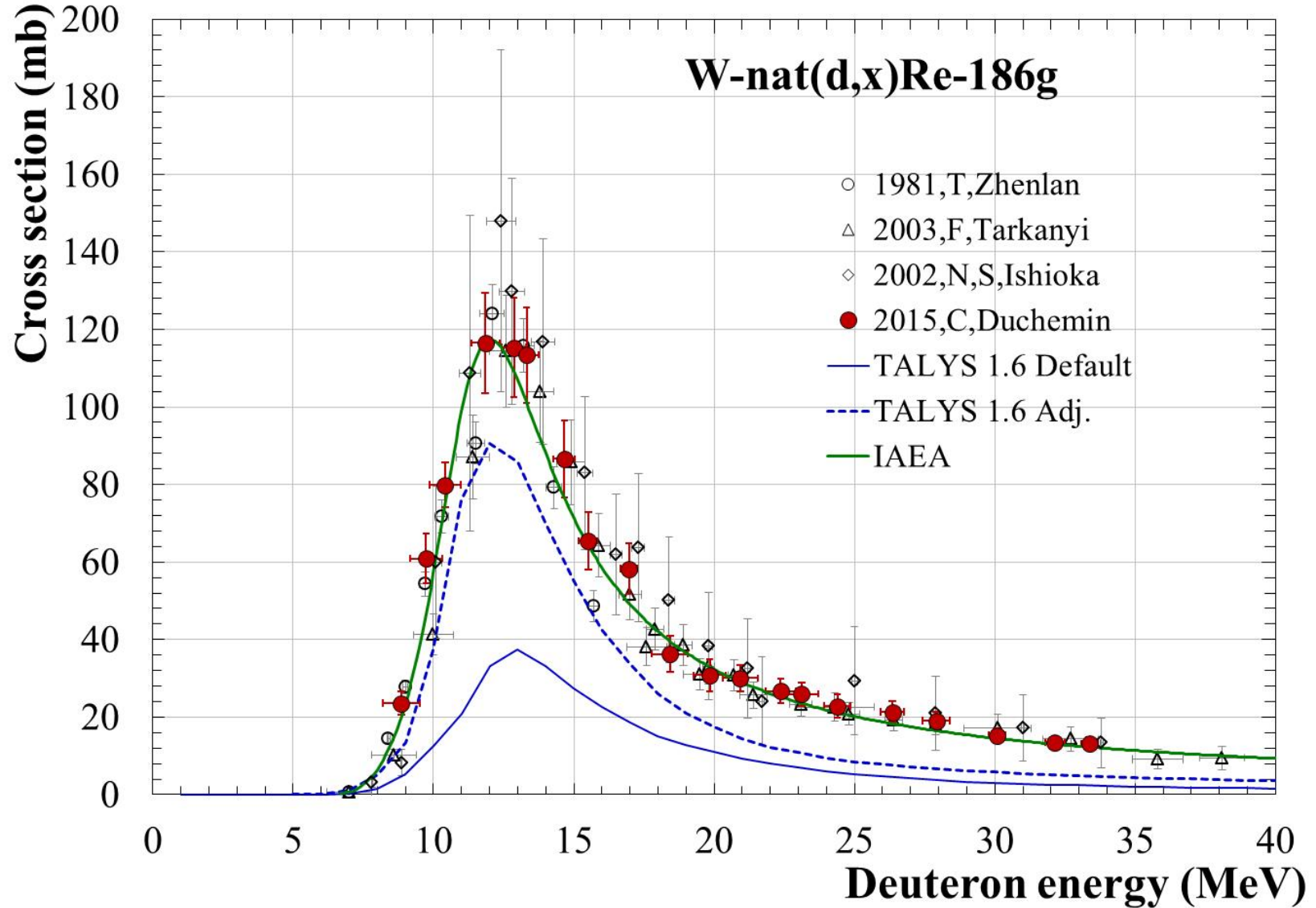


C. Duchemin et al., *Appl. Radiat. Isot.* 97 (2015) 52-58

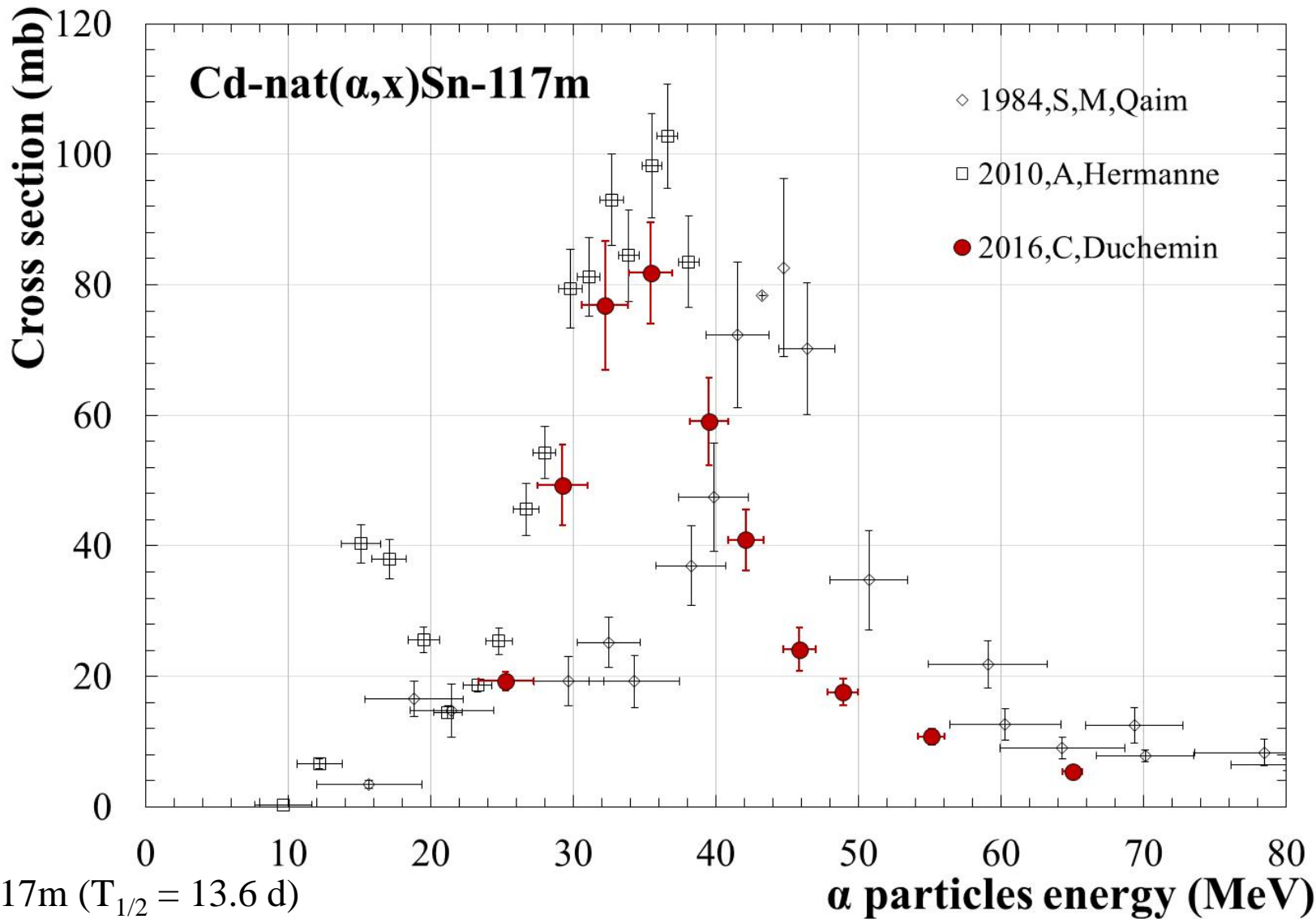
# Novel therapeutic nuclide



# Novel therapeutic nuclide



# Novel therapeutic and imaging nuclide



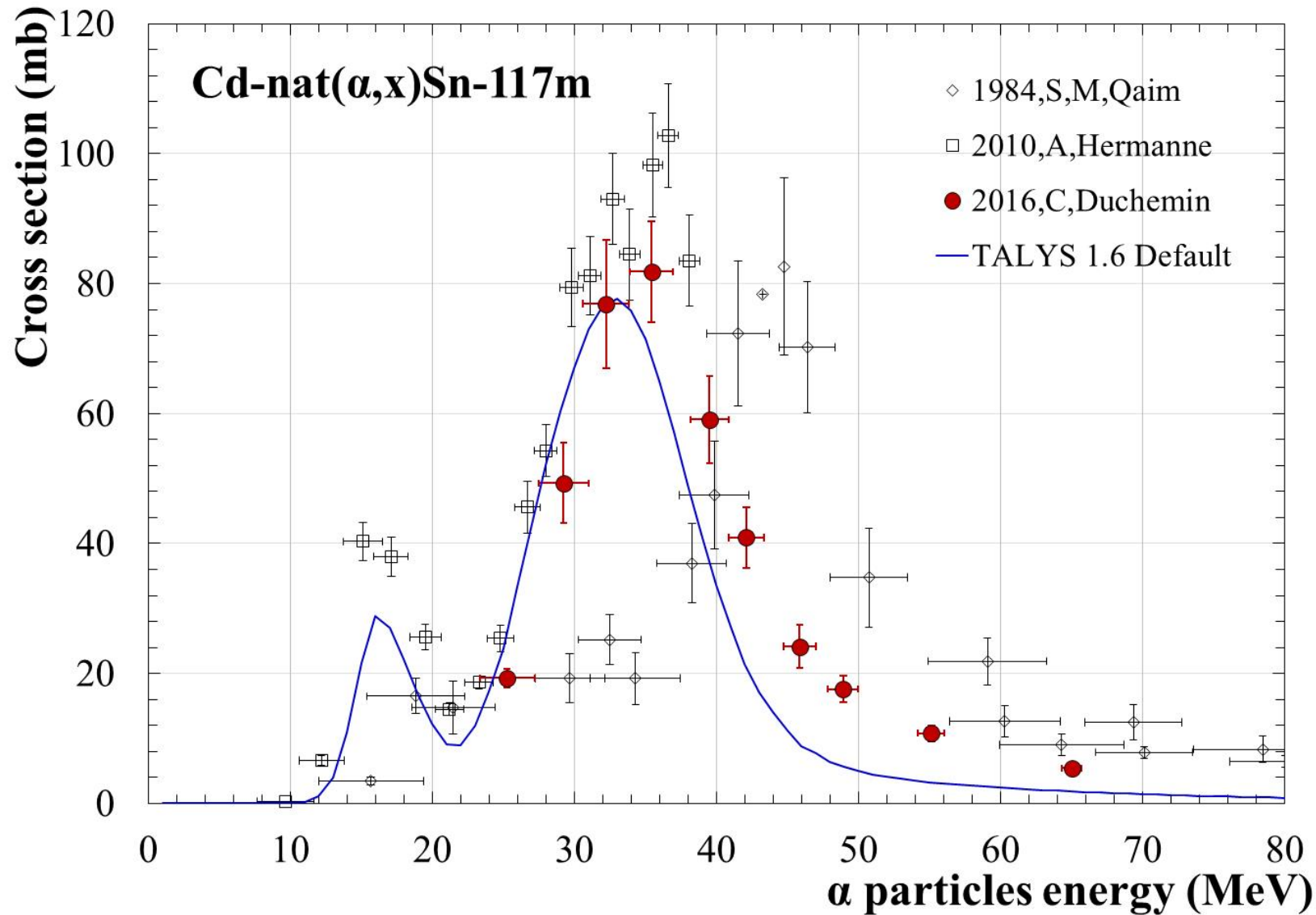
Sn-117m ( $T_{1/2} = 13.6$  d)

Conversion e- emitter used for the palliation of painful bone metastases

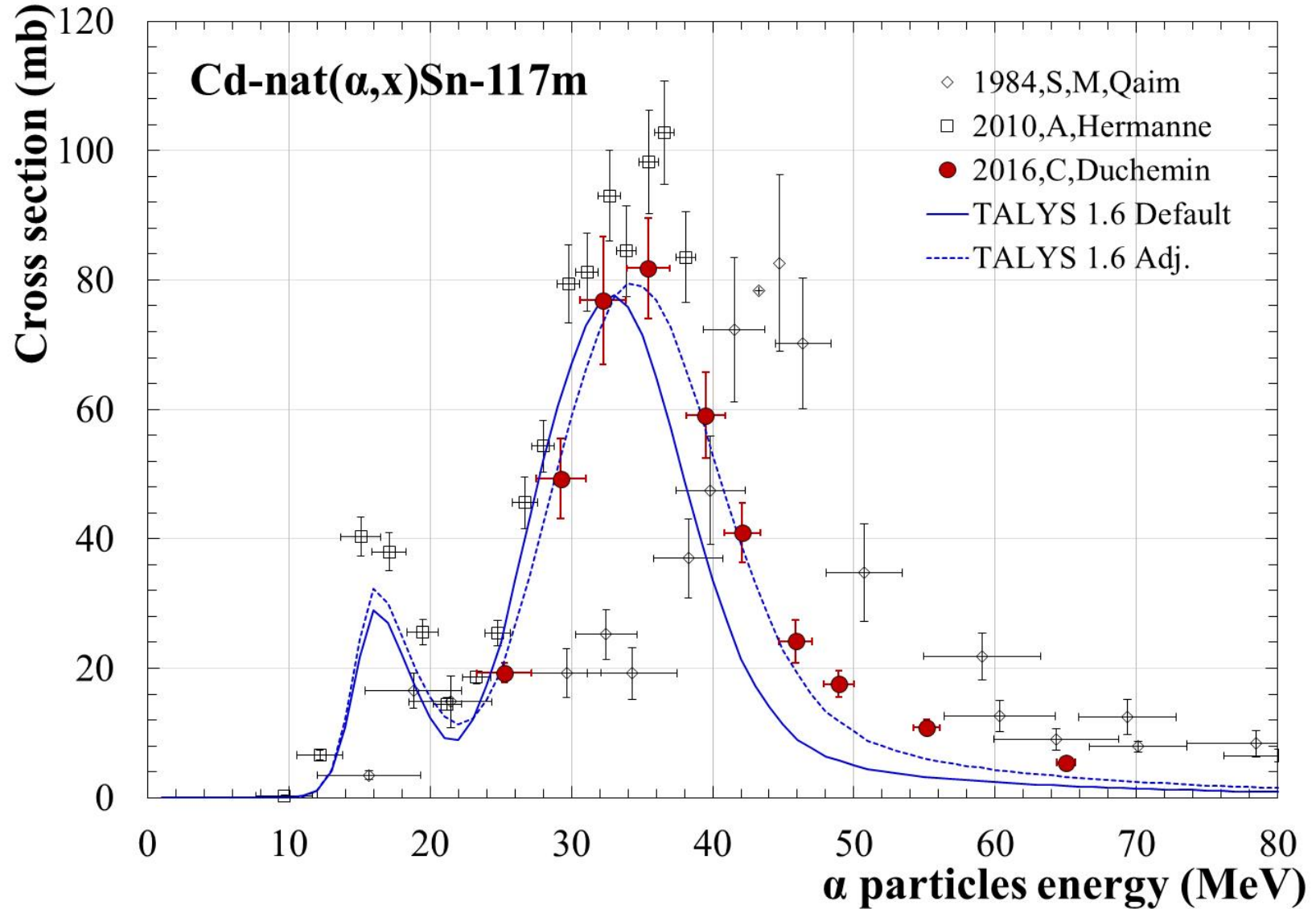
158 keV gamma ray suitable for SPECT imaging

*C. Duchemin et al., Appl. Radiat. Isot. 115 (2016) 113-124*

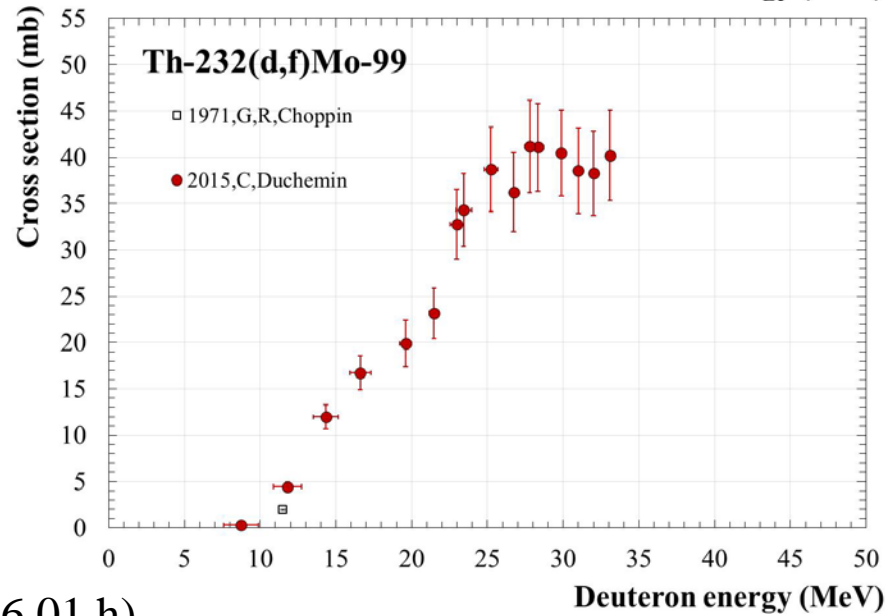
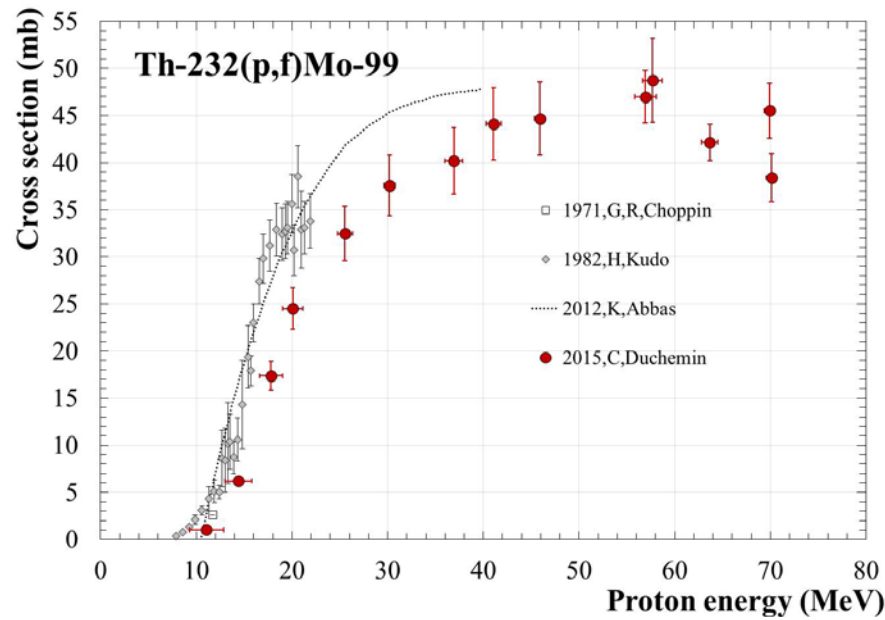
# Novel therapeutic and imaging nuclide



# Novel therapeutic and imaging nuclide



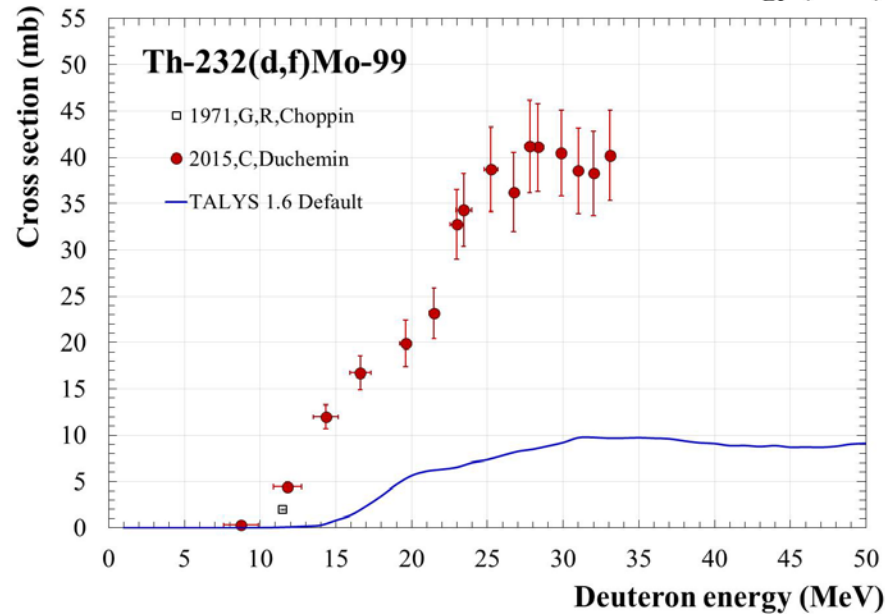
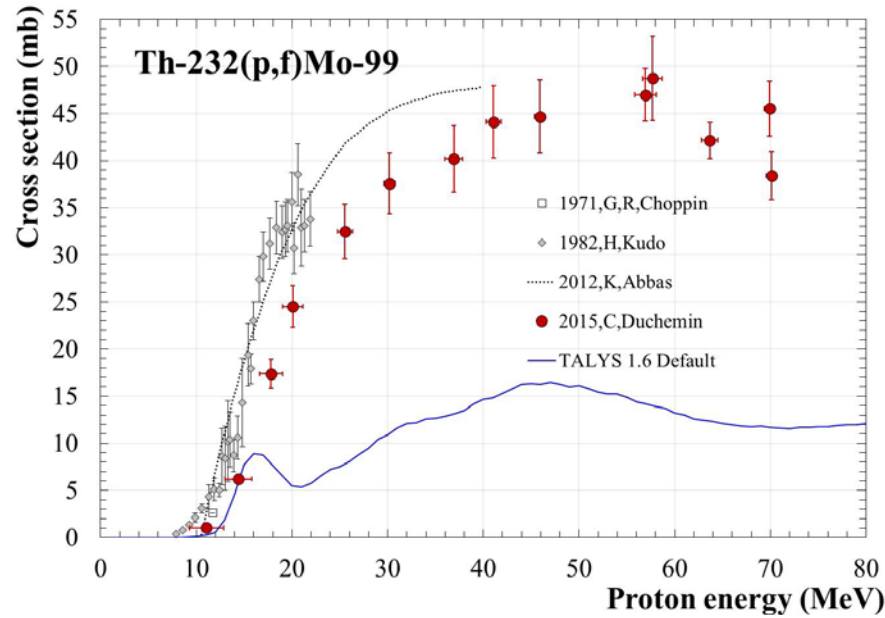
# Potential method for production of $^{99}\text{Mo}/^{99m}\text{Tc}$



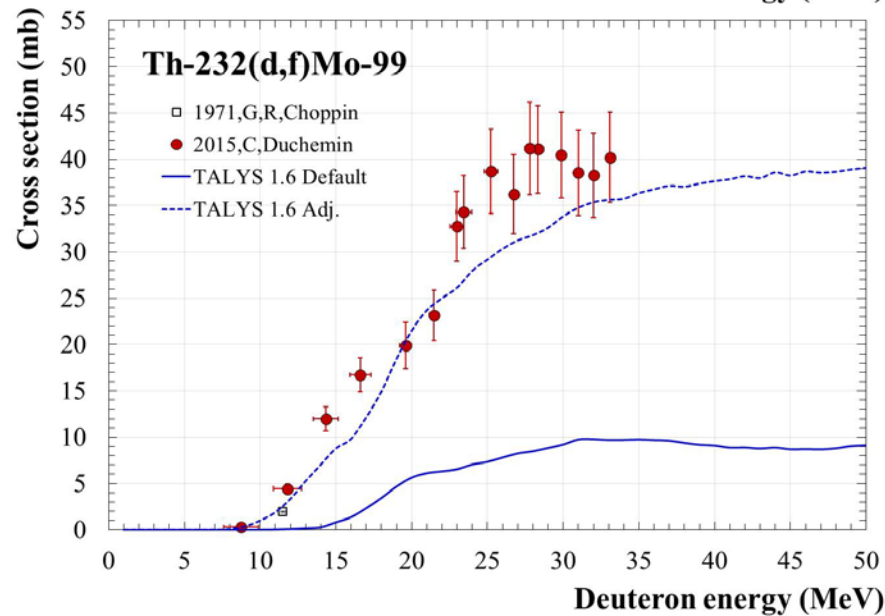
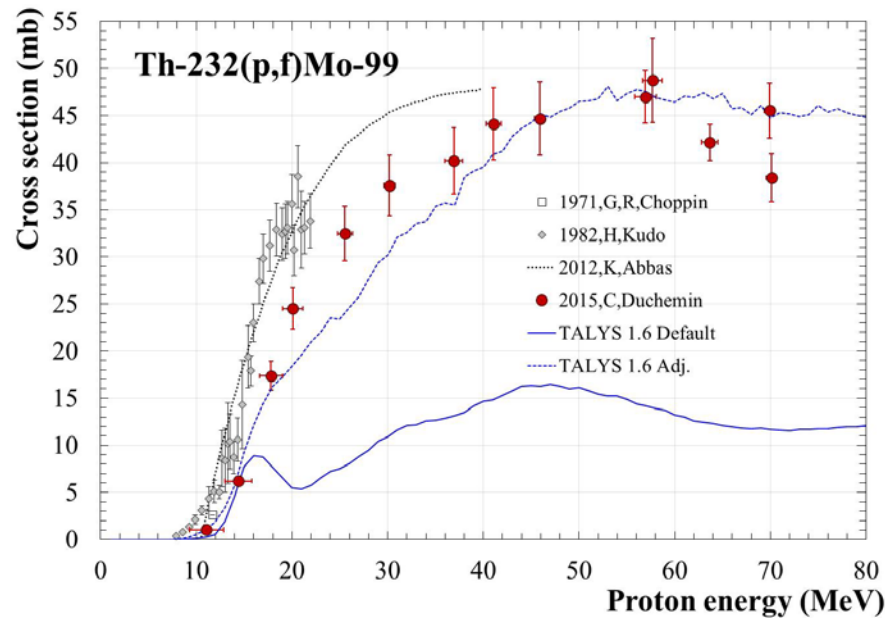
Mo-99(65.94 d)/Tc-99m(6.01 h)

*C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946*

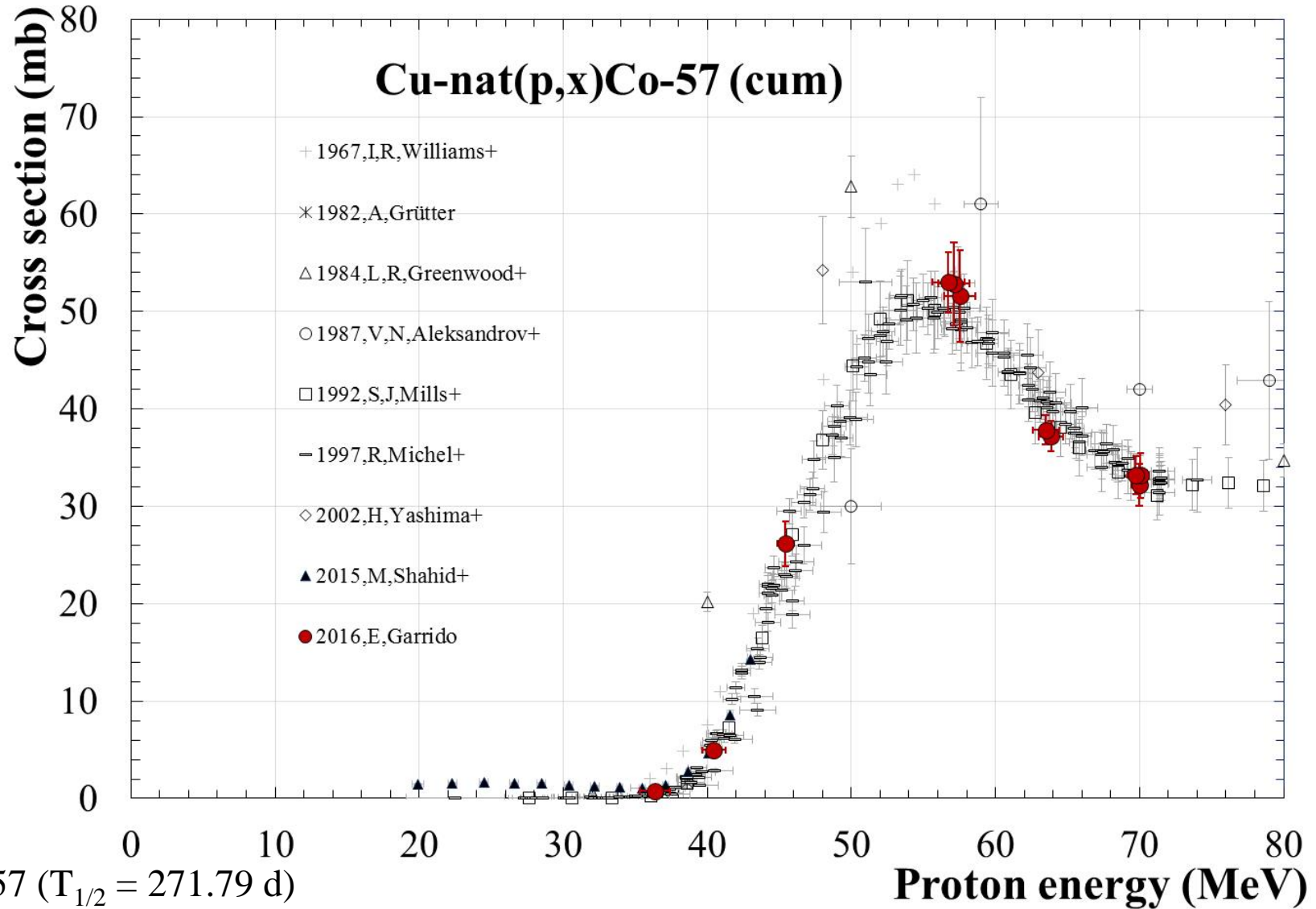
# Potential method for production of $^{99}\text{Mo}/^{99m}\text{Tc}$



*C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946*



# Monitor reaction



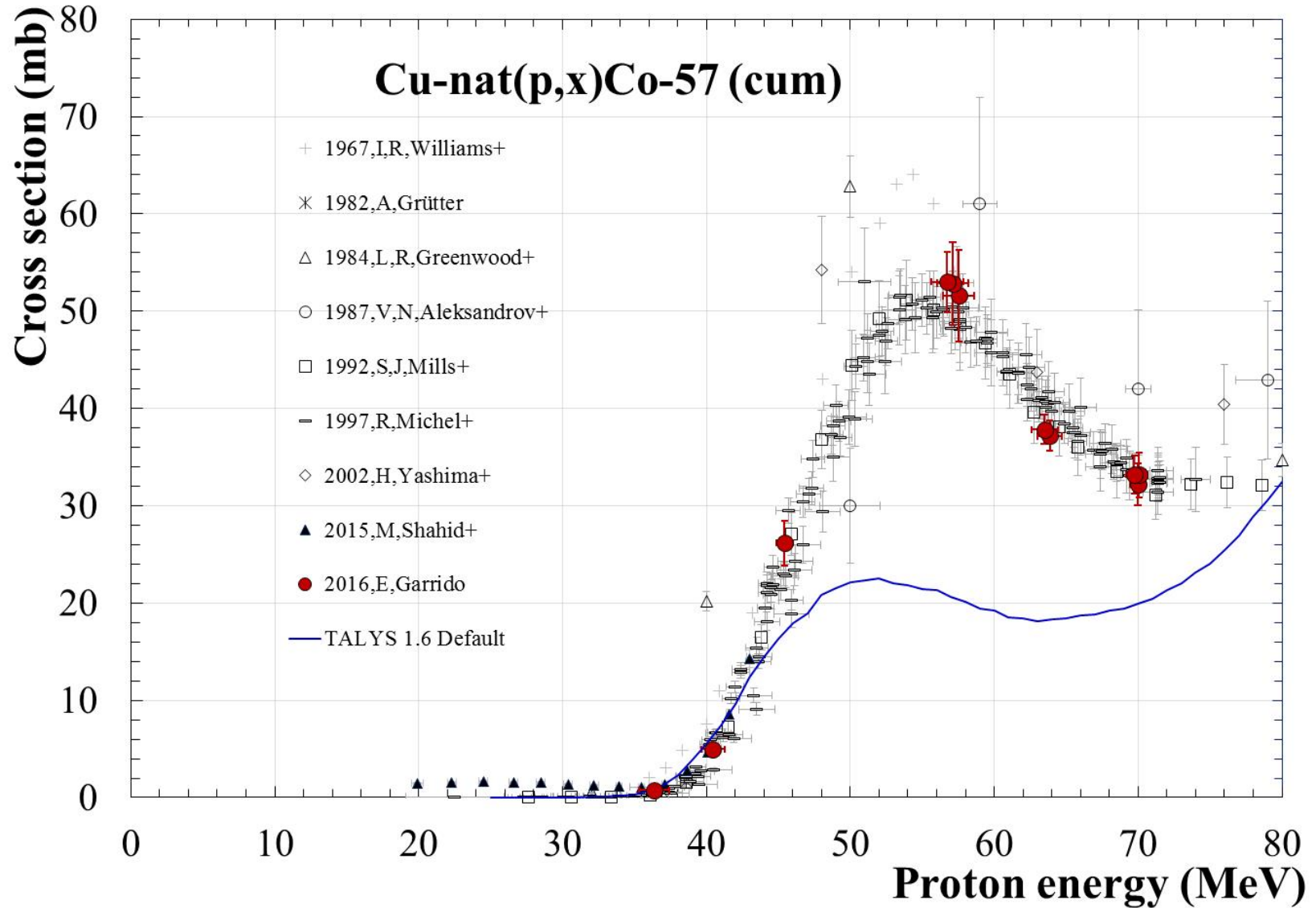
Co-57 ( $T_{1/2} = 271.79$  d)

EC process (100%) to stable Fe-57

Suitable for proton monitor reaction

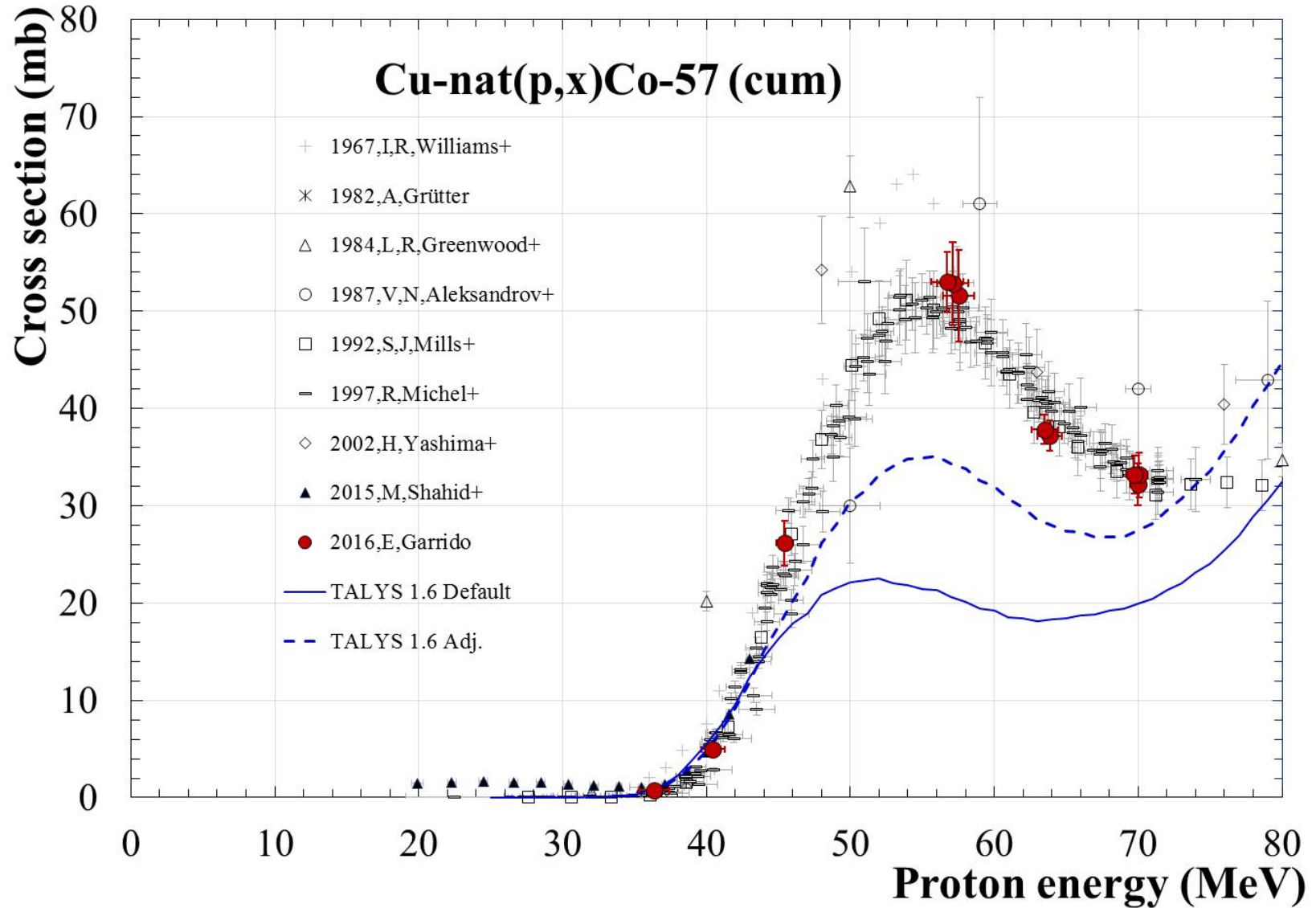
*E. Garrido et al., Nucl. Instr. Meth. Phys. Res. B 383 (2016) 191-212*

# Monitor reaction



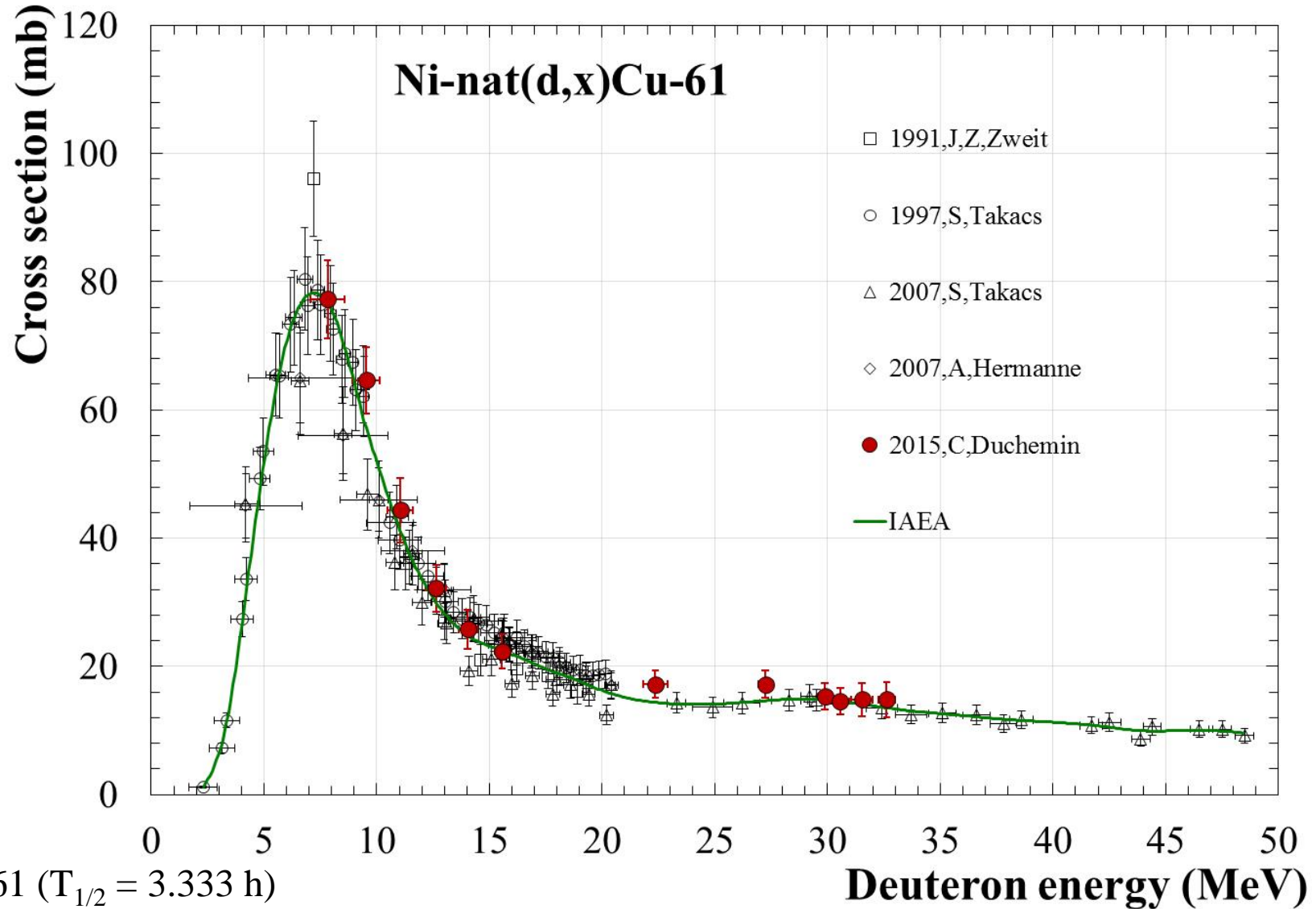
*E. Garrido et al., Nucl. Instr. Meth. Phys. Res. B 383 (2016) 191-212*

# Monitor reaction



*E. Garrido et al., Nucl. Instr. Meth. Phys. Res. B 383 (2016) 191-212*

# Monitor reaction



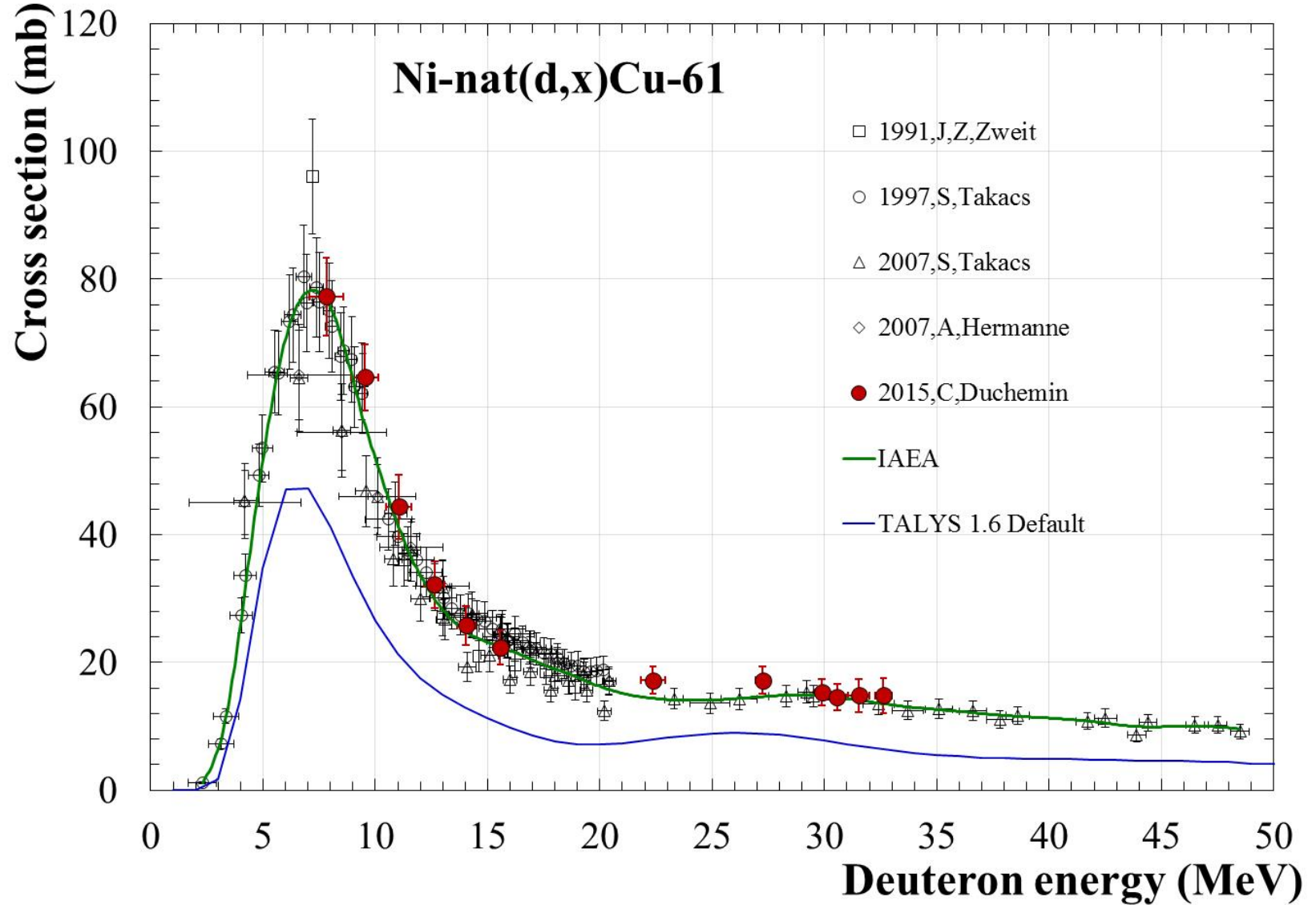
Cu-61 ( $T_{1/2} = 3.333$  h)

EC  $\beta^+$  processes to stable Ni-61

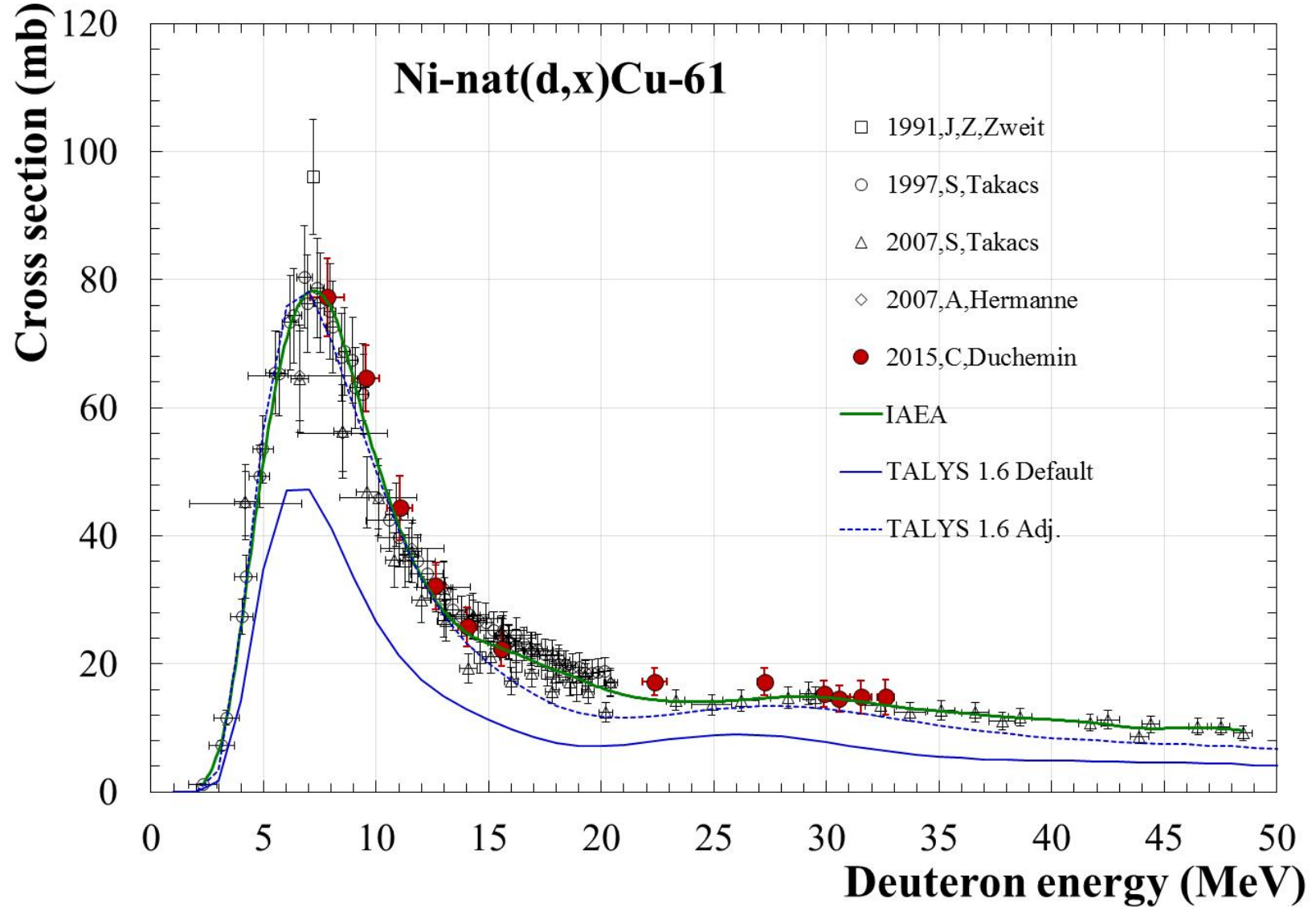
Suitable for PET imaging

*C. Duchemin et al., PhD thesis (2016)*

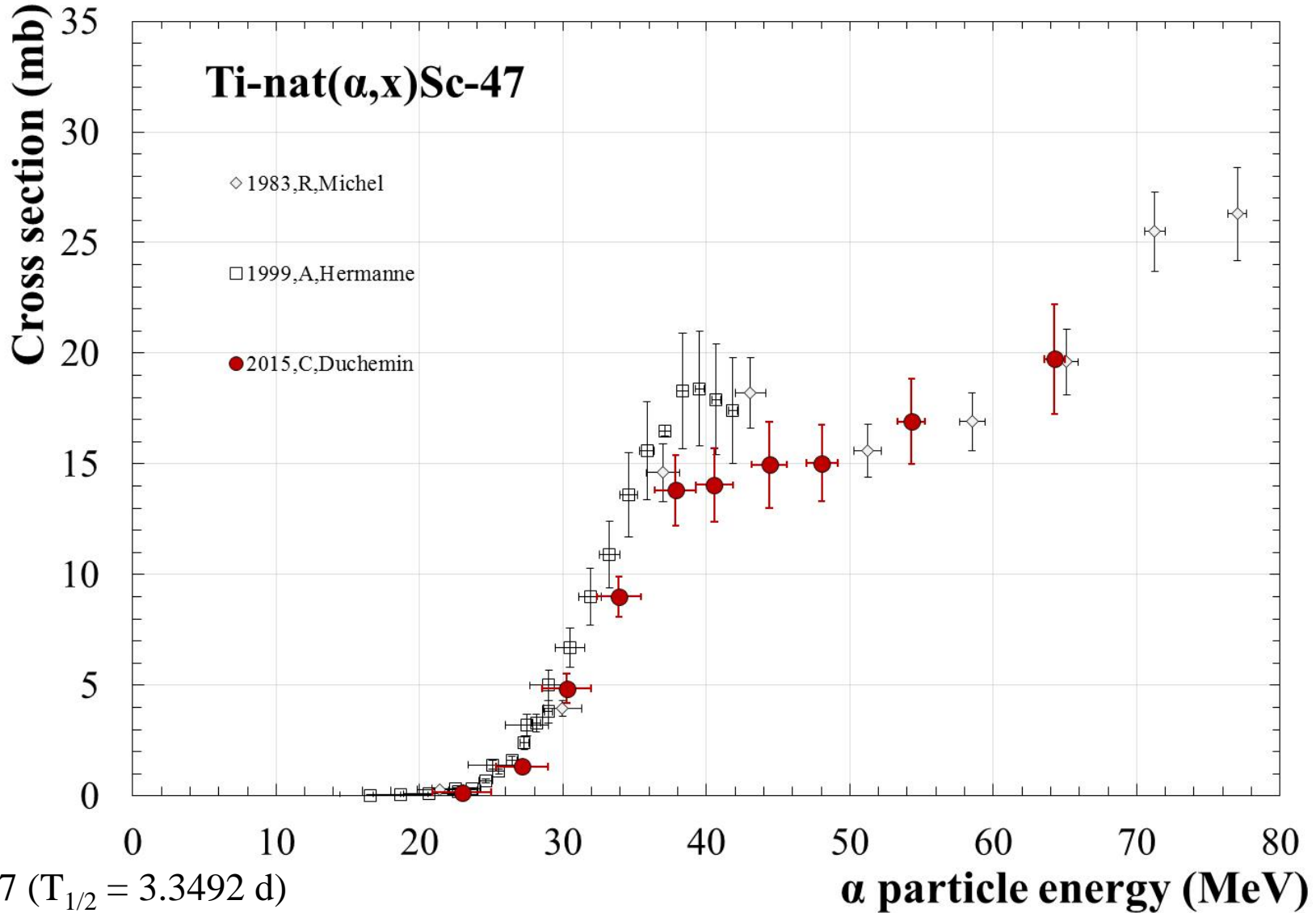
# Monitor reaction



# Monitor reaction



# Monitor reaction

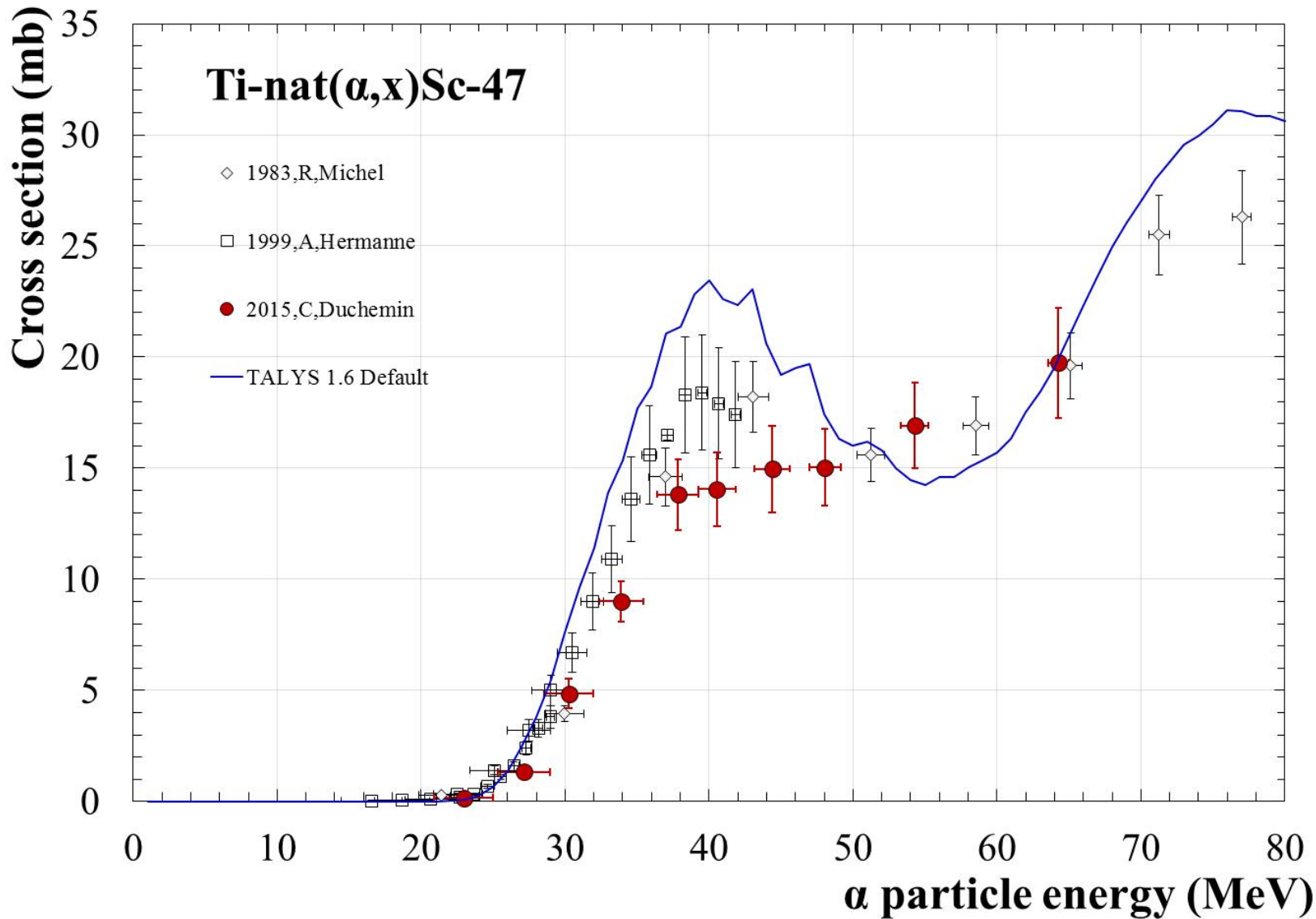


Sc-47 ( $T_{1/2} = 3.3492$  d)  
 $\beta^-$  (100%) to stable Ti-47

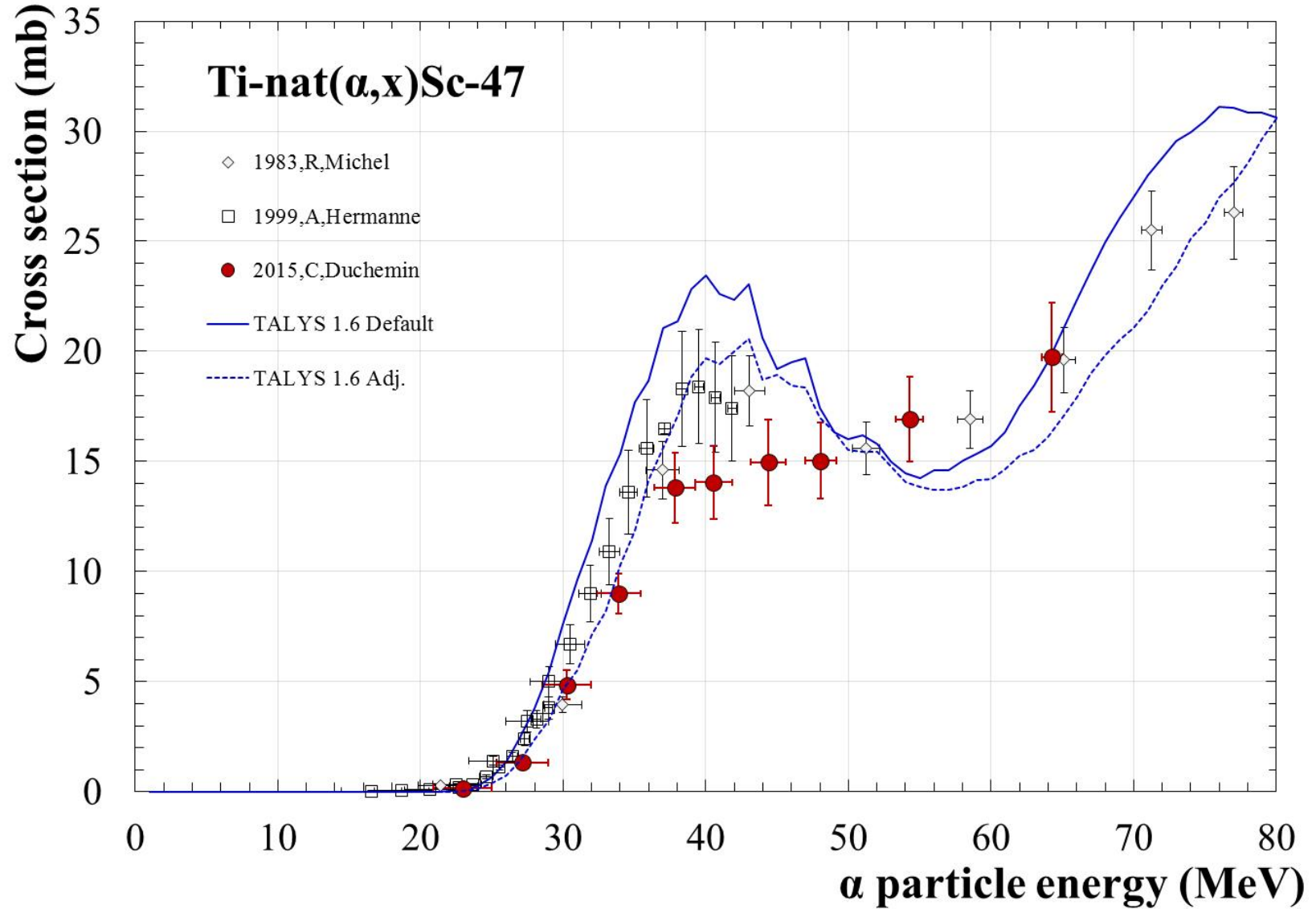
Suitable for theranostic approach with the Sc-44

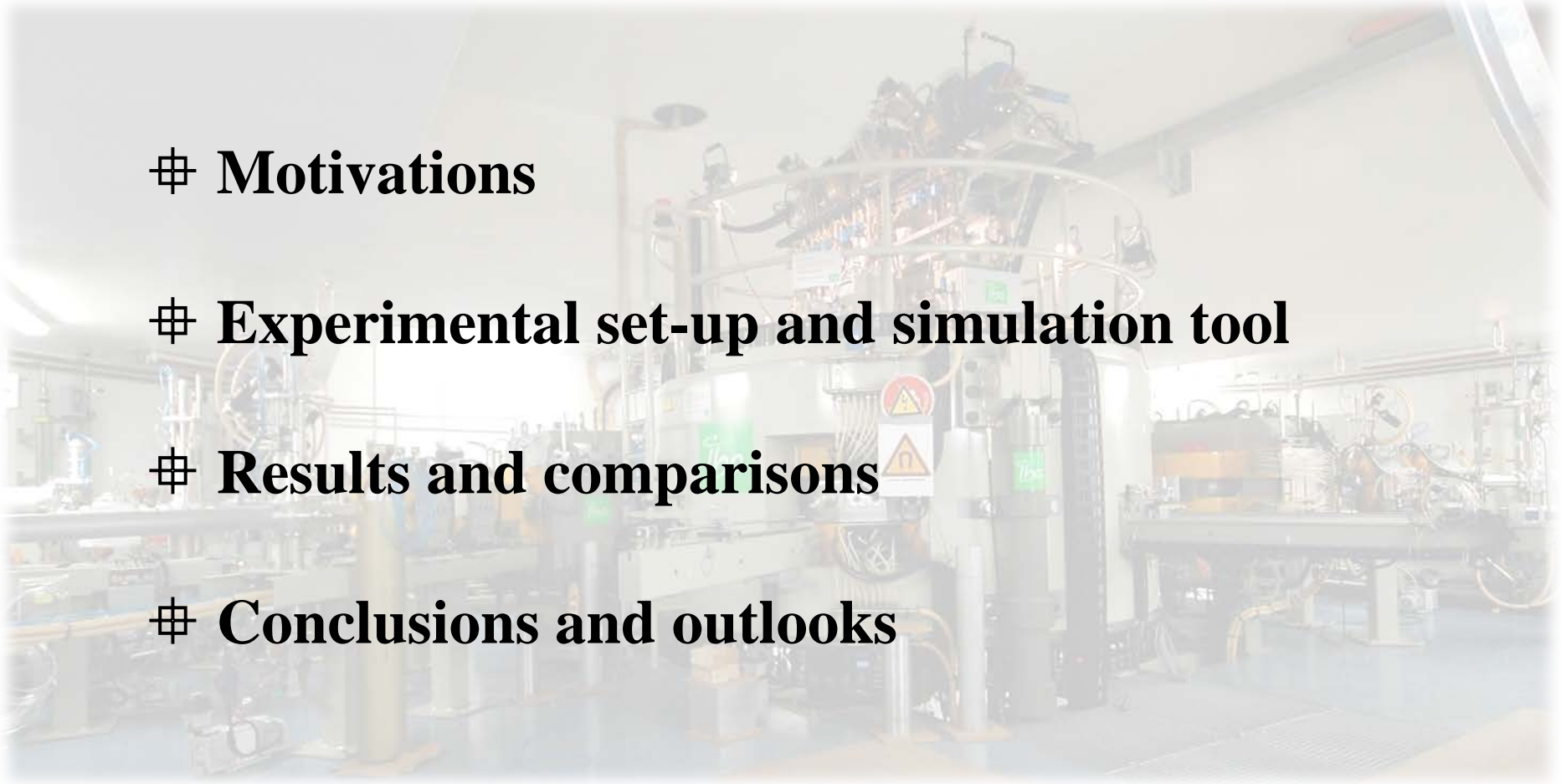
*C. Duchemin et al., PhD thesis (2016)*

# Monitor reaction



# Monitor reaction



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- # Motivations
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**A large set of data have been collected** using the stacked-foil technique at ARRONAX

- with different type of projectiles (proton, deuteron and alpha particles)
- for materials all over the mass range
- for diagnosis and therapy purposes in nuclear medicine

Comparisons have been performed systematically with the **TALYS 1.6 code**

- state of the art models included
- possibility to combine models to better describe data
- strong reactivity of authors

Three main mechanisms have been studied at the moment:

- optical potential
- level density description
- preequilibrium model

A set of models have been found to allow a **good description of all our collected data**, which is different from the suggested default models.

Further investigations will be done on other mechanisms that can affect our observable.

**Thank you for your attention**

Acknowledgments to the ND2016 organization committee

**“How nuclear data collected for medical radionuclides production could constrain nuclear codes”**

**Guertin A.<sup>1</sup>, Duchemin C.<sup>1,2</sup>, Haddad F.<sup>1,3</sup>, Michel N.<sup>1,3</sup>, Métivier V.<sup>1</sup>**

1 SUBATECH, Ecole des Mines de Nantes, Université de Nantes, CNRS/IN2P3, Nantes, France

2 CERN, Radiation Protection Group Special Project Section - HSE-RP-SP, 1211 Genève 23, Suisse

3 GIP ARRONAX, 1 rue Aronnax, 44817 Saint-Herblain cedex – France