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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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GUERTIN Arnaud

Laboratoire Subatech – UMR 6457

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 (γ , β^+)
- Therapy (β^- , α , e_{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

- 
- # Motivations
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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⁻, D⁻: multicusp
 - He²⁺, HH⁺: supernanogan ECR
- 2 extraction lines:
stripper or electrostatic deflector

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

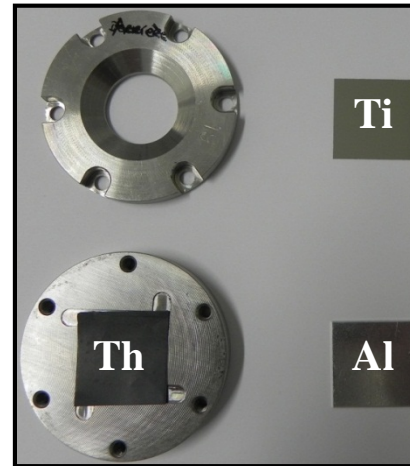
Experimental set-up

Stacked-foil technique:

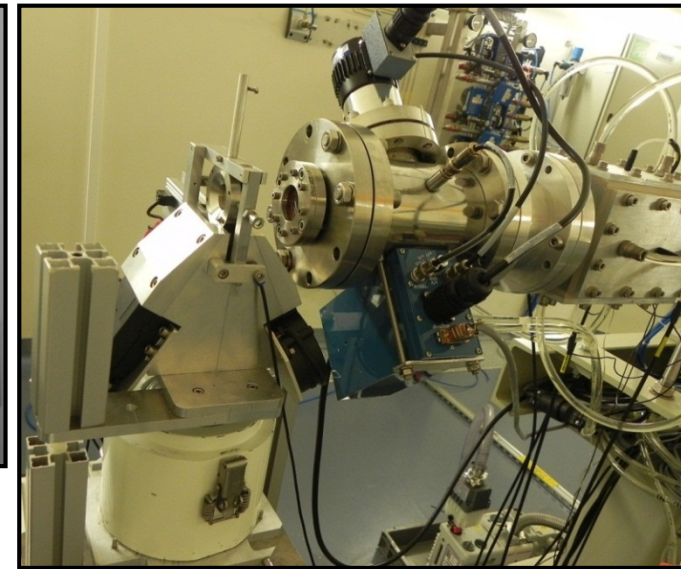
- Target/monitor/degrader **pattern**
- **Thin** foils:
E loss small and constant
- One cross section value per foil

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

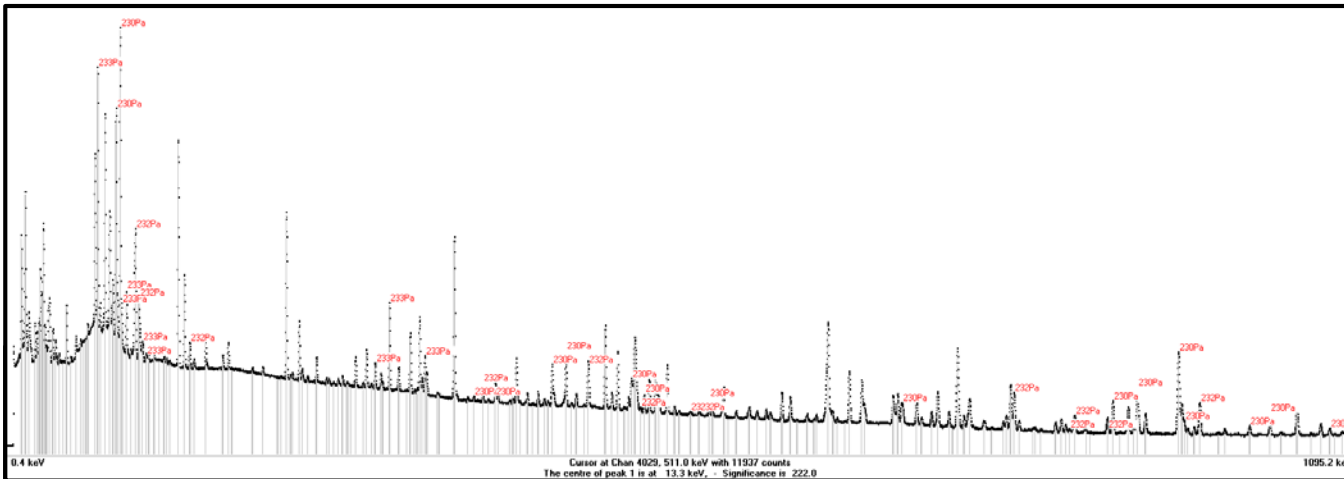
IAEA recommended cross sections:

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

Experimental set-up

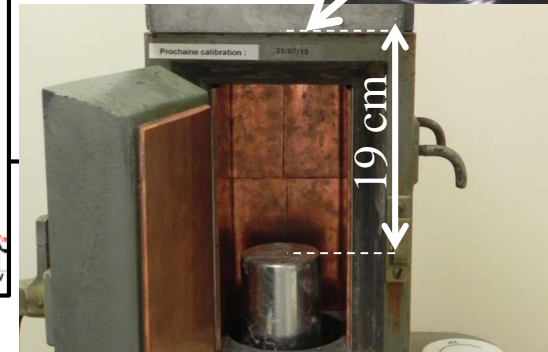
Gamma spectroscopy

- **HPGe** coaxial detector
- Dead time: < 10% (sum peak)
- Activity values: FitzPeaks
- $T_{1/2}$, E_γ , I_γ : Lund/LBNL, NNDC
- γ spectra recorded on **8192 channels**
- FWHM: 1.04 keV at 122 keV (^{57}Co)
- 1.97 keV at 1332 keV (^{60}Co)
- Energy and efficiency calibrations: Co and Eu

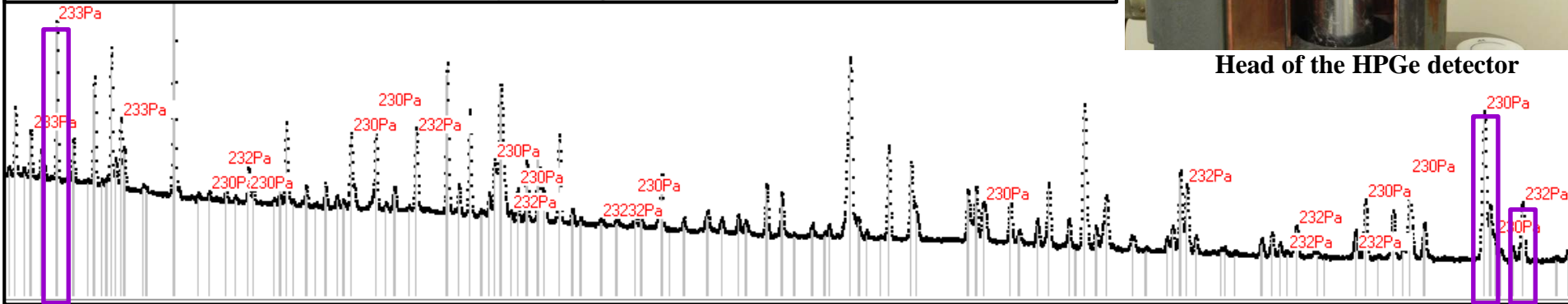


Target and monitor:

- ✓ counted twice
- ✓ during > 24 hrs
- ✓ 2 week delay



Head of the HPGe detector



γ rays used to extract production cross section of $^{230,232,233}\text{Pa}$

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(α ,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, α particles

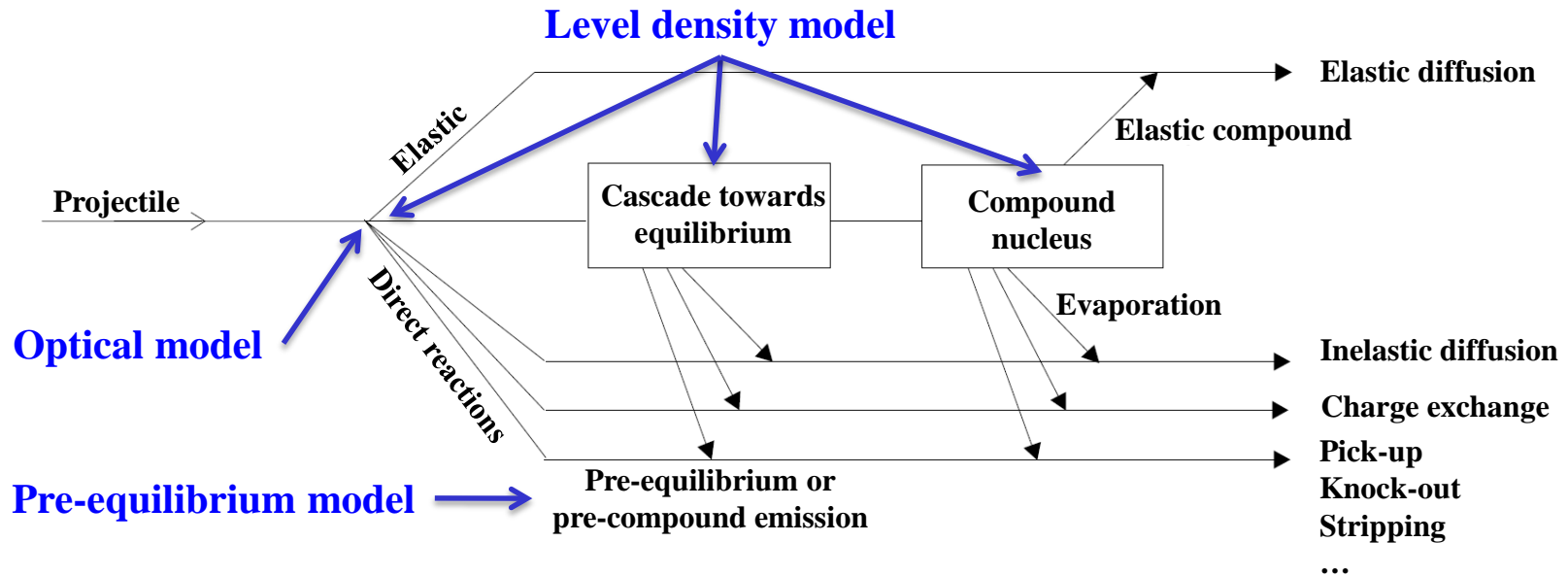
Many state-of-the-art nuclear models

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. Koning¹, D. Rochman², J. Kopecky³, J.Ch. Sublet⁴, M. Fleming⁴, E. Bauge⁷, S. Hilaire⁷, P. Romain⁷, B. Morillon⁷, H. Duarte⁷, S.C van der Marck⁶, S. Pomp⁵, H. Sjostrand⁵, R. Forrest¹, H. Henriksson⁸, O. Cabellos⁹, S. Goriely¹⁰, J. Leppanen¹¹, H. Leeb¹², A. Plompen¹³, and R. Mills¹⁴

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

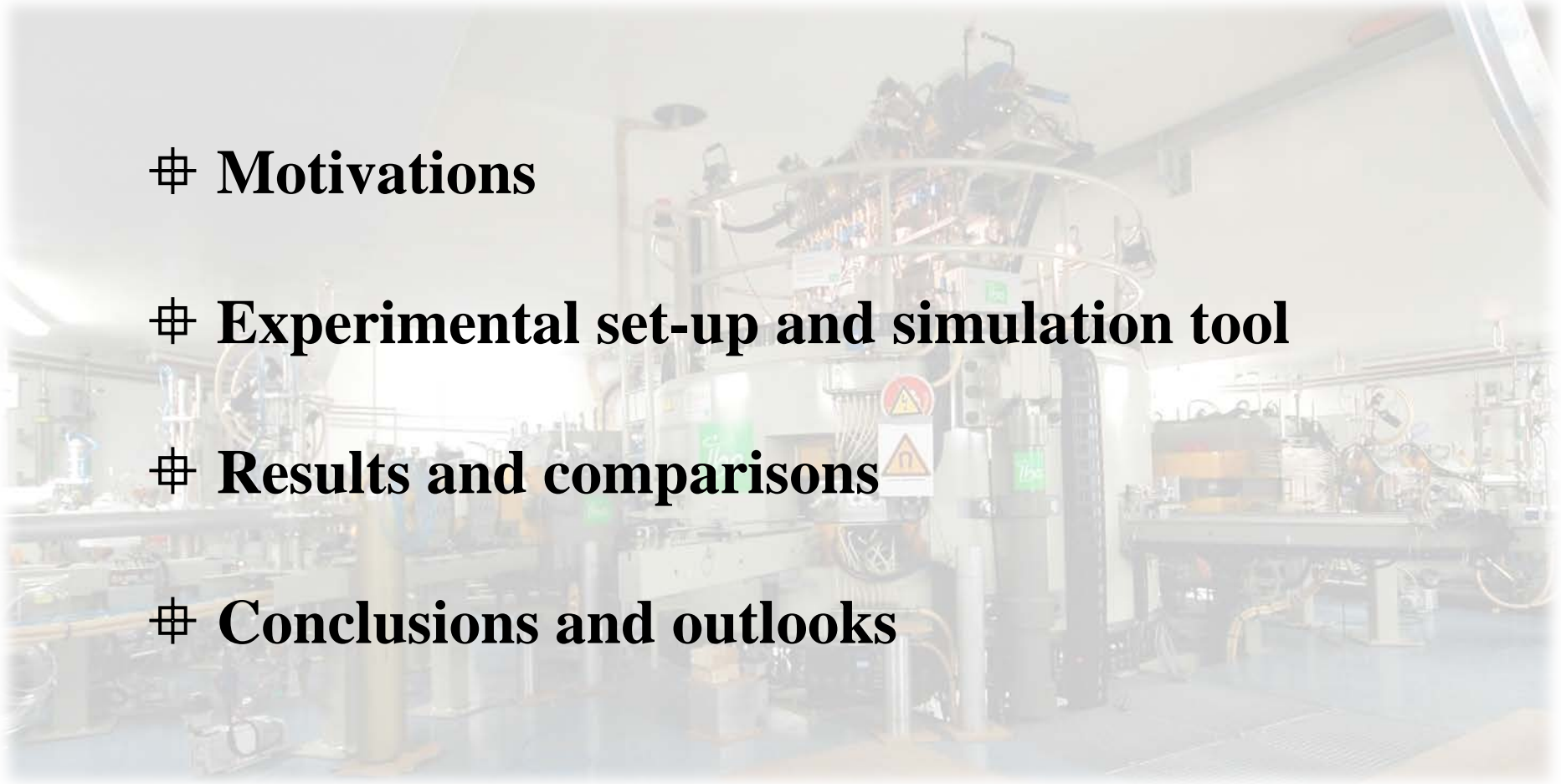
TENDL2015 contains evaluations for :

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

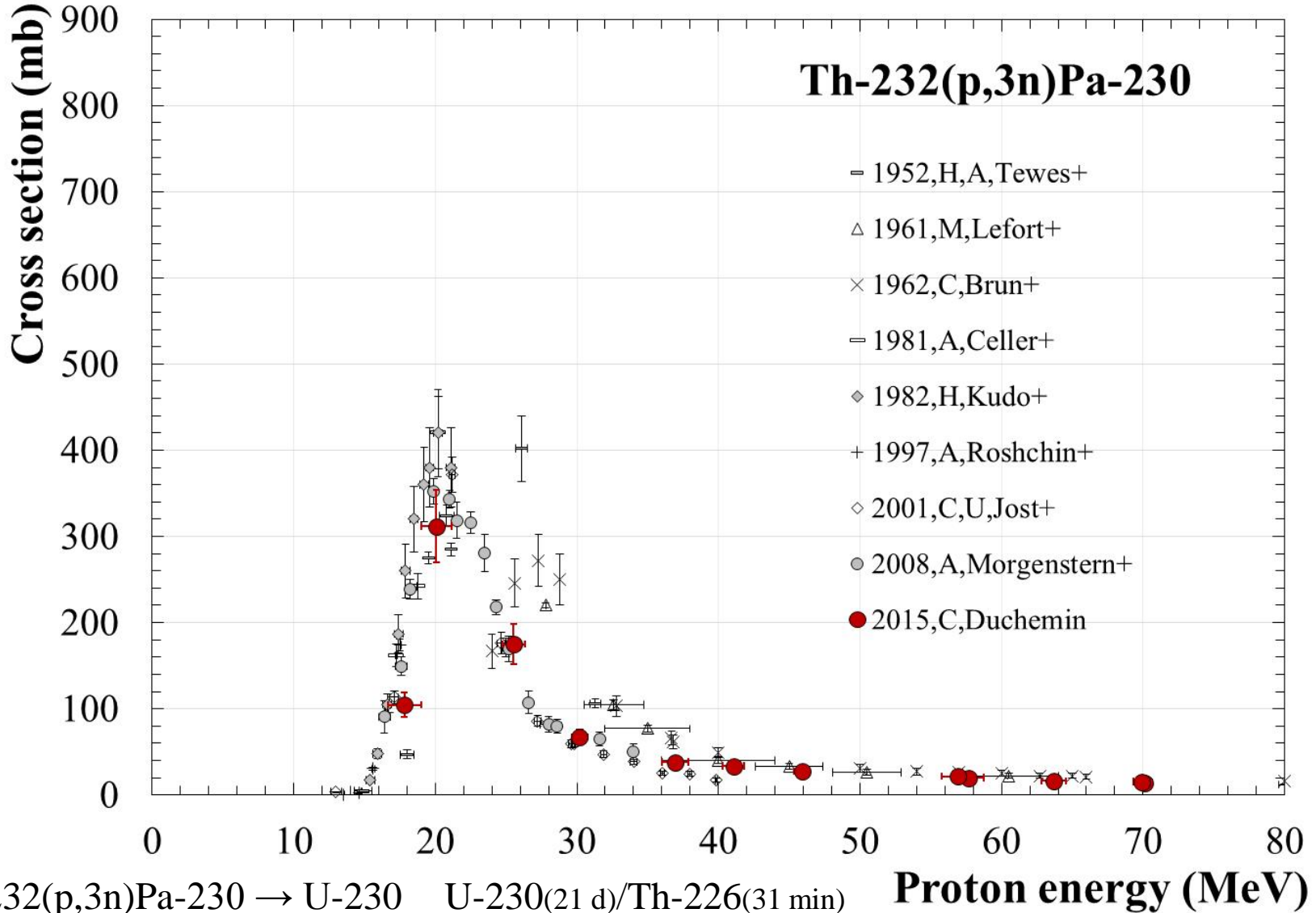
all isotopes living more than 1 second (~ 2800 isotopes)

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

Koning A.J. et al., 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

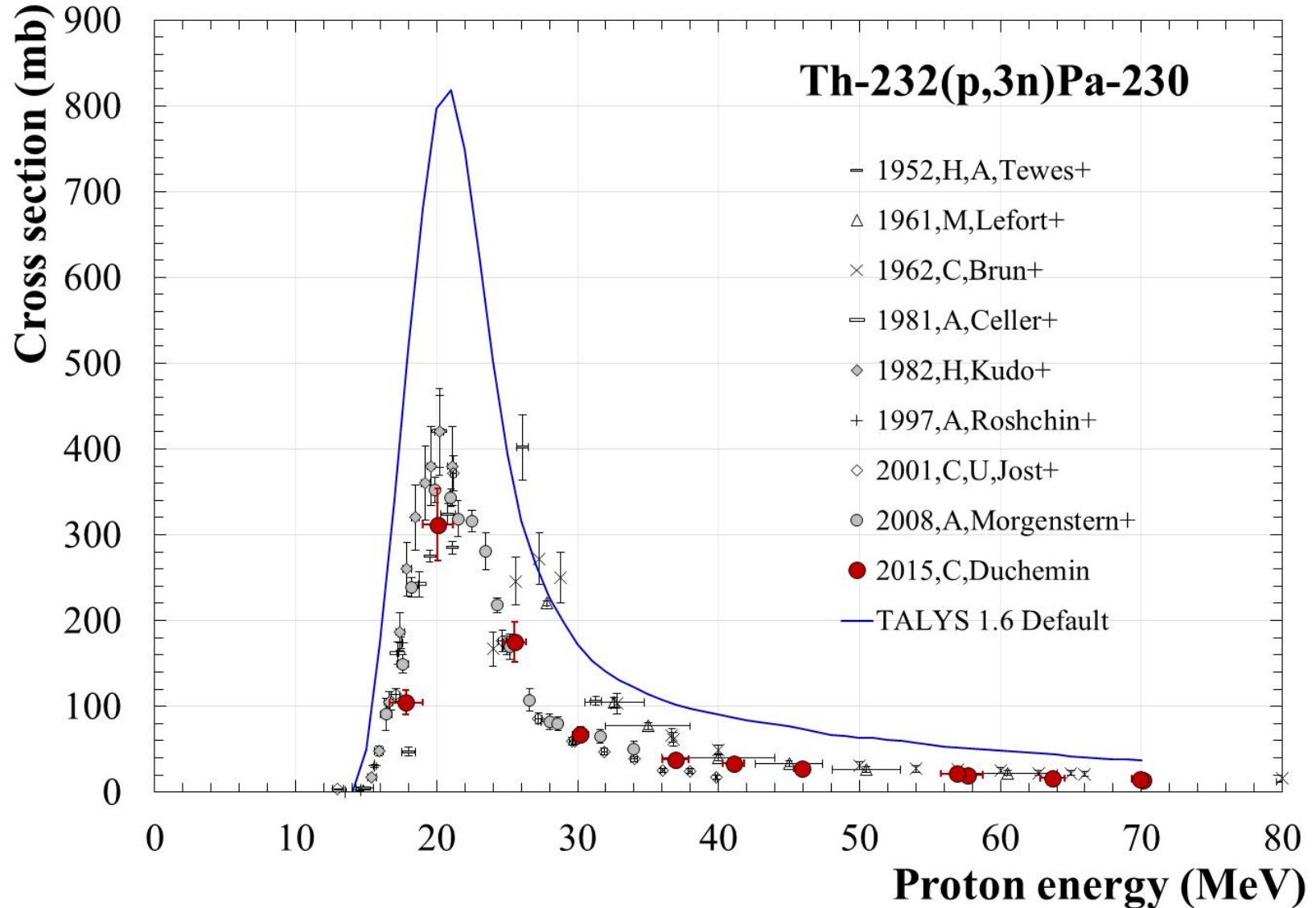


α RIT for leukaemia treatment

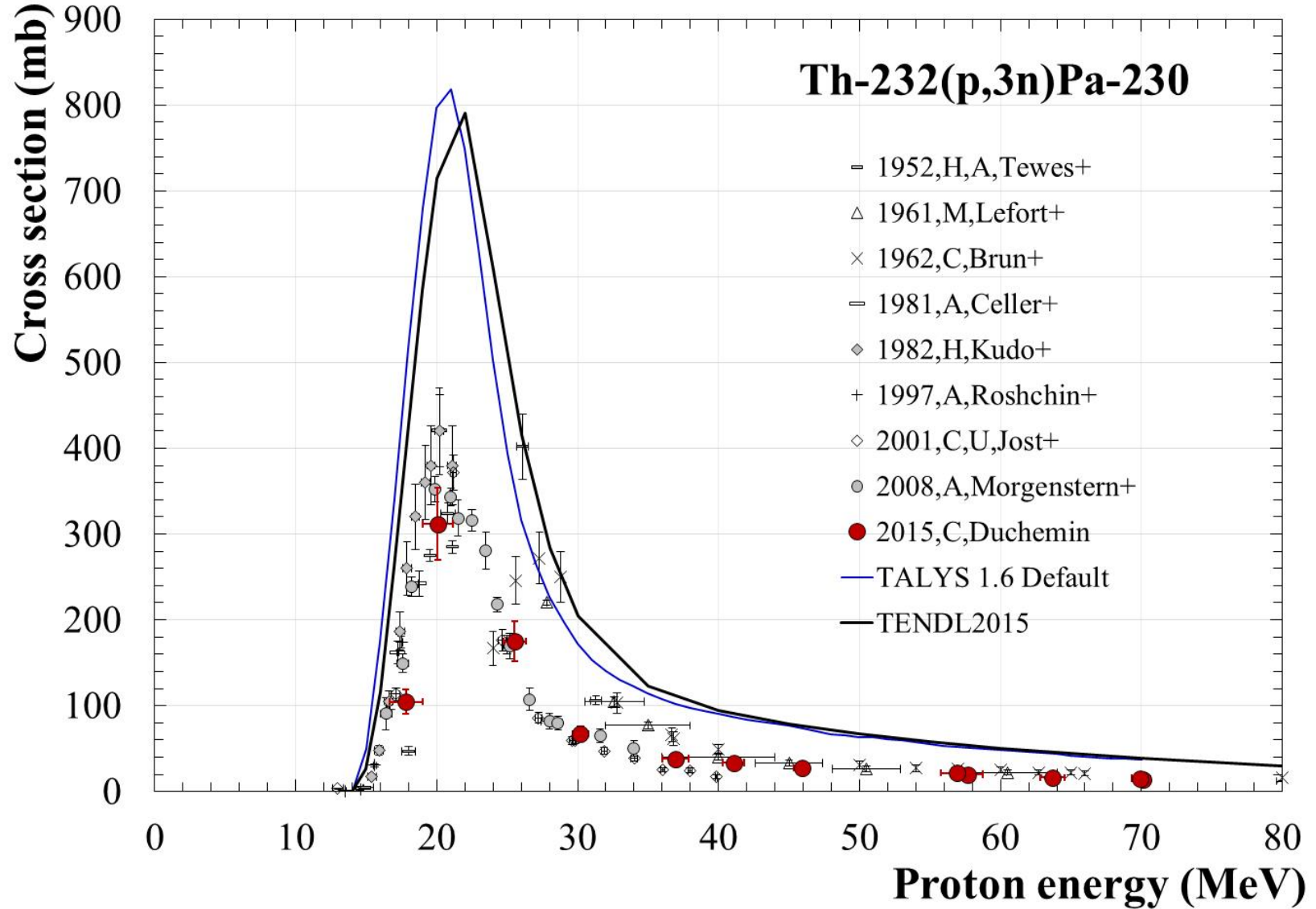
4 α cascade of 27.7 MeV

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

Novel therapeutic nuclide



Novel therapeutic nuclide



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

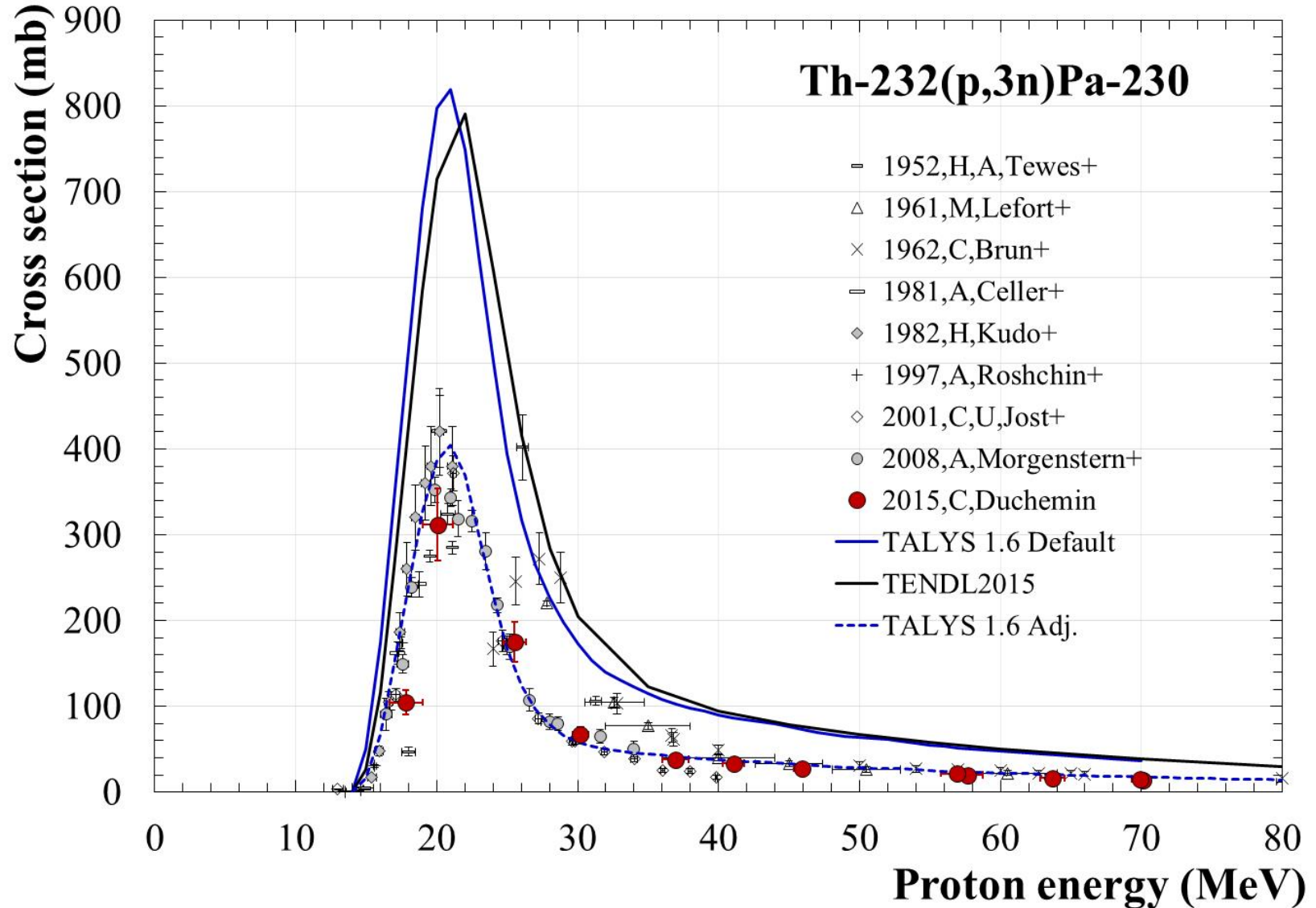
⇒ TALYS 1.6 Default

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.

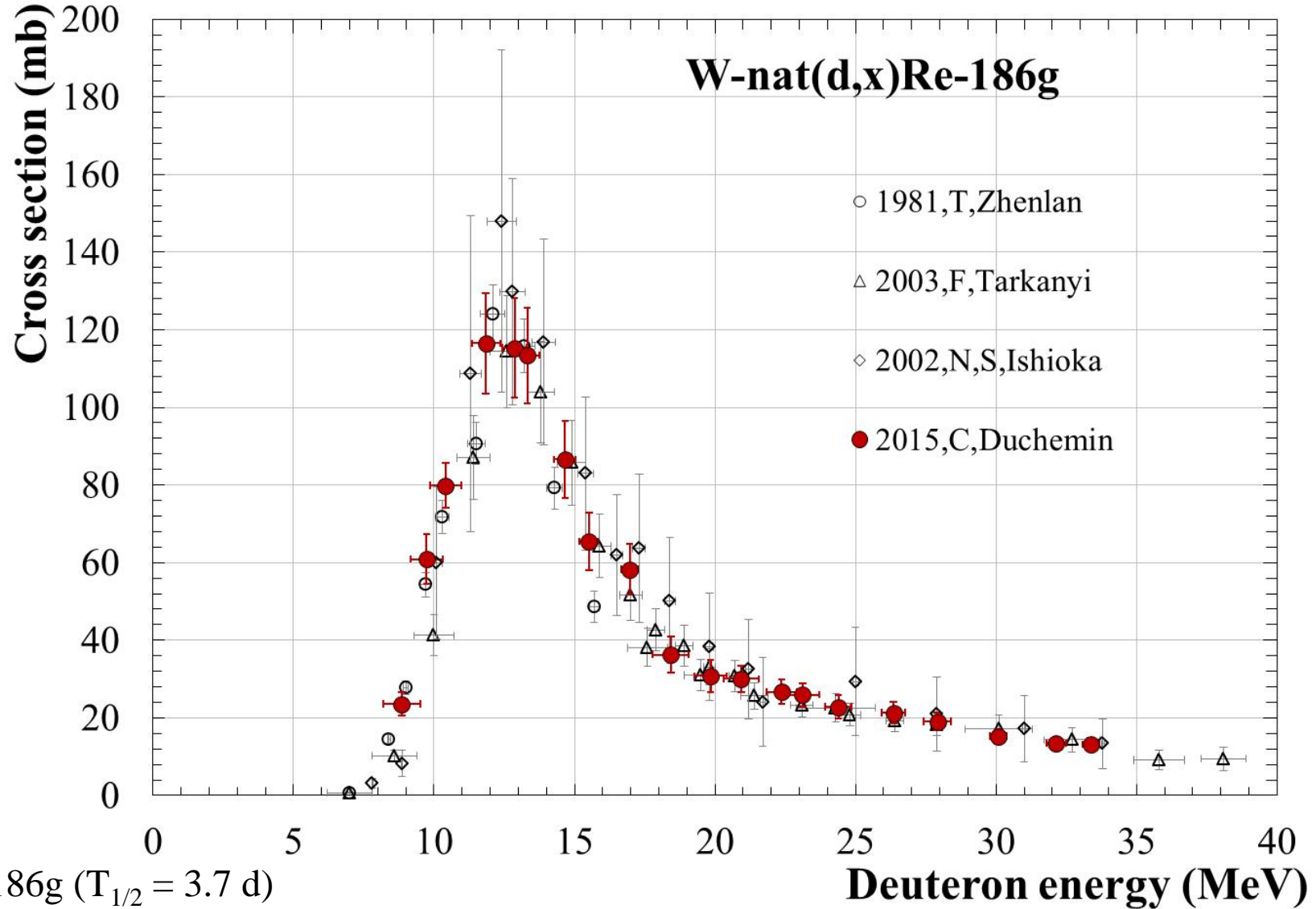
Models	Projectile	Default	Adj.
Optical	p (1)	A.J. Koning and J.P. Delaroche (2003)	A.J. Koning and J.P. Delaroche (2003)
	d (5)	S. Watanabe (1958) ...	Y. Han et al. (2006) ...
	α (5)	L. McFadden and G.R. Satchler (1966) ...	Demetriou et al. (2002) ...
Pre-equilibrium	All (4)	2 components exciton A.J.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

Novel therapeutic nuclide



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

Novel therapeutic nuclide

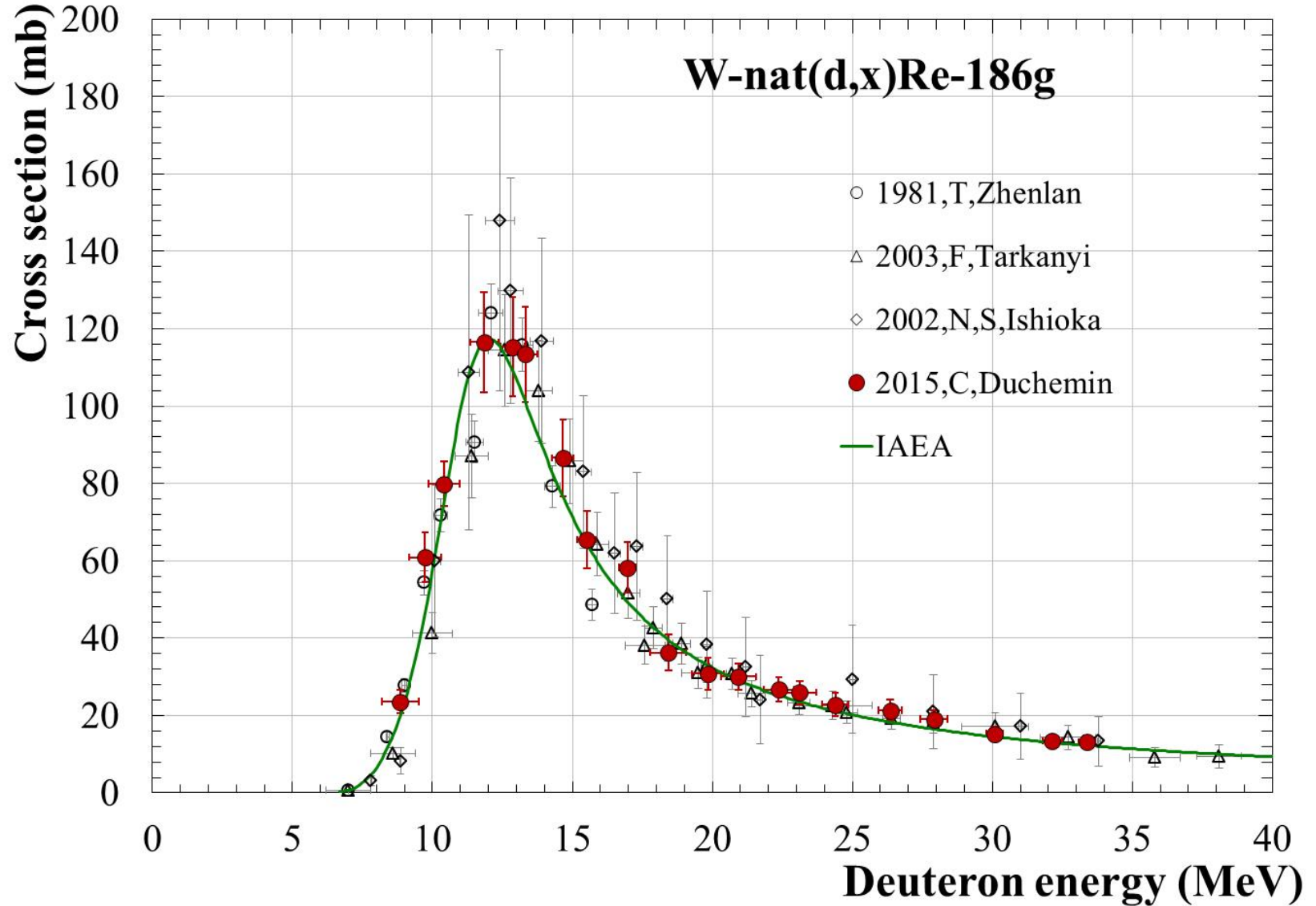


Re-186g ($T_{1/2} = 3.7$ d)

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

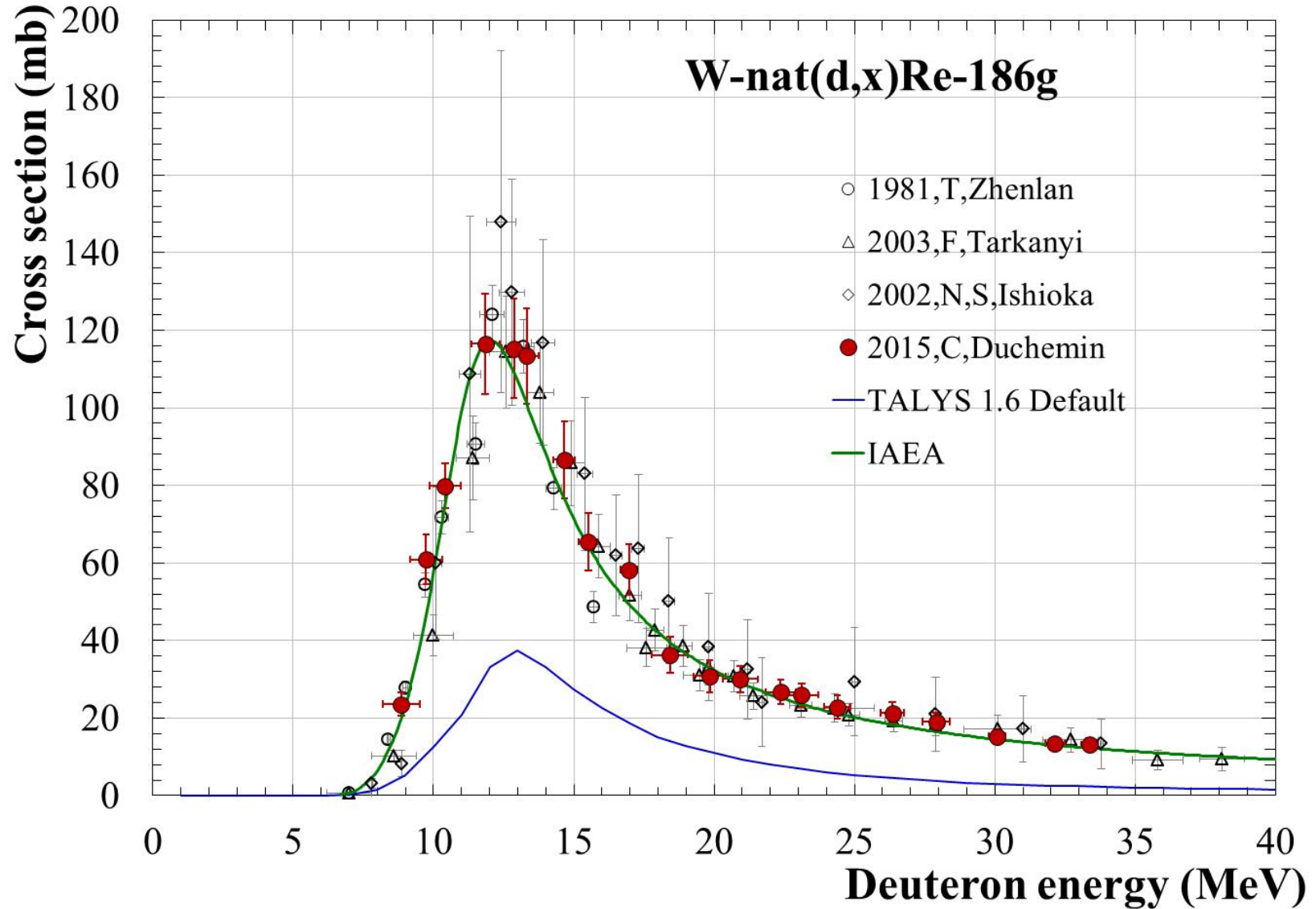
C. Duchemin et al., Appl. Radiat. Isot. 97 (2015) 52-58

Novel therapeutic nuclide

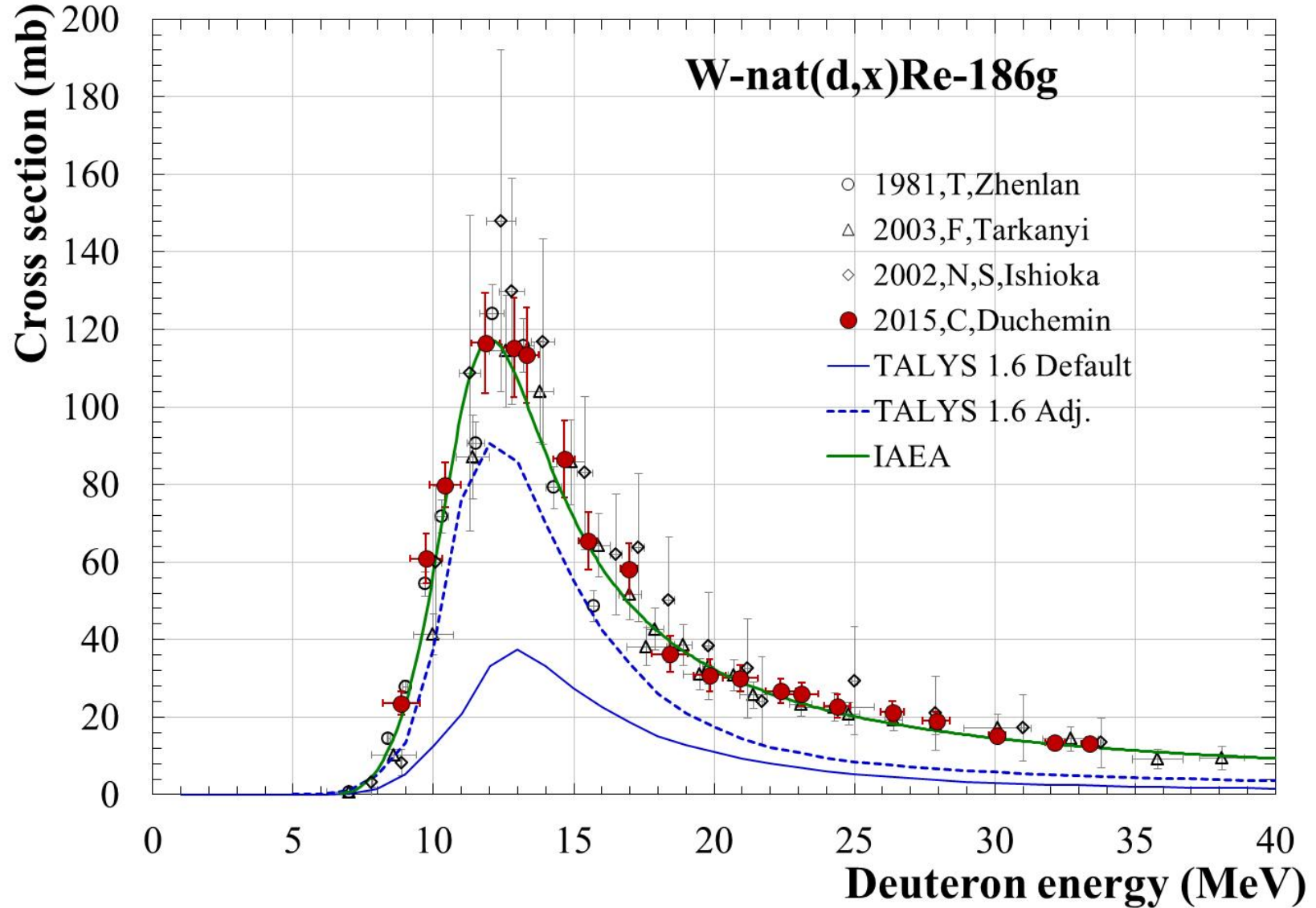


C. Duchemin et al., *Appl. Radiat. Isot.* 97 (2015) 52-58

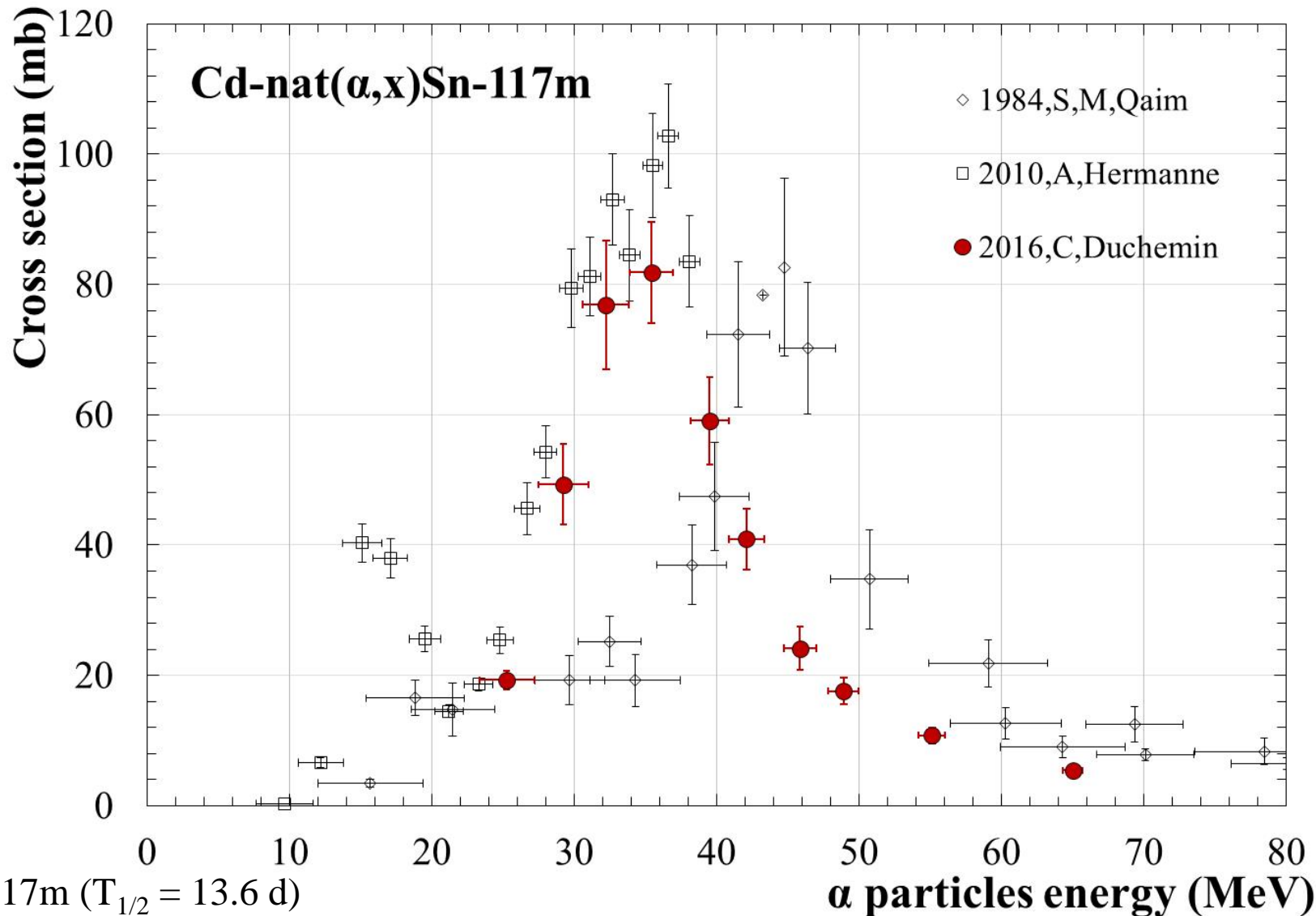
Novel therapeutic nuclide



Novel therapeutic nuclide



Novel therapeutic and imaging nuclide



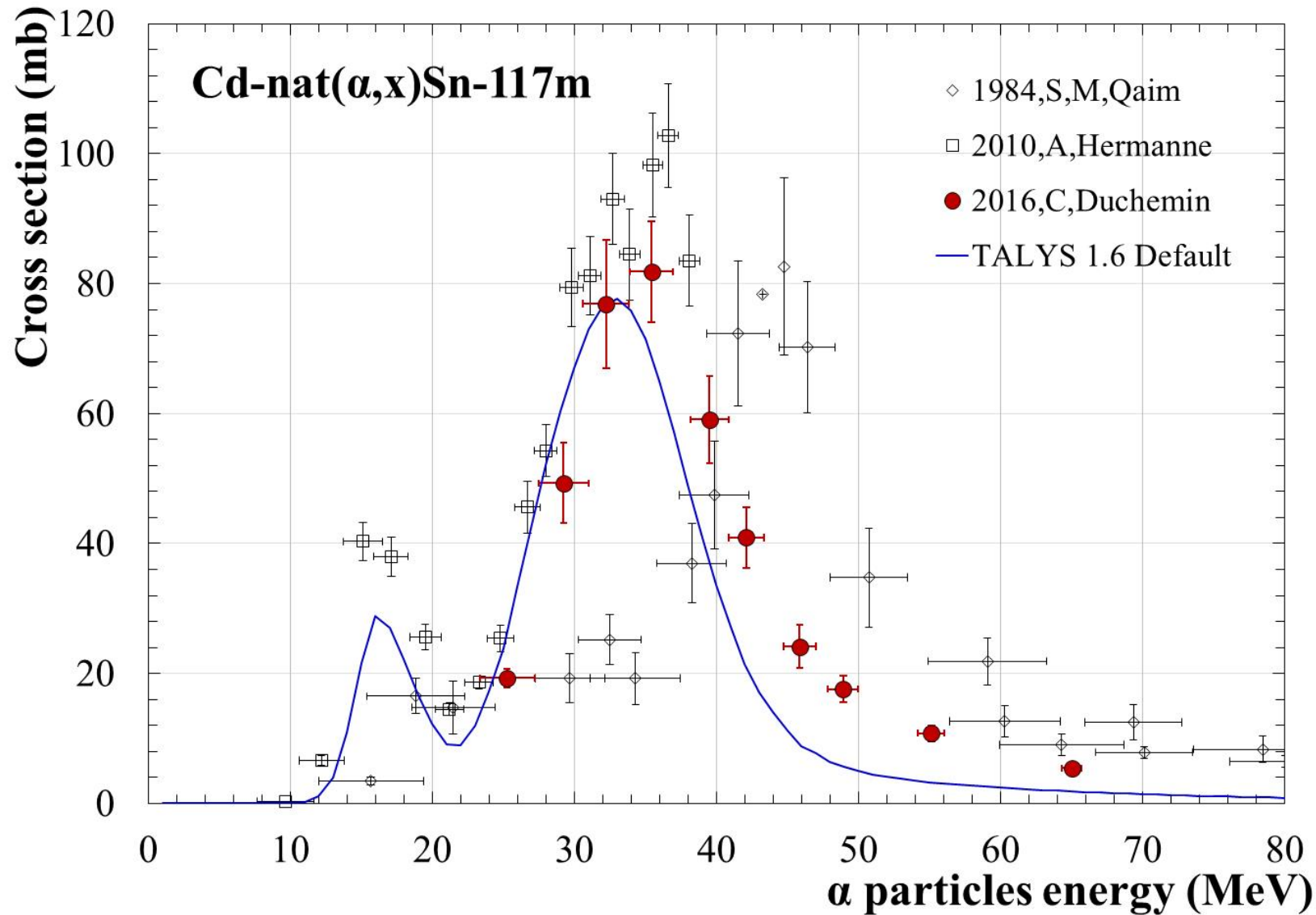
Sn-117m ($T_{1/2} = 13.6$ d)

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

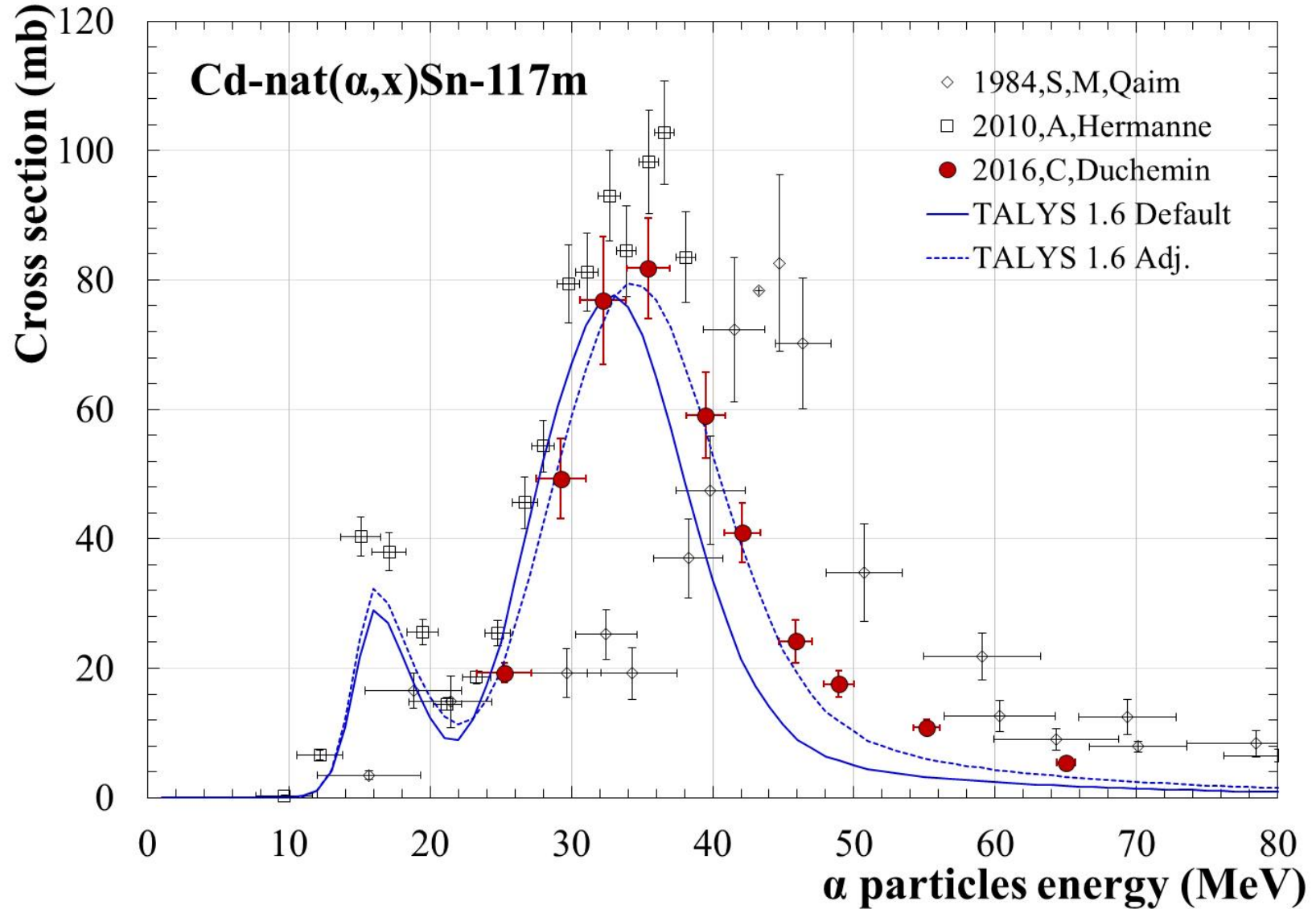
158 keV gamma ray suitable for SPECT imaging

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

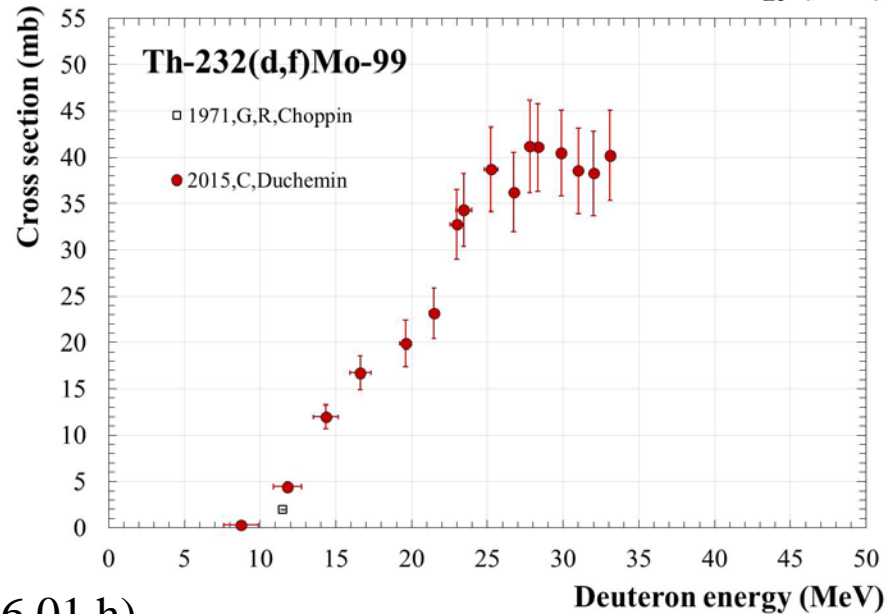
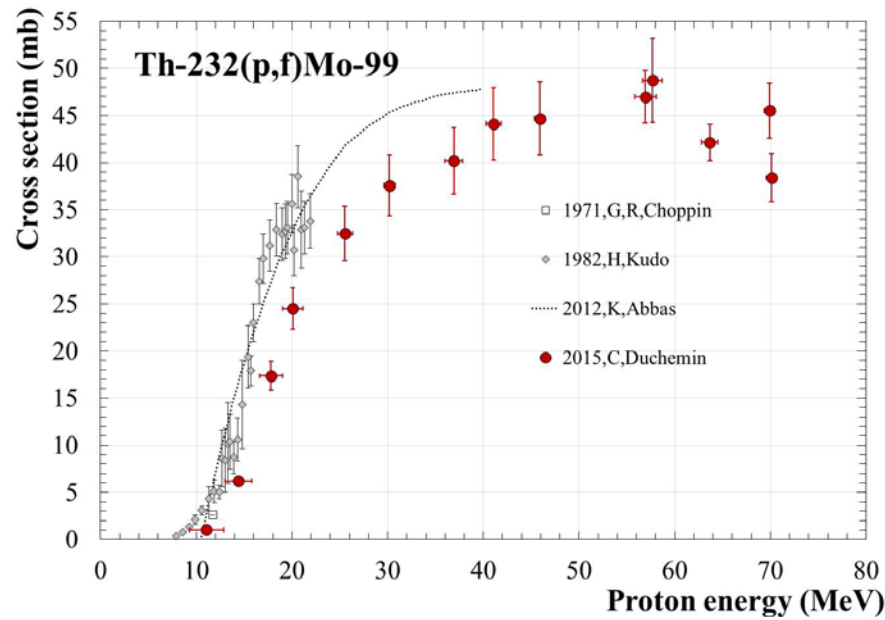
Novel therapeutic and imaging nuclide



Novel therapeutic and imaging nuclide



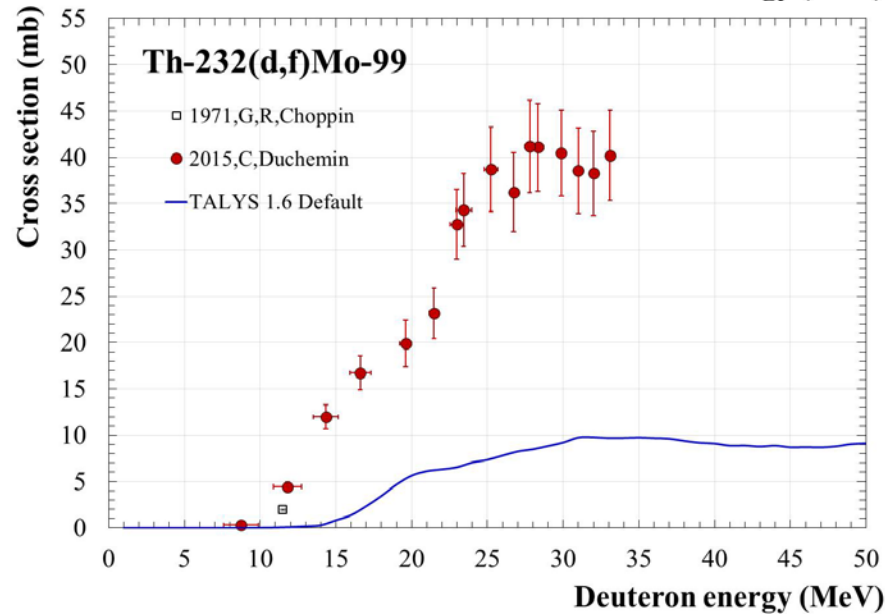
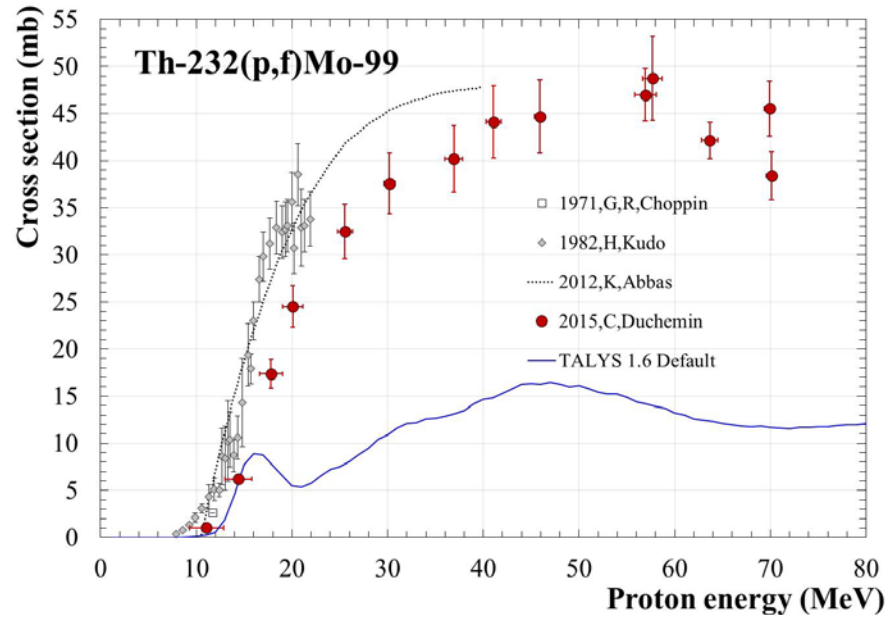
Potential method for production of $^{99}\text{Mo}/^{99m}\text{Tc}$



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

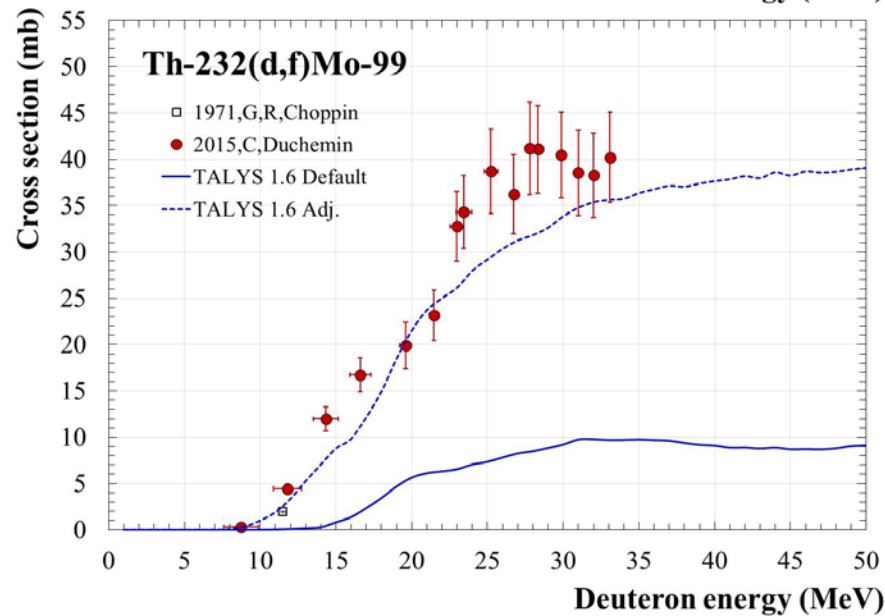
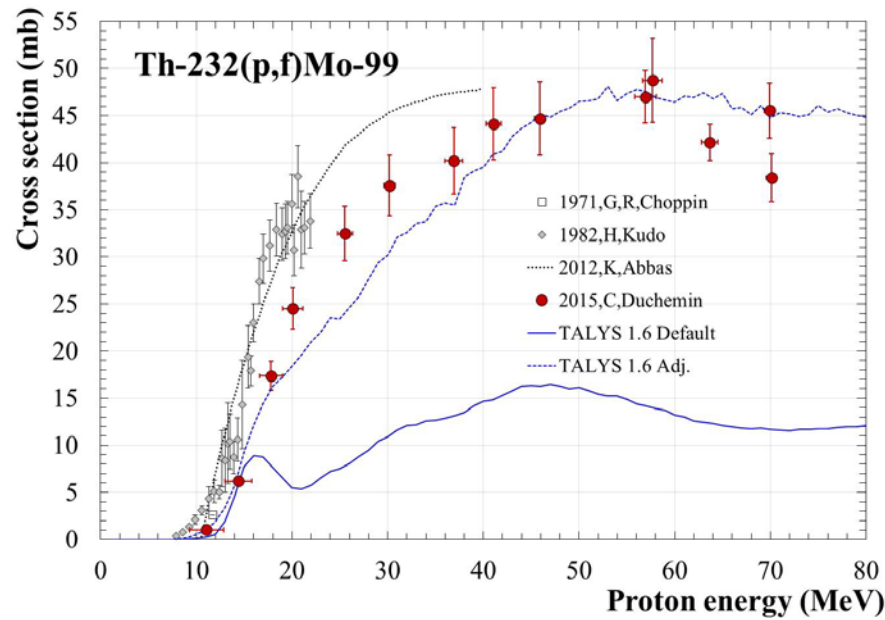
C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946

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



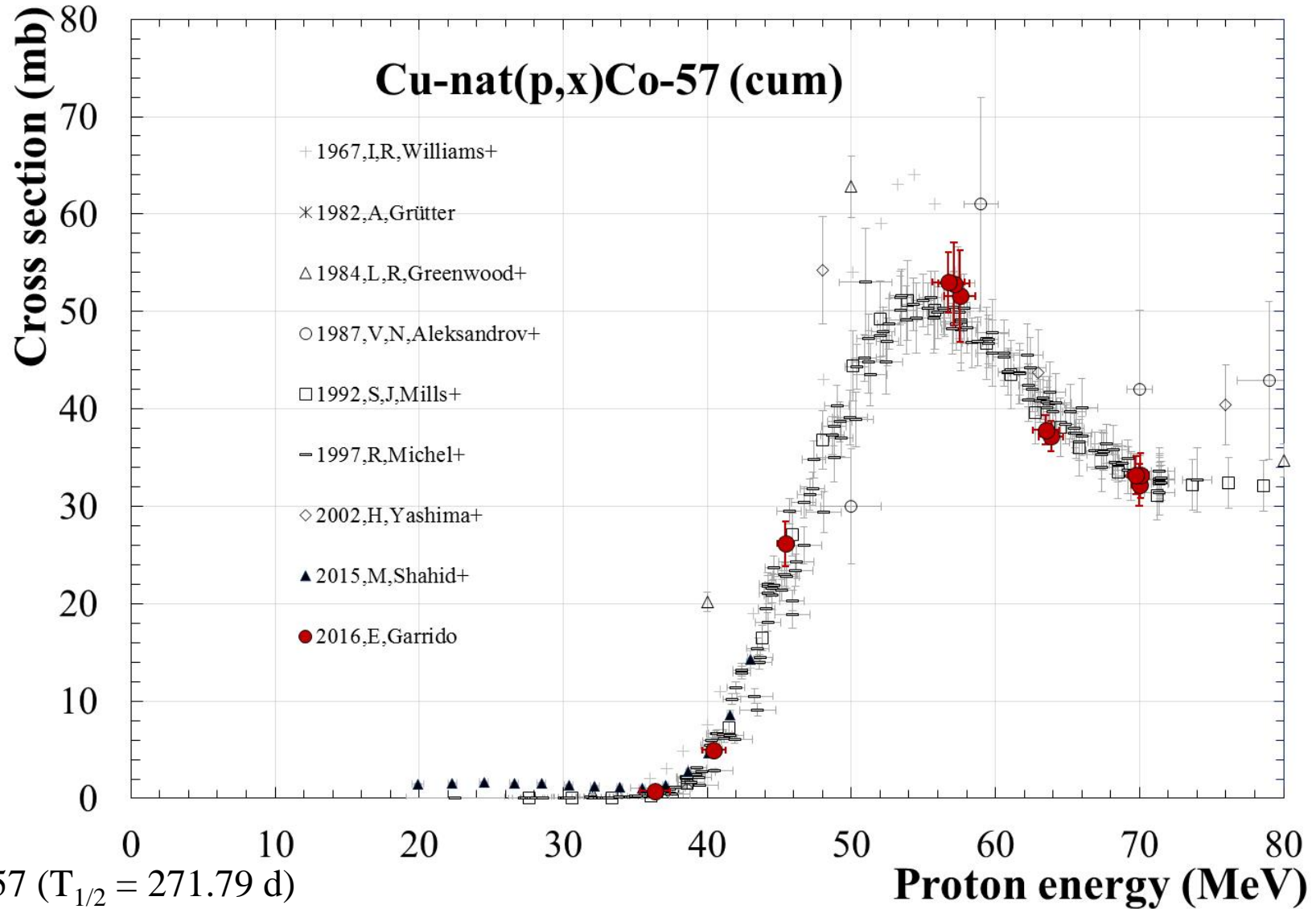
C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946

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



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

Monitor reaction



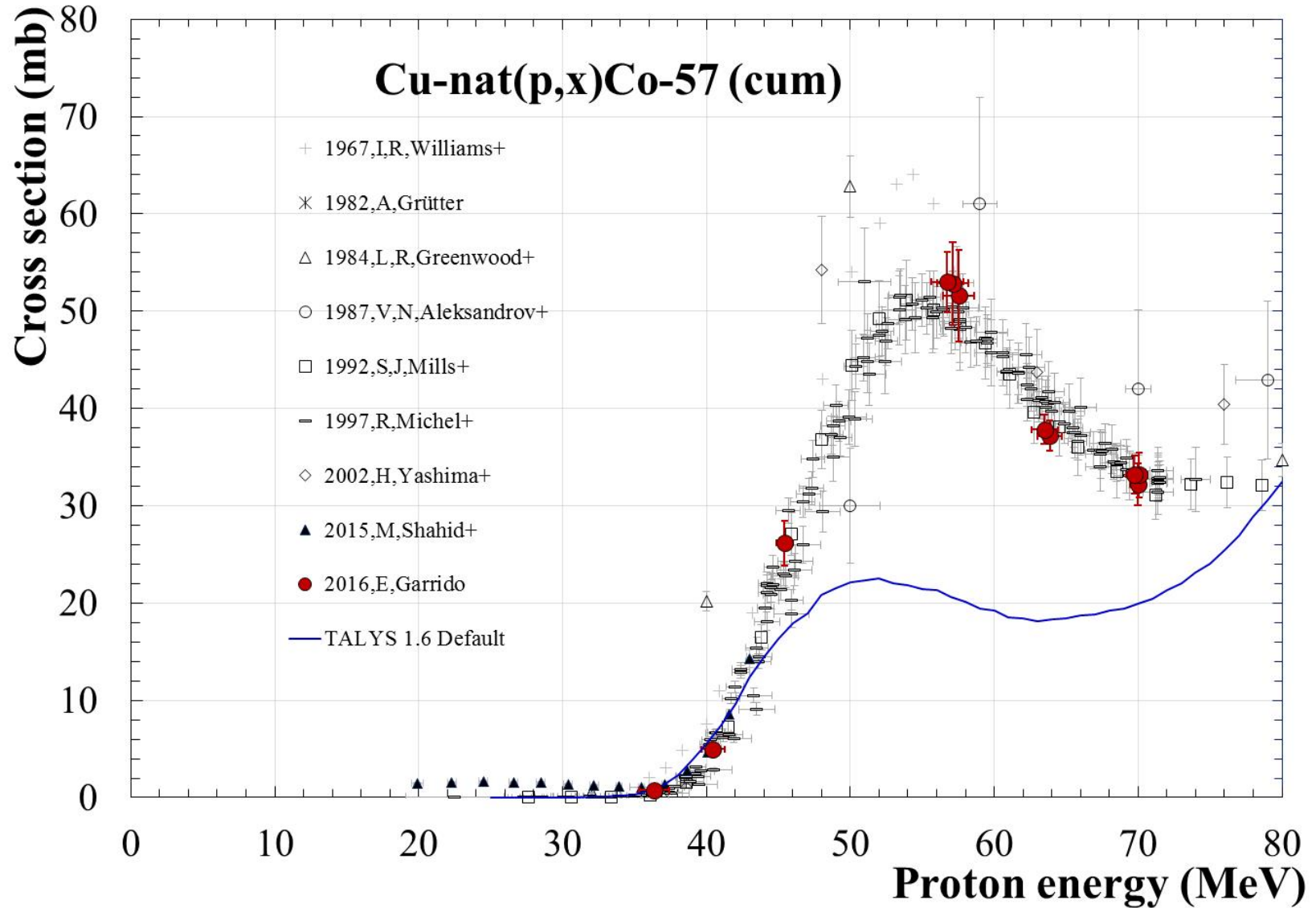
Co-57 ($T_{1/2} = 271.79$ d)

EC process (100%) to stable Fe-57

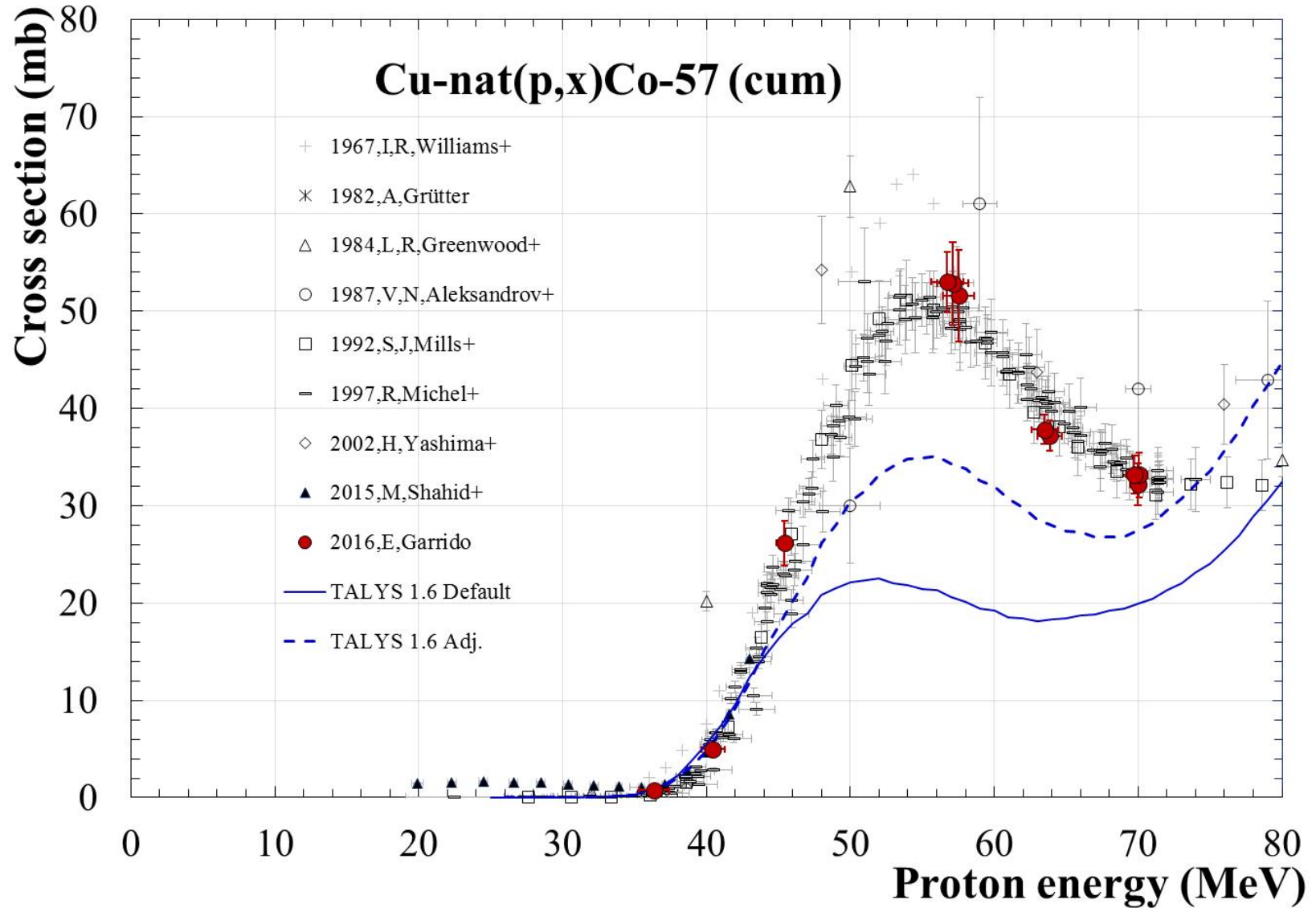
Suitable for proton monitor reaction

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

Monitor reaction

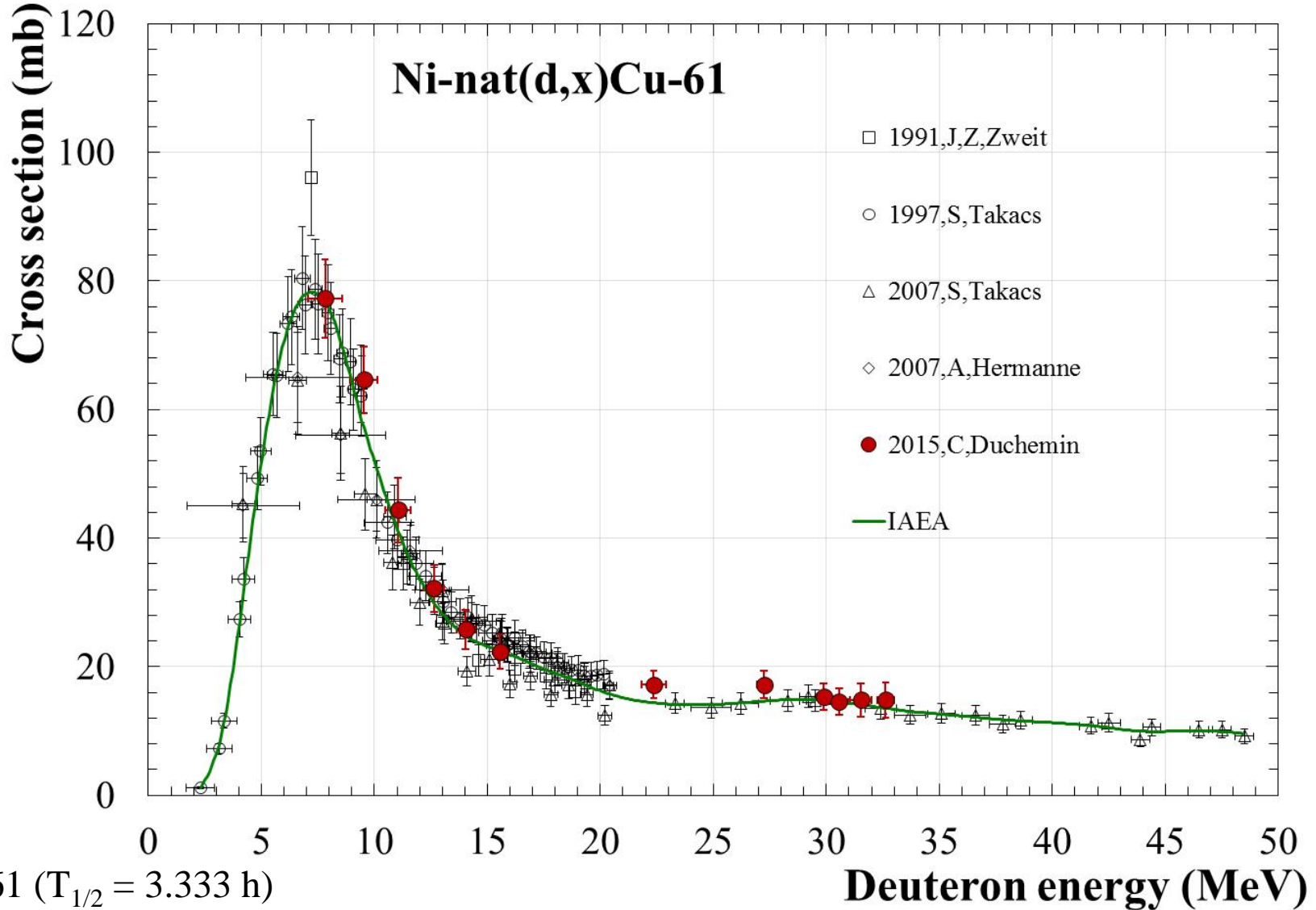


Monitor reaction



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

Monitor reaction



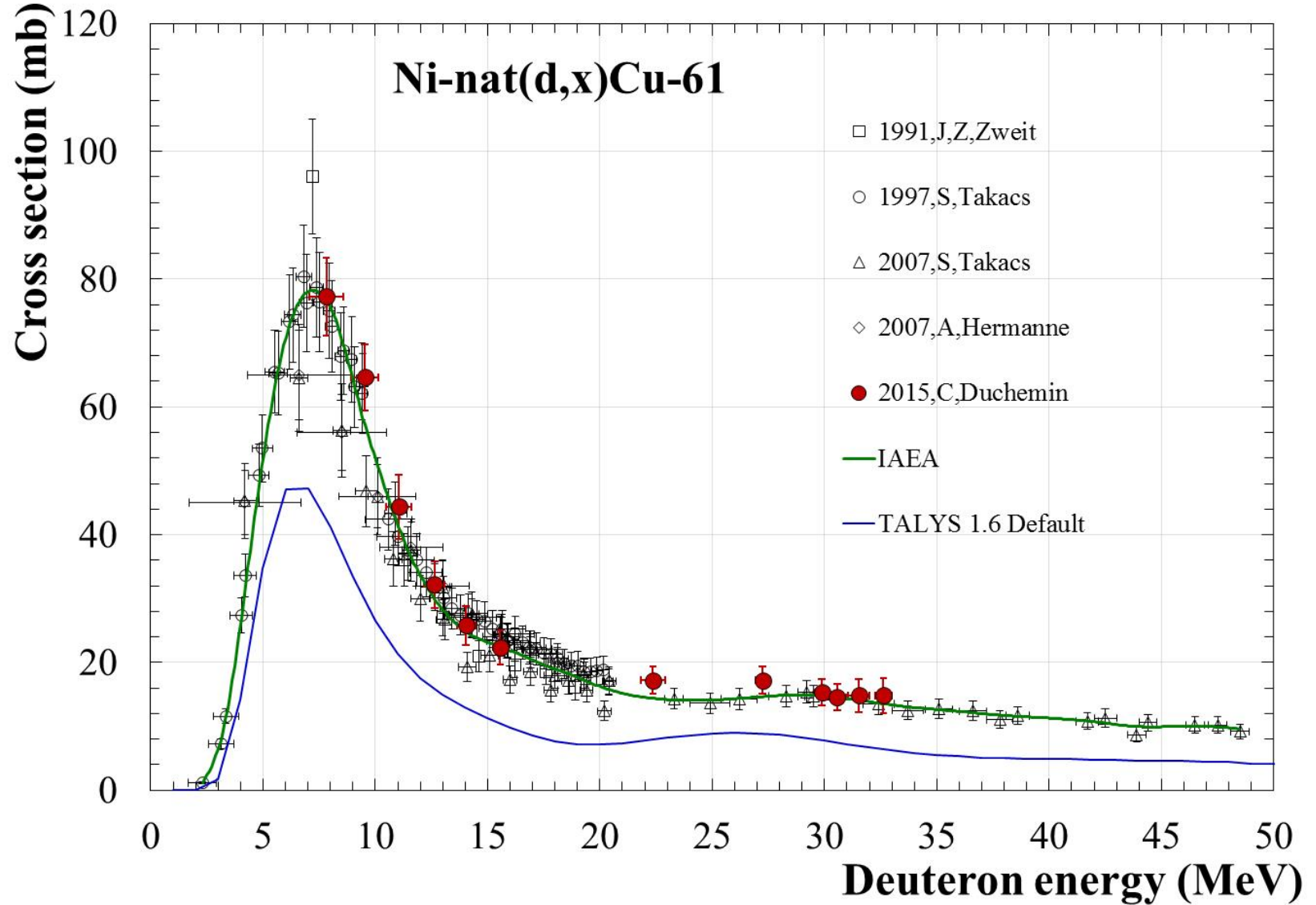
Cu-61 ($T_{1/2} = 3.333$ h)

EC β^+ processes to stable Ni-61

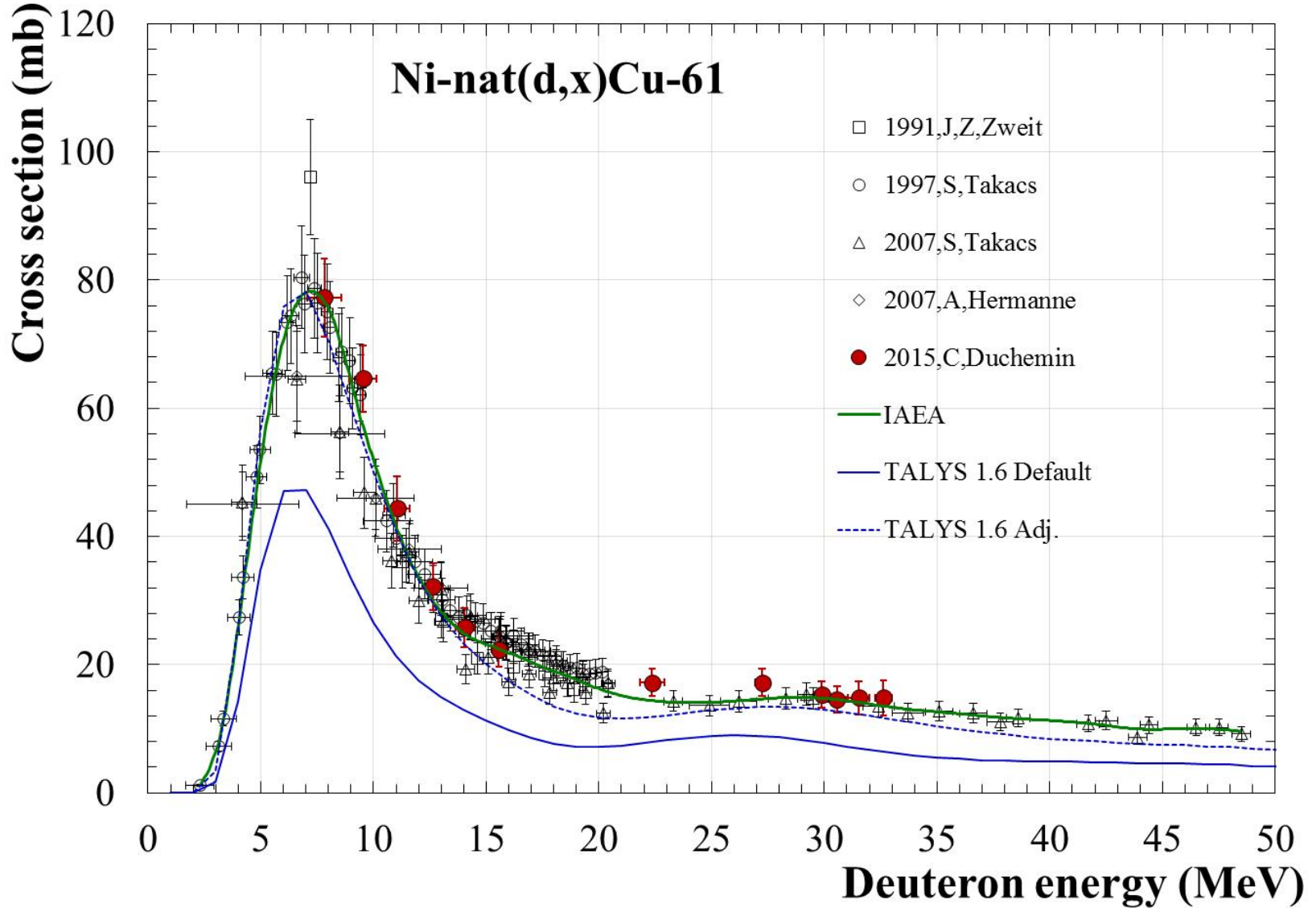
Suitable for PET imaging

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

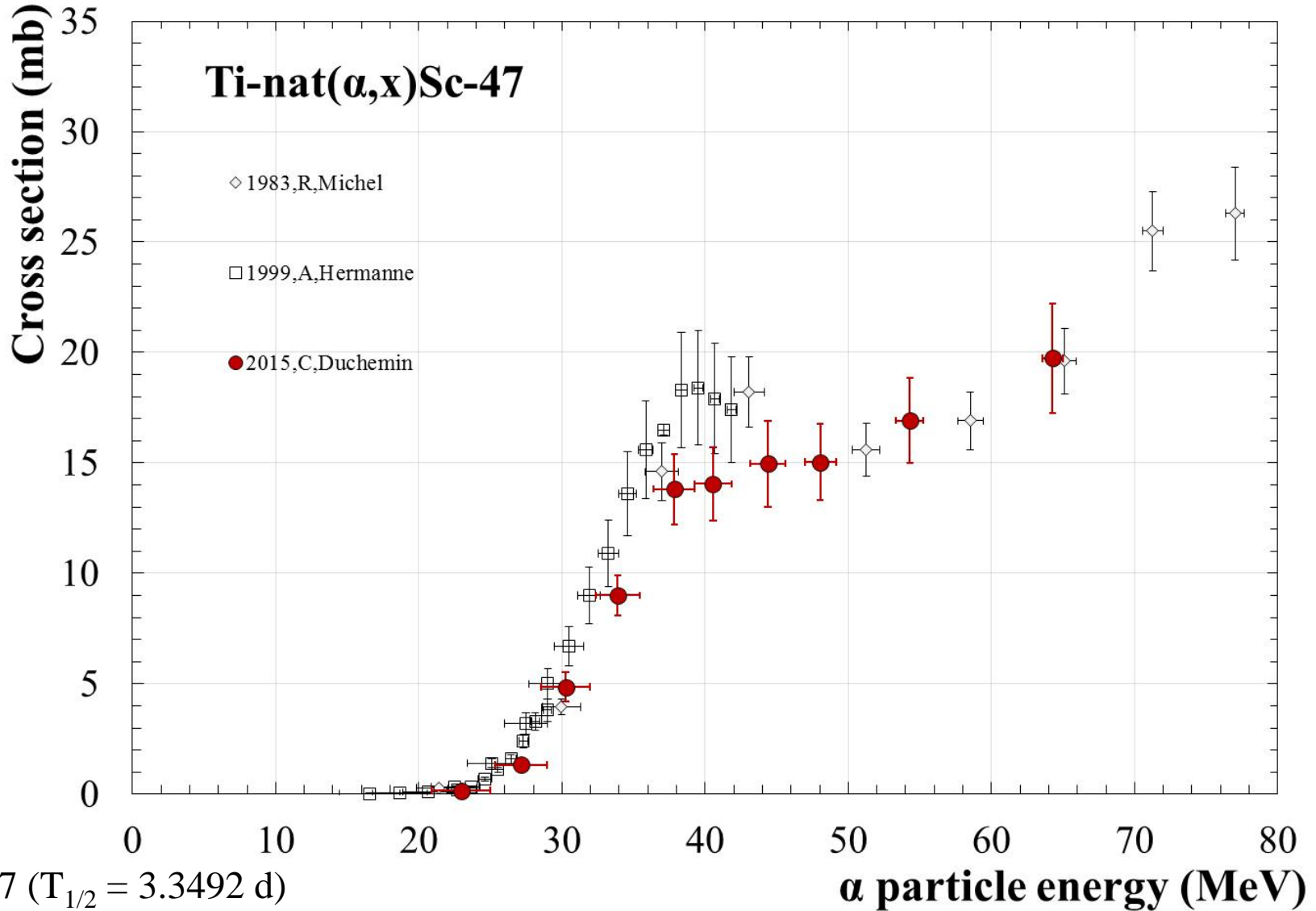
Monitor reaction



Monitor reaction



Monitor reaction

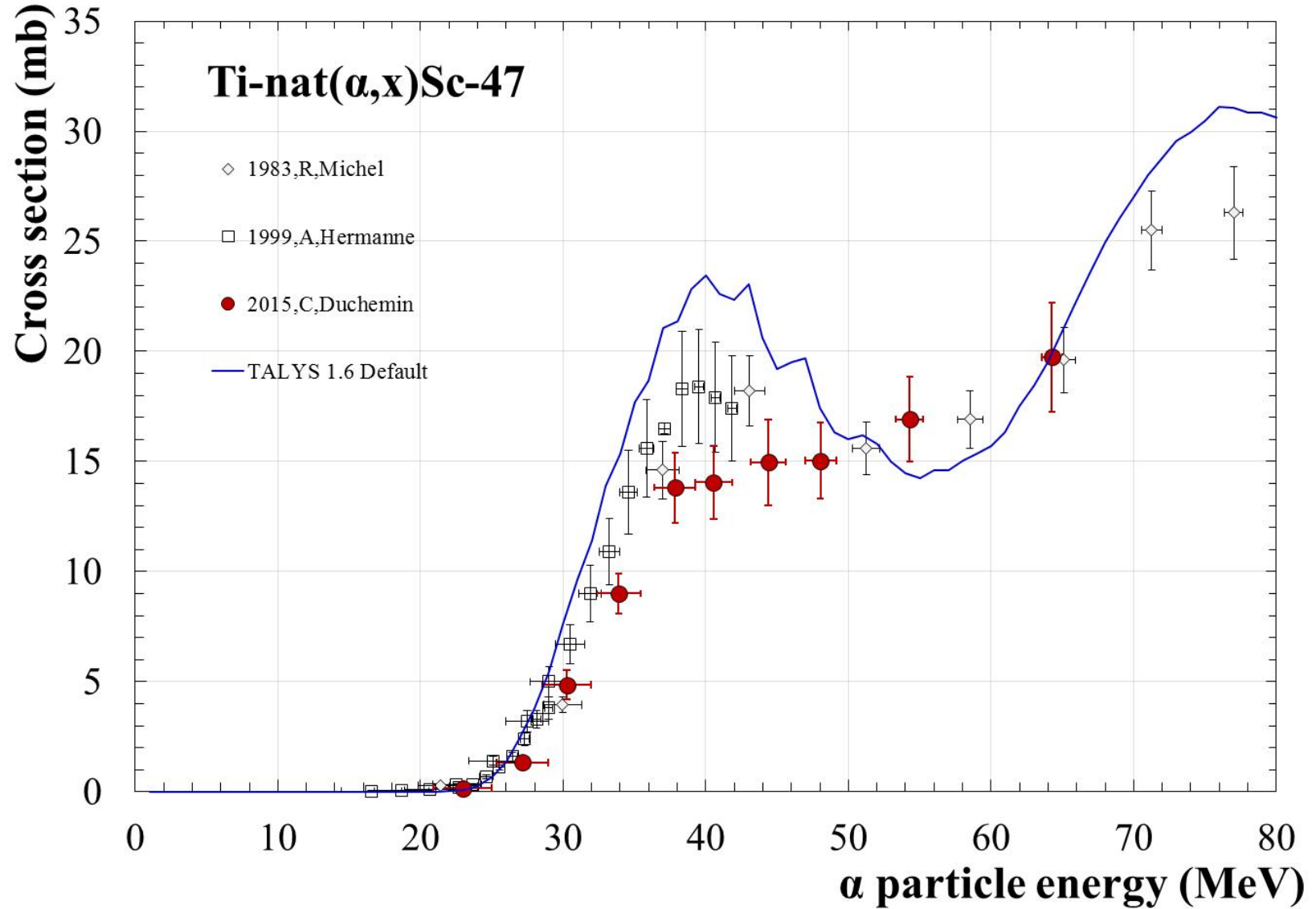


Sc-47 ($T_{1/2} = 3.3492$ d)
 β^- (100%) to stable Ti-47

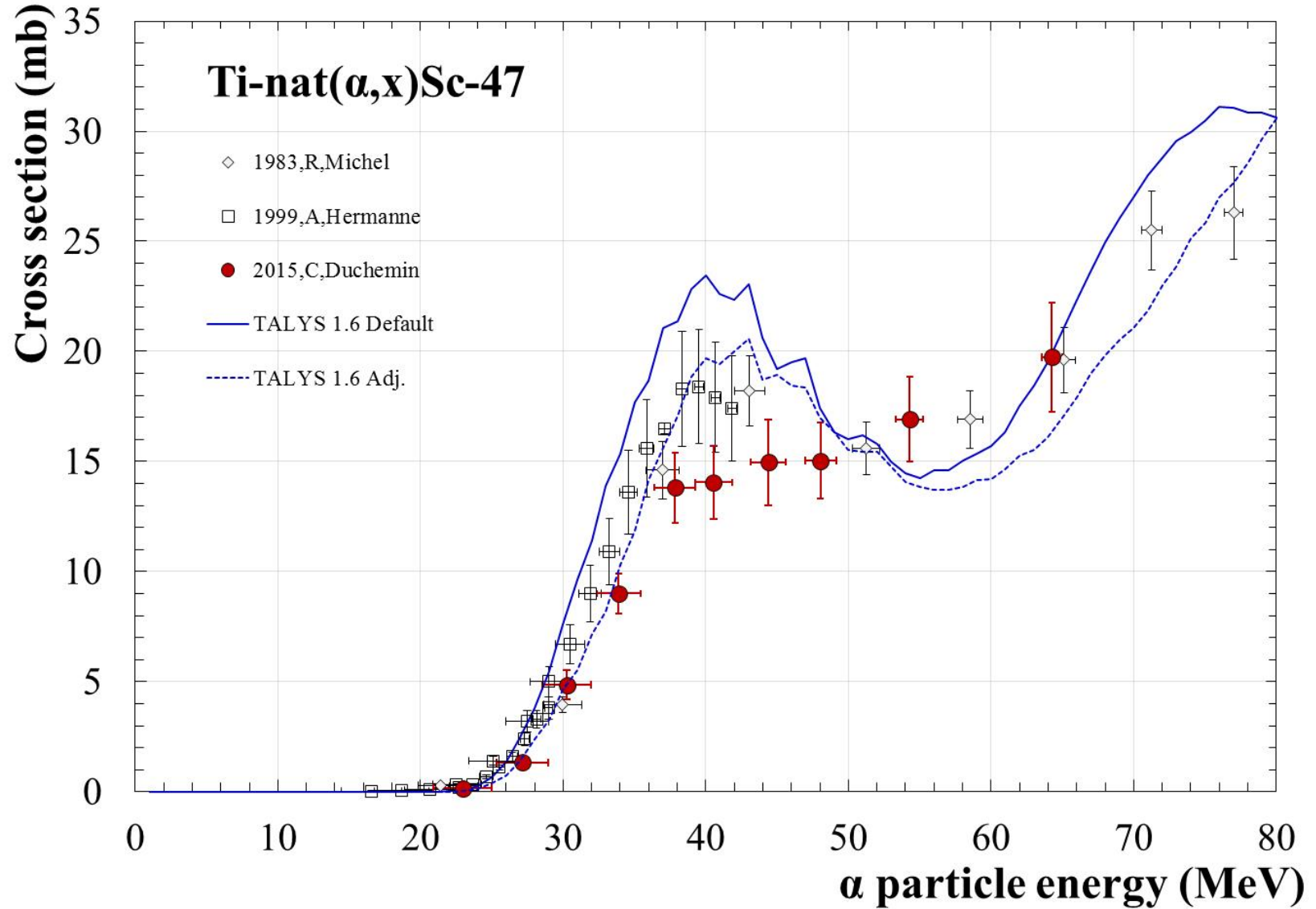
Suitable for theranostic approach with the Sc-44

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

Monitor reaction



Monitor reaction



- 
- # 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.¹, Duchemin C.^{1,2}, Haddad F.^{1,3}, Michel N.^{1,3}, Métivier V.¹

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