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

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

UNIVERSITÉ DE NANTES

MINES Nantes
Graduate School of Engineering

Outline

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

Motivations

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

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Experimental facility

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

C70 Cyclotron build by IBA:

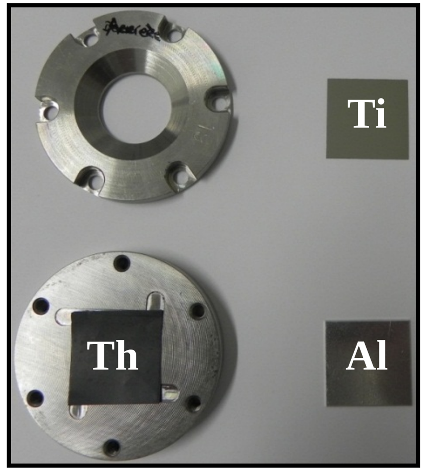
- **4 sectors** isochron **cyclotron**
- **2 multi-particle sources**:
  - H^-,D^-: multicusp
  - He^{2+},HH^+: supernanogan ECR
- **2 extraction lines**:
  stripper or electrostatic deflector

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

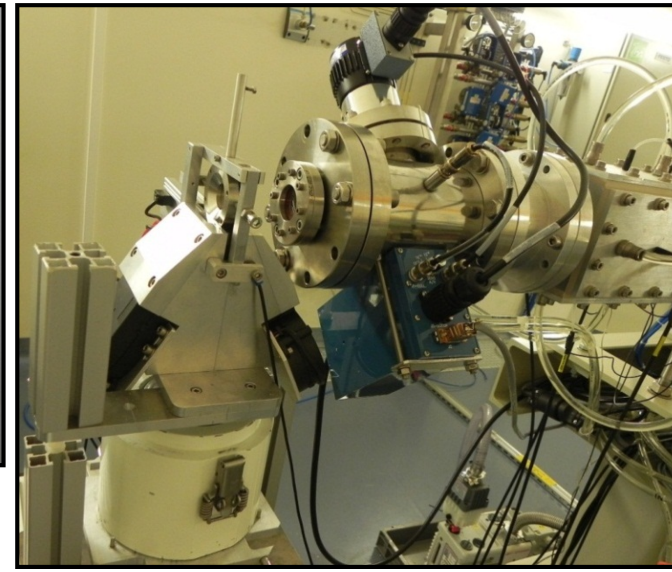
Experimental set-up

Stacked-foil technique:

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



Capsule and foils



Irradiation station and beam line

Activity and cross section:

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

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:

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

- error on e, e': ≤ 1%
- error on t: negligible

IAEA recommended cross sections:

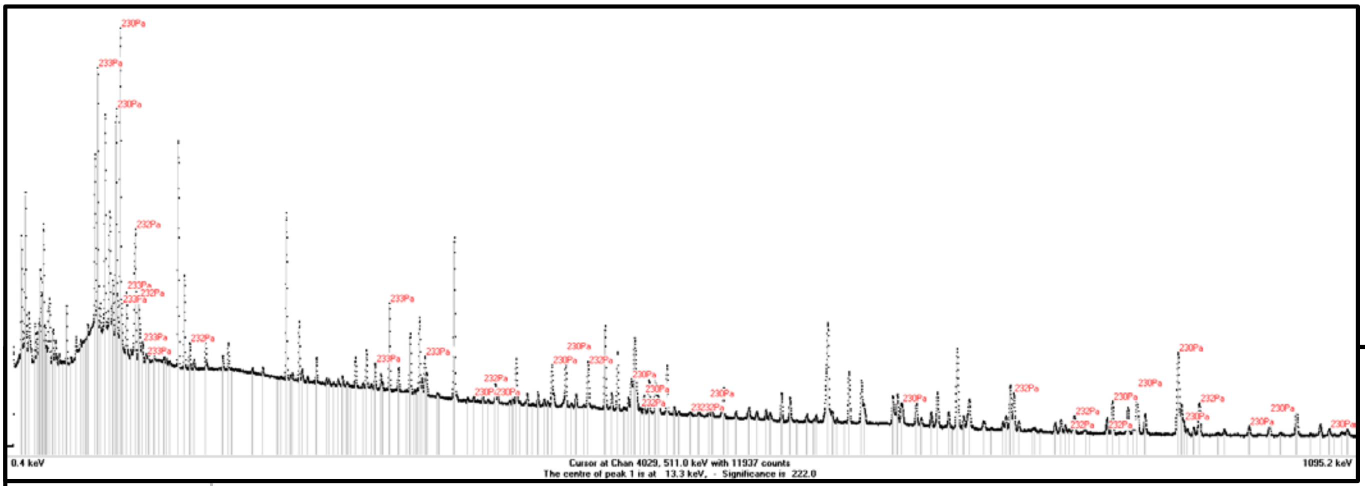
- 8 reactions available for protons
  ^{27}Al (2), ^{nat}Ni, ^{nat}Ti and ^{nat}Cu (4)
- 5 reactions available for deuterons
  ^{27}Al (2), ^{nat}Fe, ^{nat}Ni and ^{nat}Ti
- 6 reactions available for alpha-particles
  ^{27}Al (2), ^{nat}Ti and ^{nat}Cu (3)

Experimental set-up

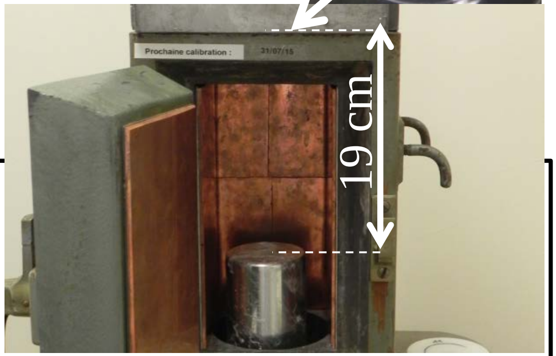
Gamma spectroscopy

- **HPGe** coaxial detector
- Dead time: < 10% (sum peak)
- Activity values: FitzPeaks
- $T_{1/2}$, E_γ, I_γ: Lund/LBNL, NNDC

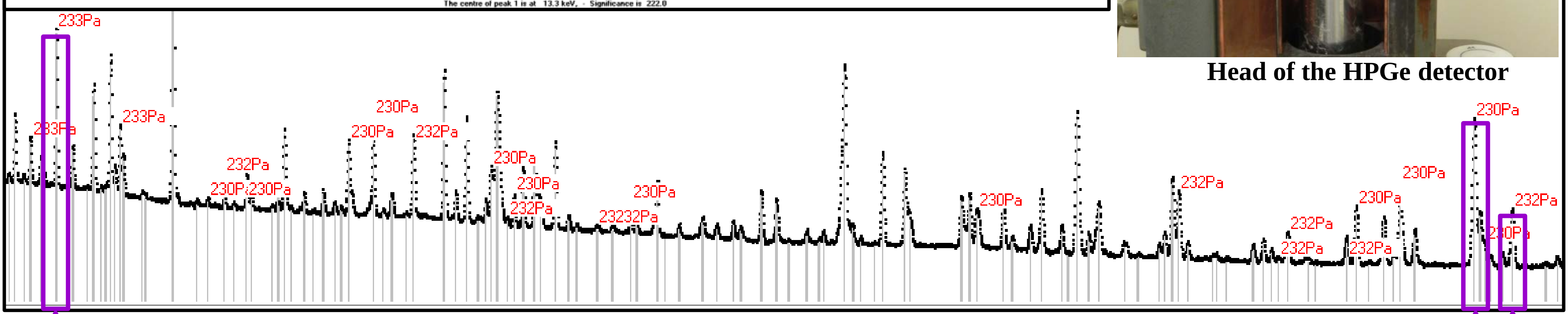
- γ spectra recorded on **8192 channels**
- FWHM: 1.04 keV at 122 keV (^{57}Co)
  1.97 keV at 1332 keV (^{60}Co)
- Energy and efficiency calibrations: Co and Eu



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}Pa

Collected data sets

Proton induced reactions:

Ac-225 from Th-232(p,x) — C. Duchemin et al, Phys Med Biol **60** (2015) **931-946**
Ra-223 from Th-232(p,x) — C. Duchemin et al, Phys Med Biol **60** (2015) **931-946**
Fission fragment distribution from Th-232(p,x)* — To be published
Monitor reactions on Ti, Ni and Cu — 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) — C. Duchemin et al, Phys Med Biol **60** (2015) **6847-6864**
Tb-155 New data set for Gd-nat(d,x) — Submitted to Appl Radiat Isot
Re-186g New data set for W-186(d,x) — C. Duchemin et al, Appl Radiat Isot **97** (2015) **52-58**
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, Appl Radiat Isot **103** (2015)**160-165**

Alpha induced reactions:

Sn-117m from Cd-116(α,x) — C.Duchemin et al, Appl Radiat Isot **115** (2016) **113-124**
Monitor reactions on Cu, Ti, Ni

* S415, Ambassadeur, 15/09/16, 15:45 and P120, Witte Roos 8:30-18:10
V. Métivier et al., Thorium-232 fission induced by light charged particles up to 70 MeV

TALYS

Code for the simulation of nuclear reactions

Many state-of-the-art nuclear models

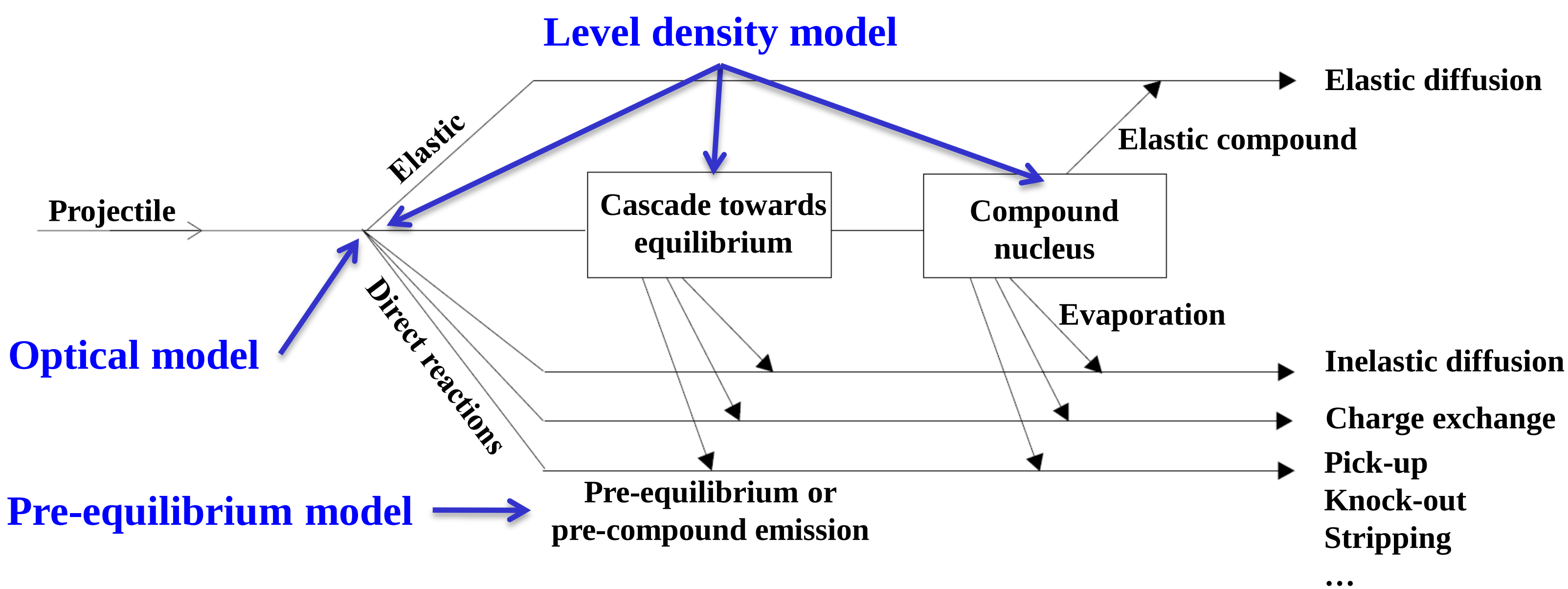
Provide a complete description of all reactions channels and observables

Projectiles : n, p, d, t, He-3, α particles

Energy : 1 keV to 1 GeV

Targets : Z = 3 to 110

Nuclear reactions



Adapted from S. Benck, PhD thesis, 1999

⇨ Influence on the calculated production cross section values

Koning A.J. and Rochman D., Nucl. Data Sheets, 113, 2012

TENDL2015

TENDL2015

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

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

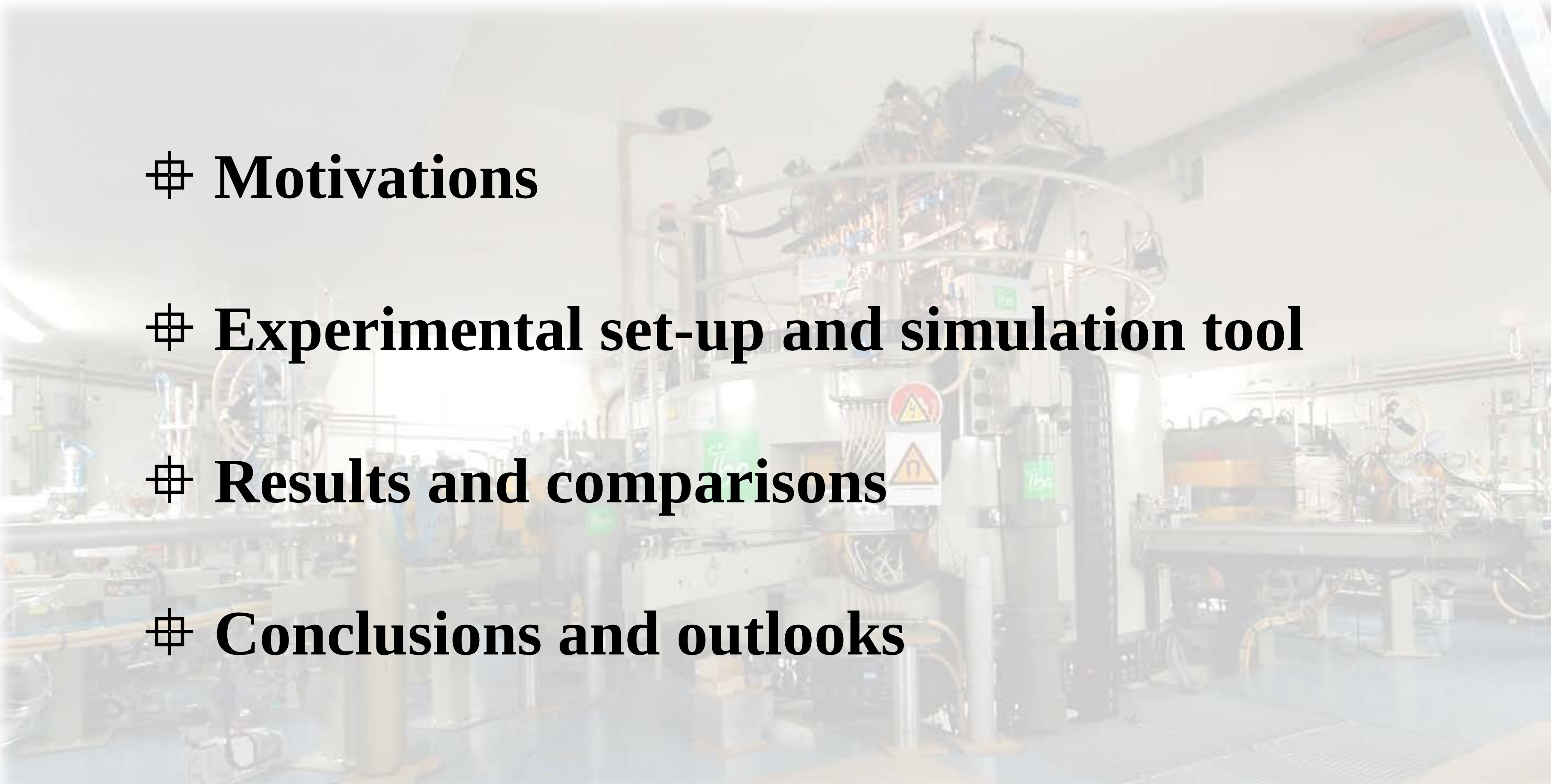
[1] IAEA, [2] PSI,[3] JUKO Research, [4]CCFE, [5]Uppsala Univ., [6]NRG, [7]CEA,[8]Vattenfall, [9]NEA, [10]ULB, [11]VTT, [12]ATI, [13]IRMM, [14]NNL.

TENDL2015 contains evaluations for :

- seven types of incidents 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, ^{9}Be, ^{14,15}N, ^{16}O and ^{19}F 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

Outline



- Motivations
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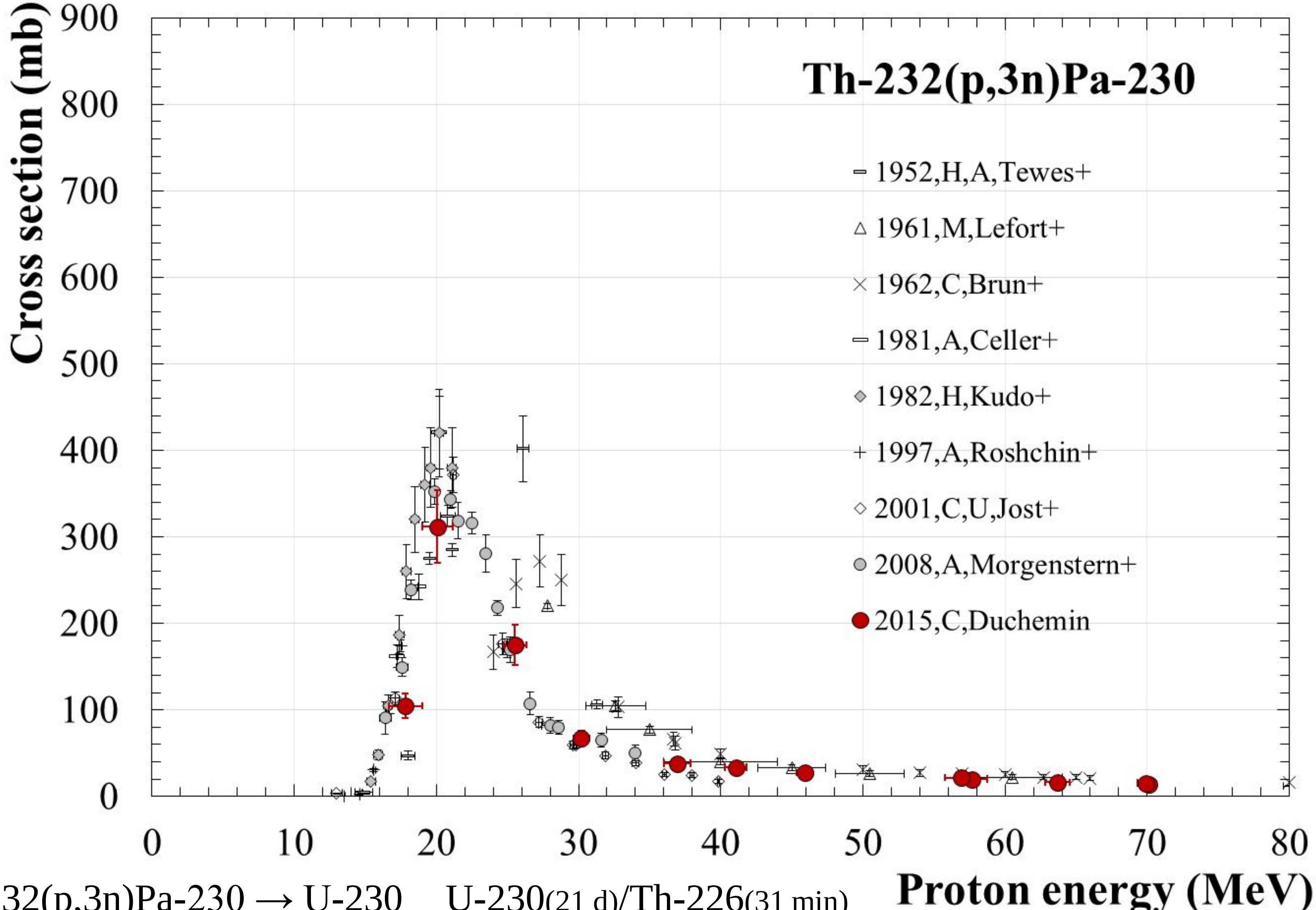
- Results and comparisons
- Conclusions and outlooks

Novel therapeutic nuclide

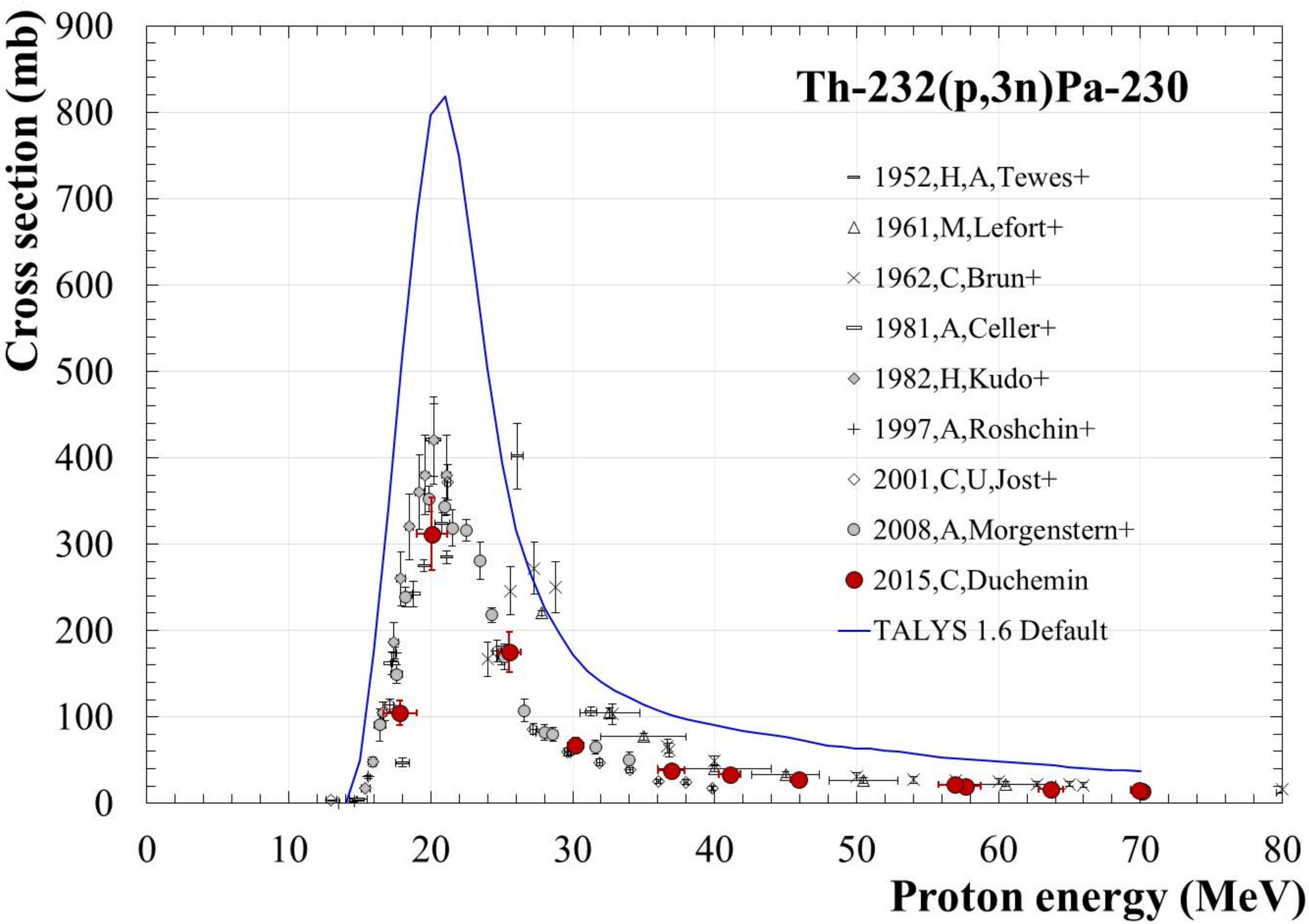


Th-232(p,3n)Pa-230 → U-230 U-230(21 d)/Th-226(31 min)

α RIT for leukaemia treatment 4 α cascade of 27.7 MeV

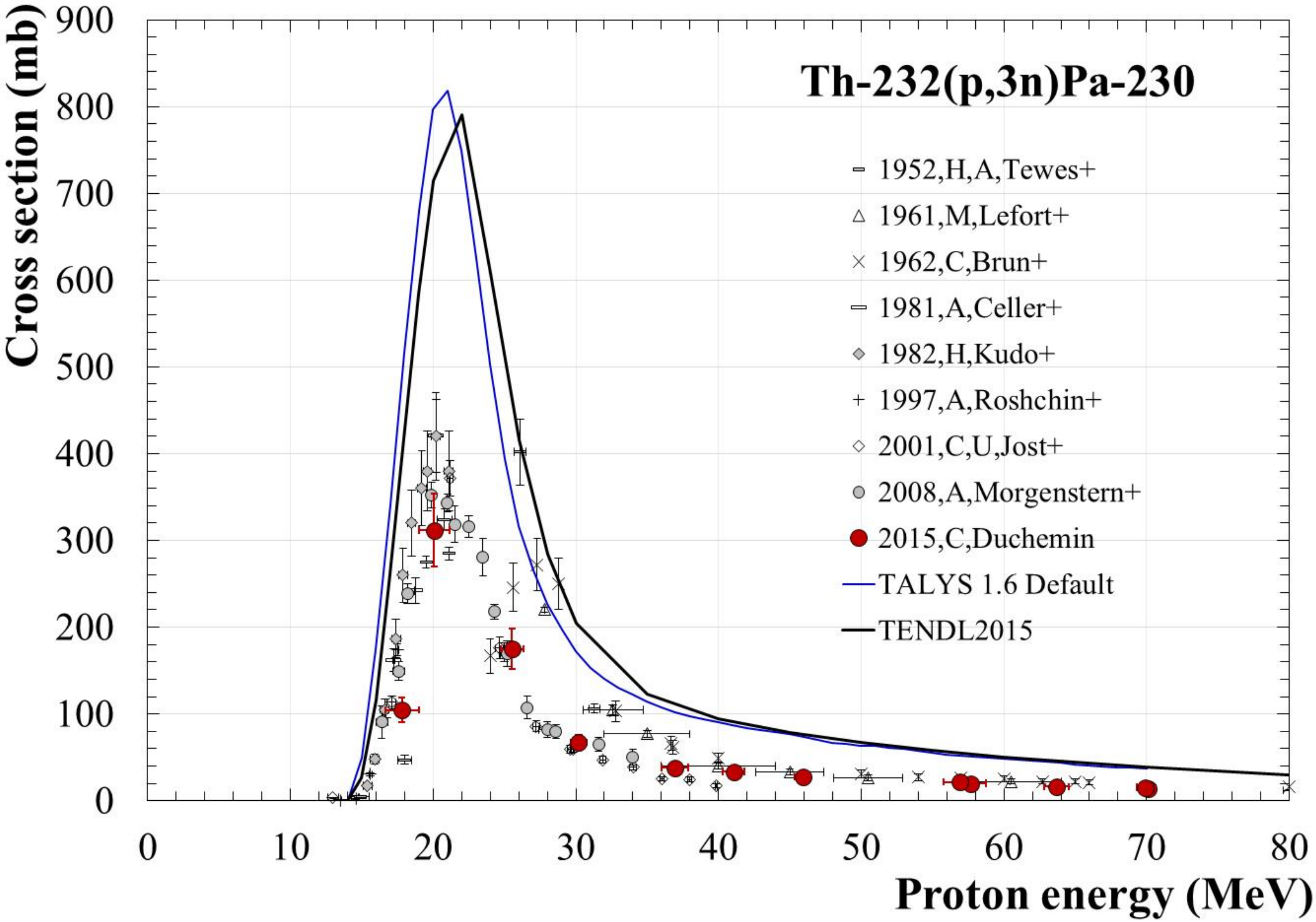
C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946

Novel therapeutic nuclide



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

Novel therapeutic nuclide



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

TALYS

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

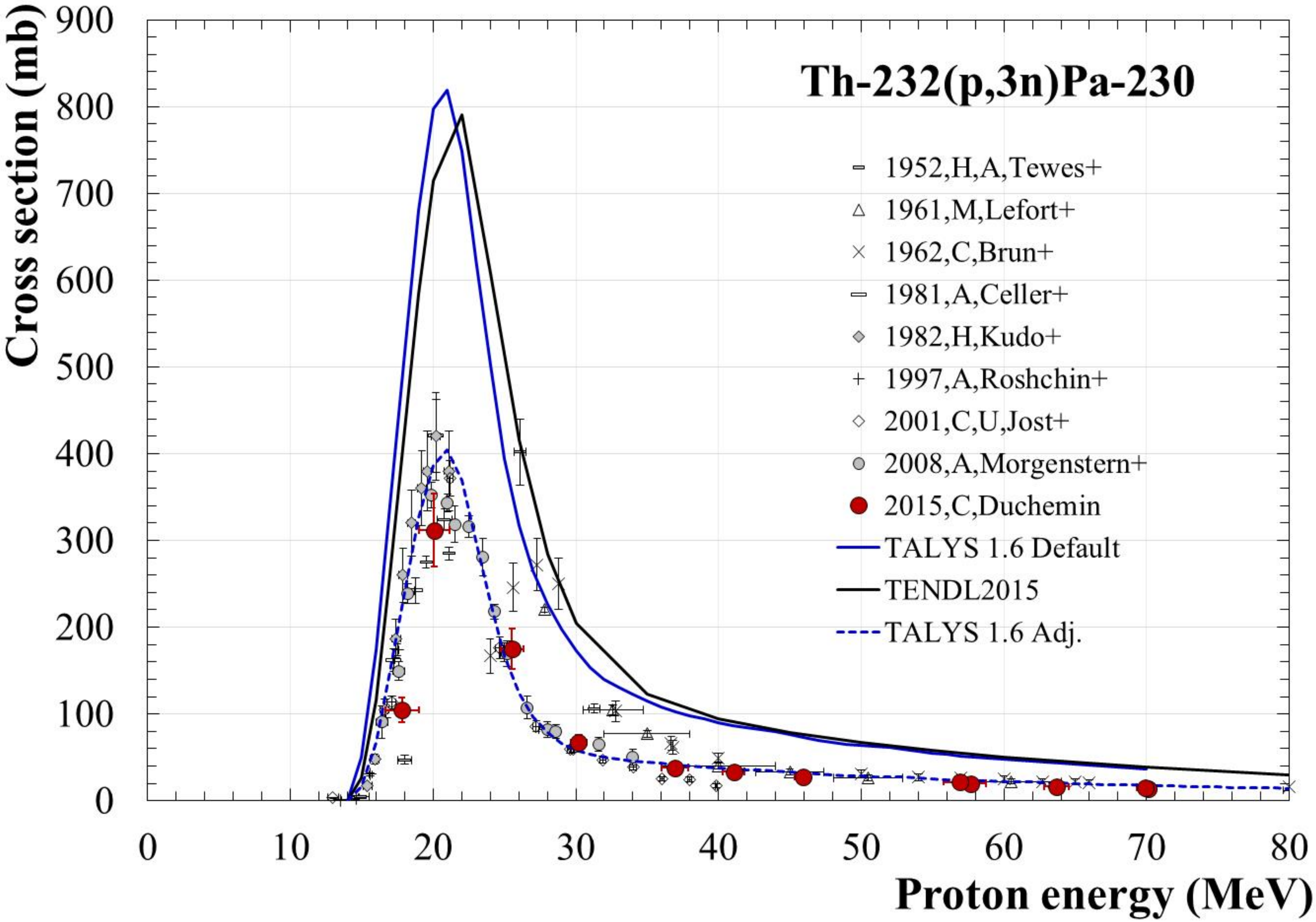
⇨TALYS 1.6 Default

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.

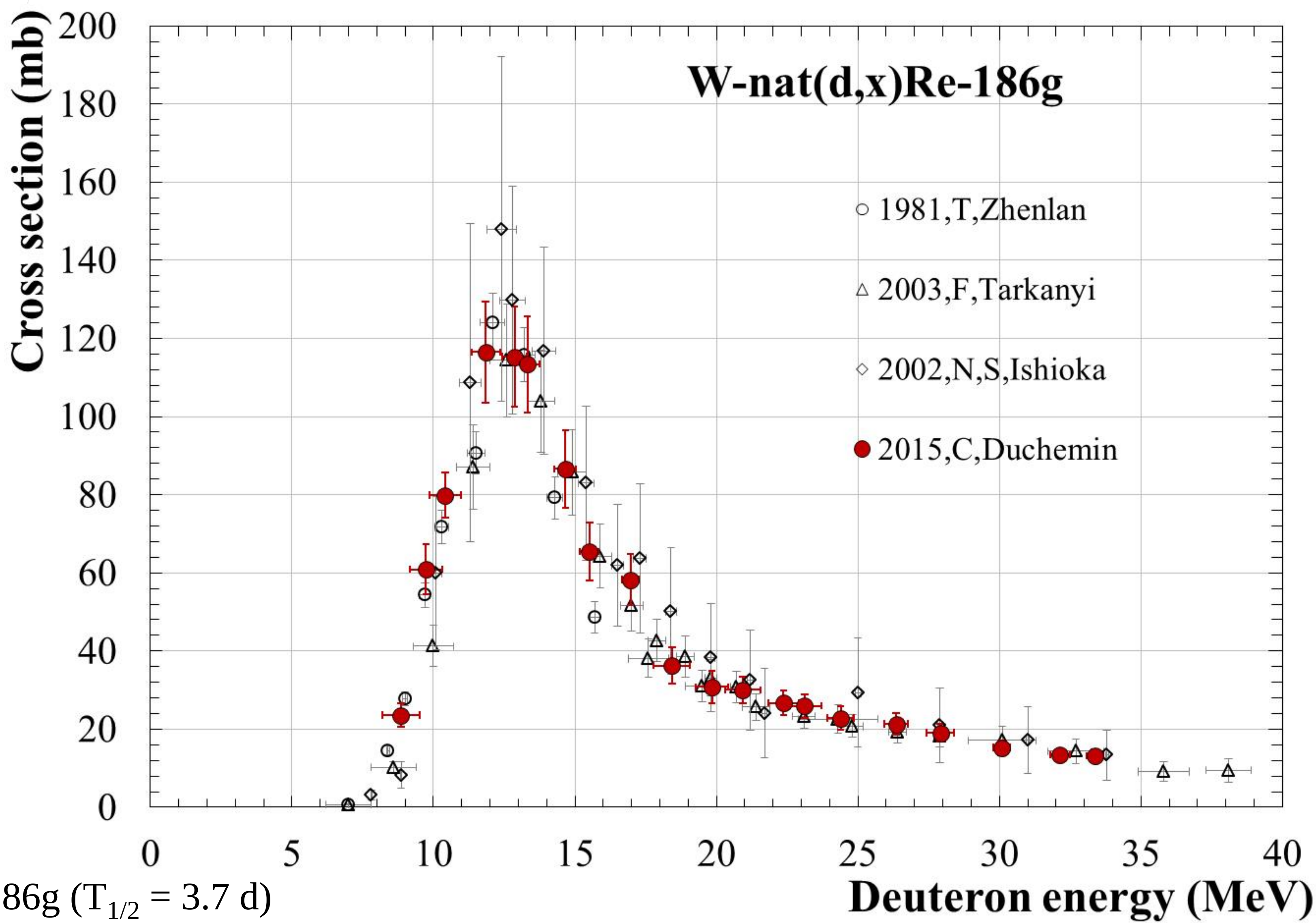
Models	Projectile	Default	Adj.
Optical	p (1)	A.J. Koning and J.P. Delaroche (2003)	A.J. Koning and J.P. Delaroche (2003)
	d (5)	S. Watanabe (1958) …	Y. Han et al. (2006) …
	α (5)	L. McFadden and G.R. Satchler (1966) …	Demetriou et al. (2002) …
Pre-equilibrium	All (4)	2 components exciton A.J. Koning 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

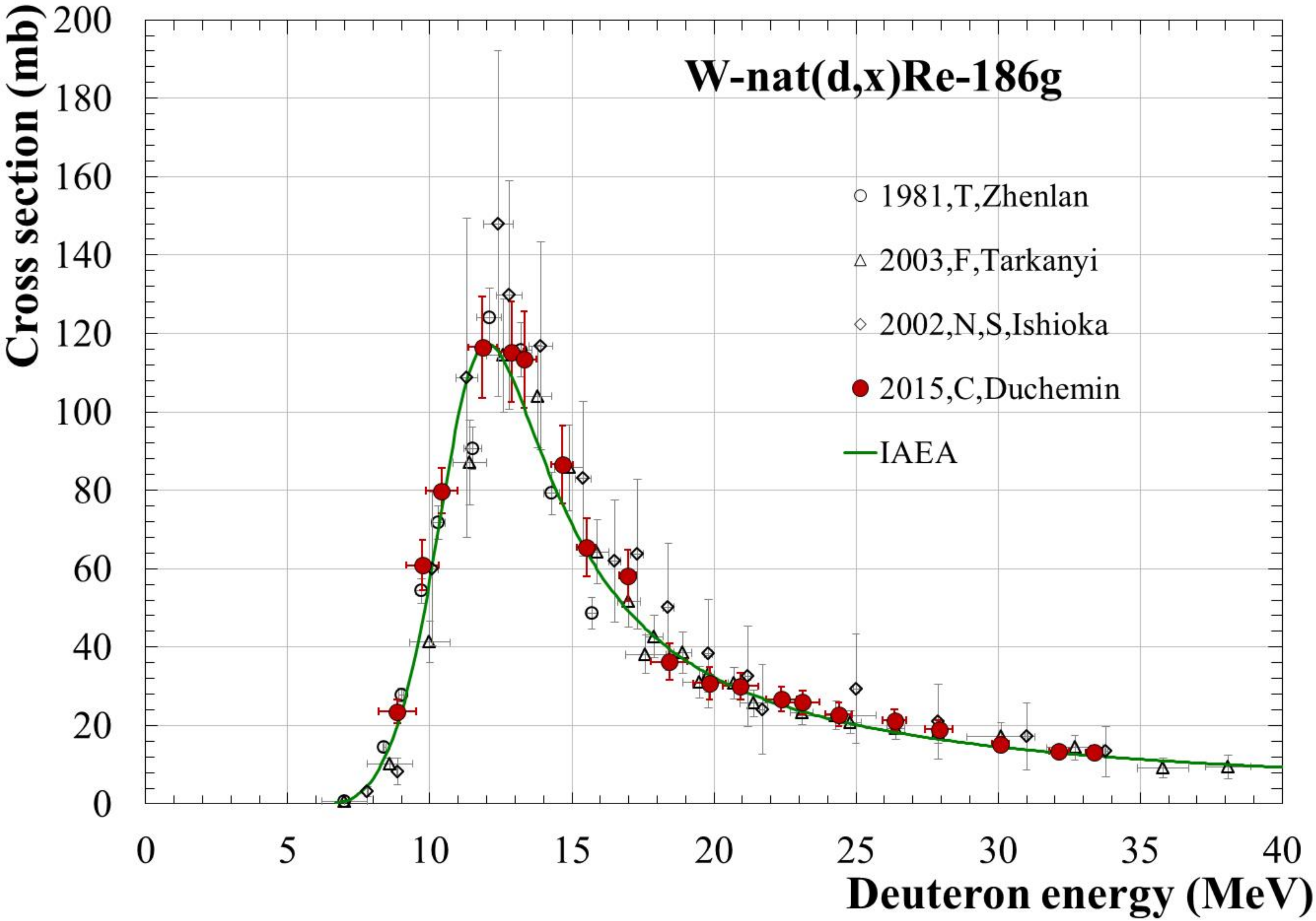


Re-186g ($T_{1/2}$ = 3.7 d)

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

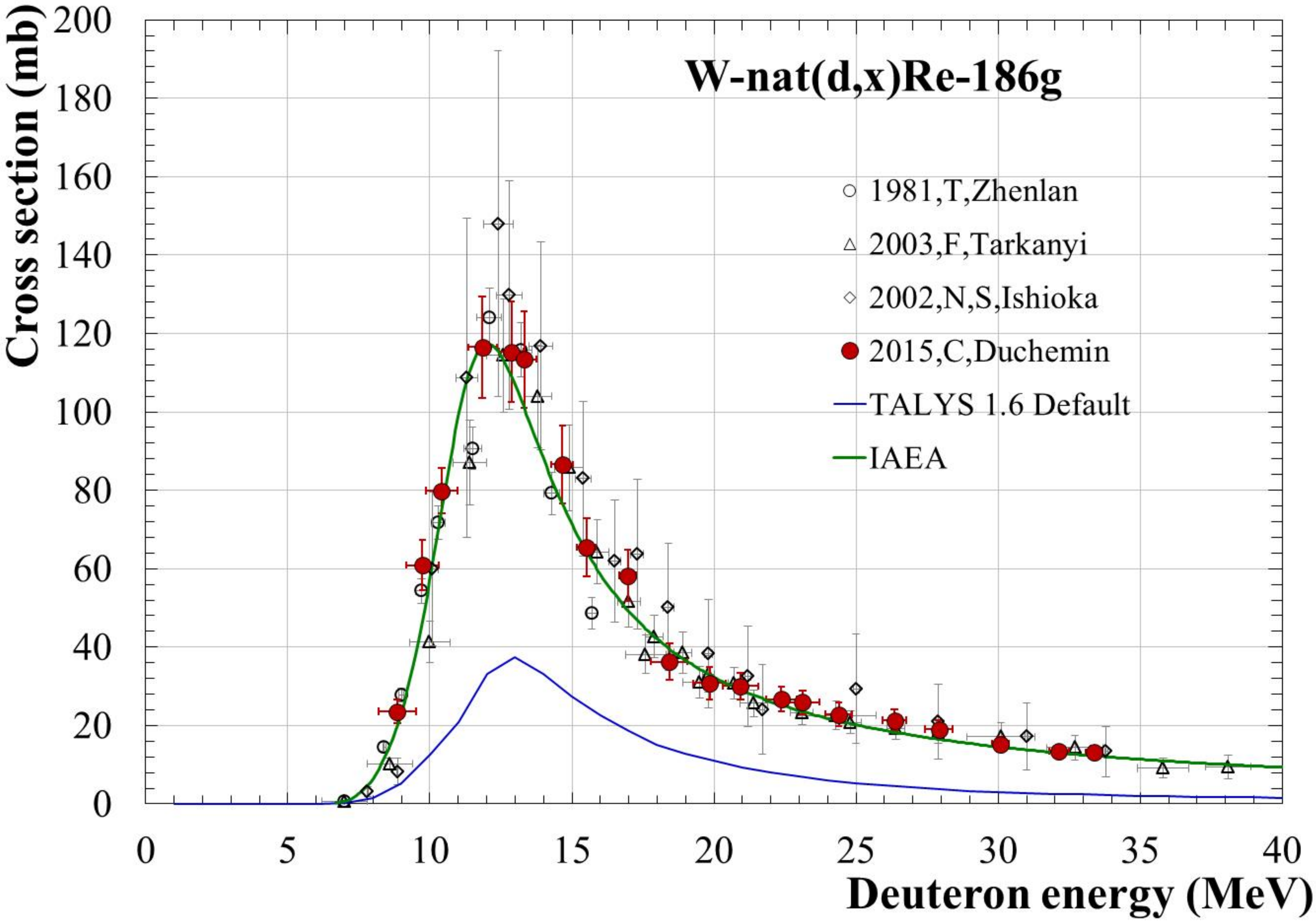
C. Duchemin et al., Appl. Radiat. Isot. 97 (2015) 52-58

Novel therapeutic nuclide



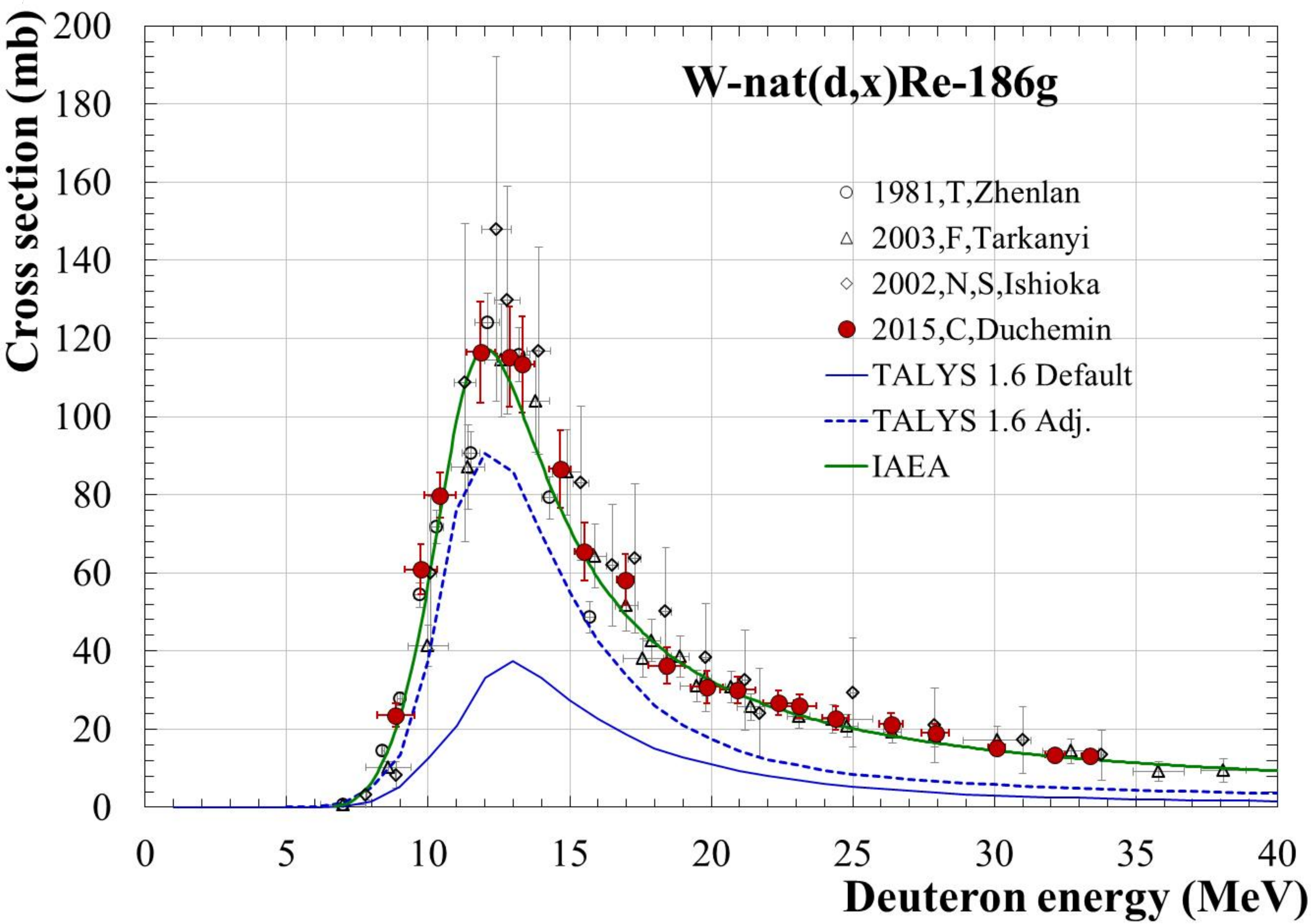
C. Duchemin et al., Appl. Radiat. Isot. 97 (2015) 52-58

Novel therapeutic nuclide



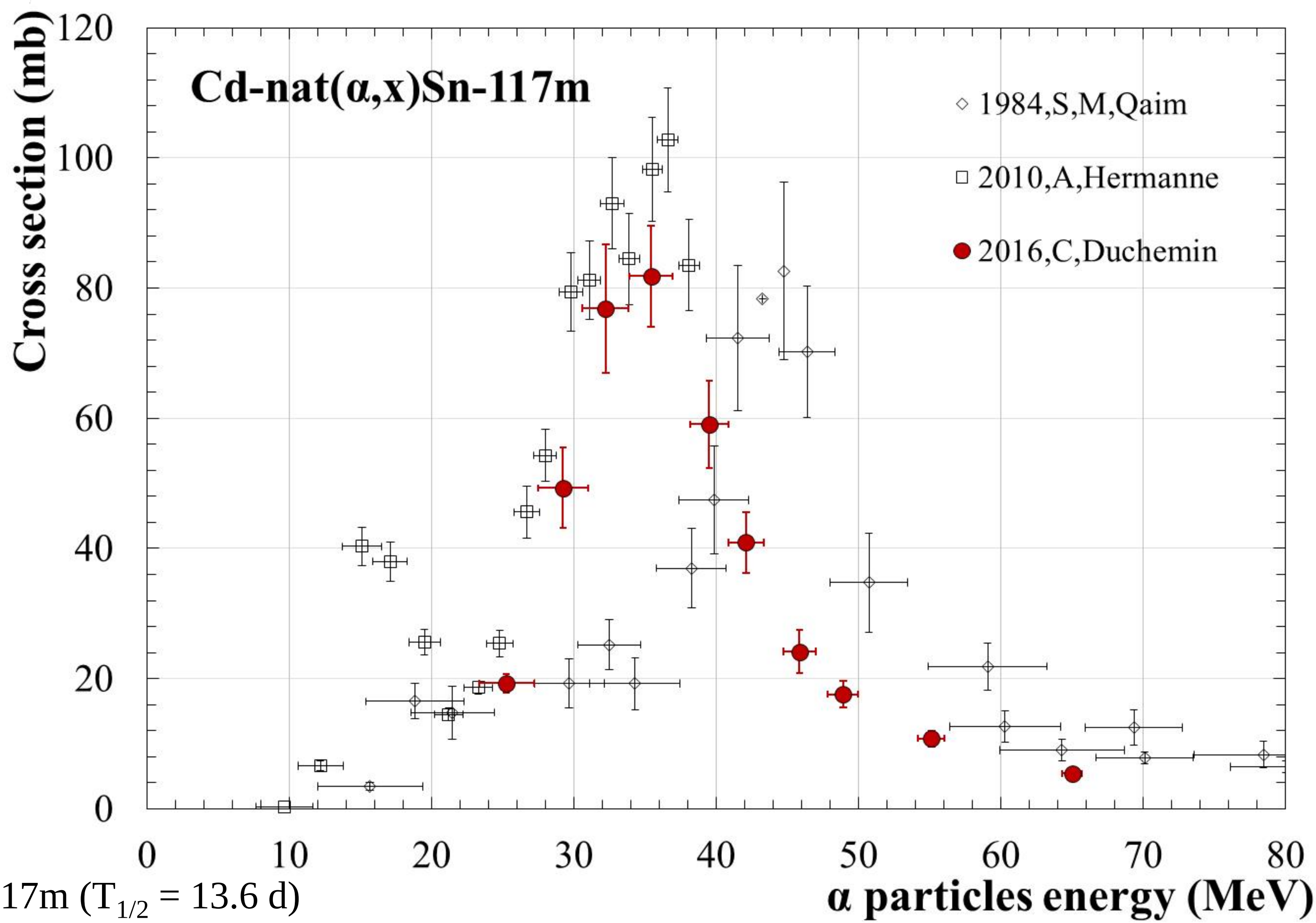
C. Duchemin et al., Appl. Radiat. Isot. 97 (2015) 52-58

Novel therapeutic nuclide



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

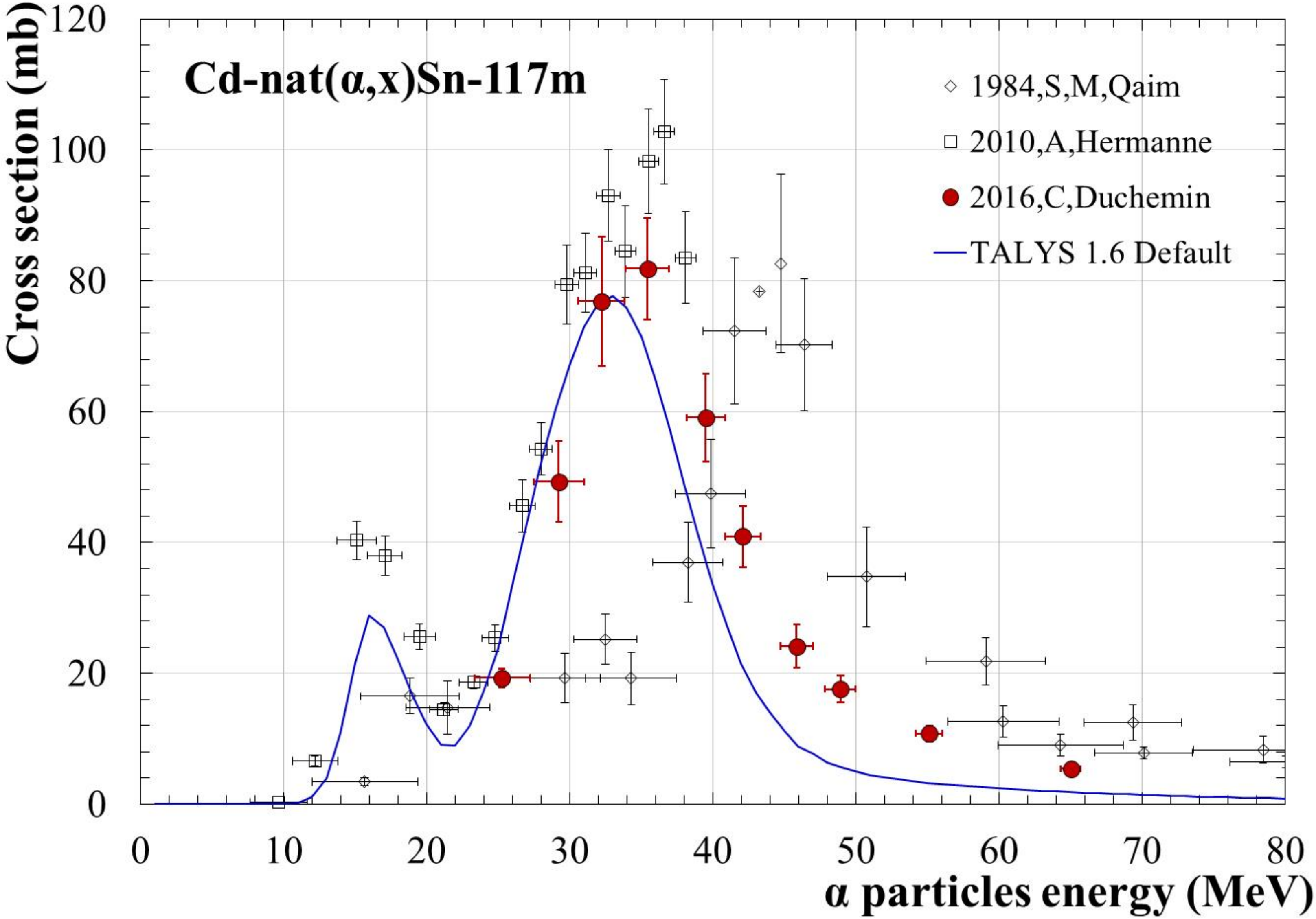
Novel therapeutic and imaging nuclide



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

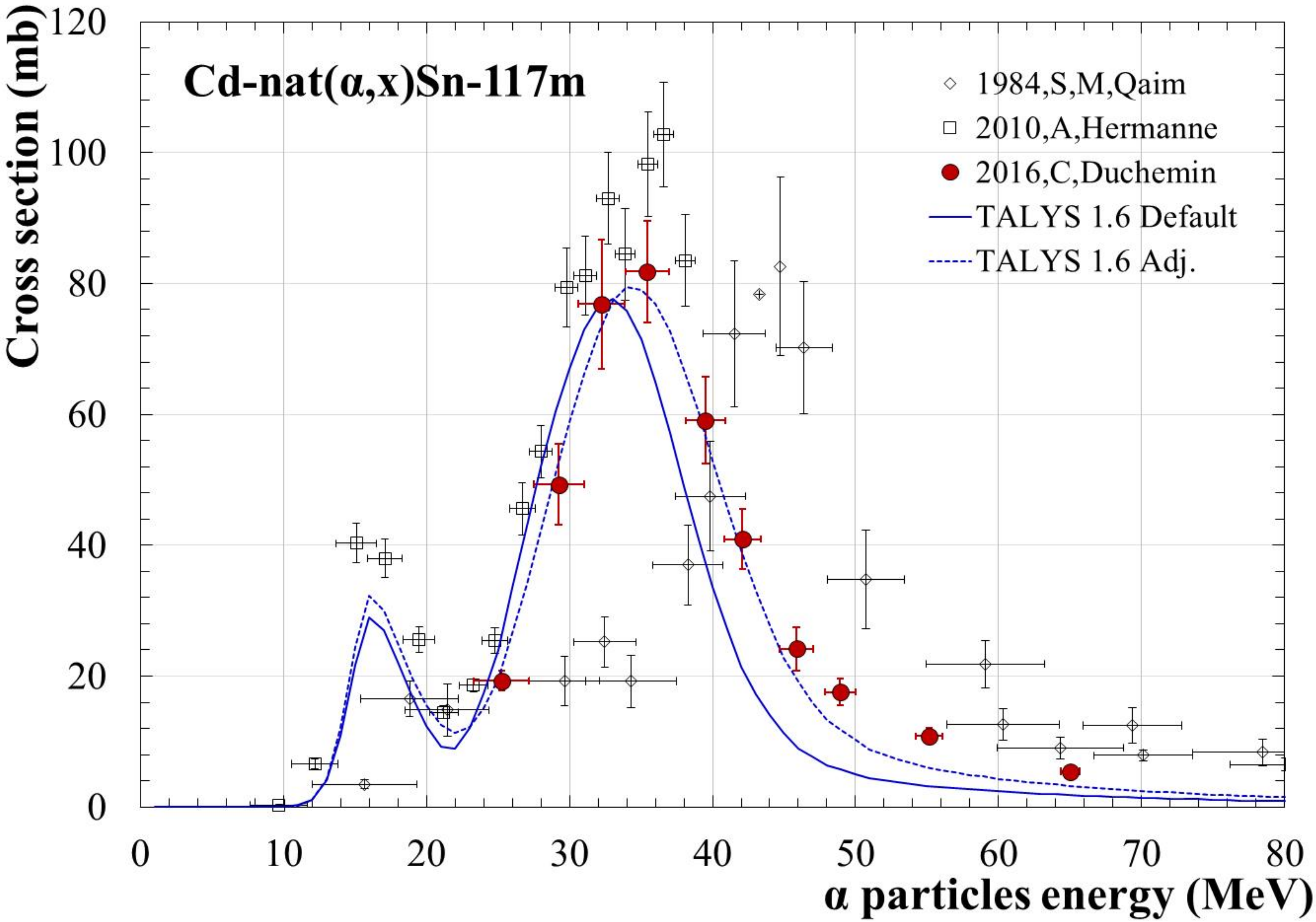
C. Duchemin et al., Appl. Radiat. Isot. 115 (2016) 113-124

Novel therapeutic and imaging nuclide



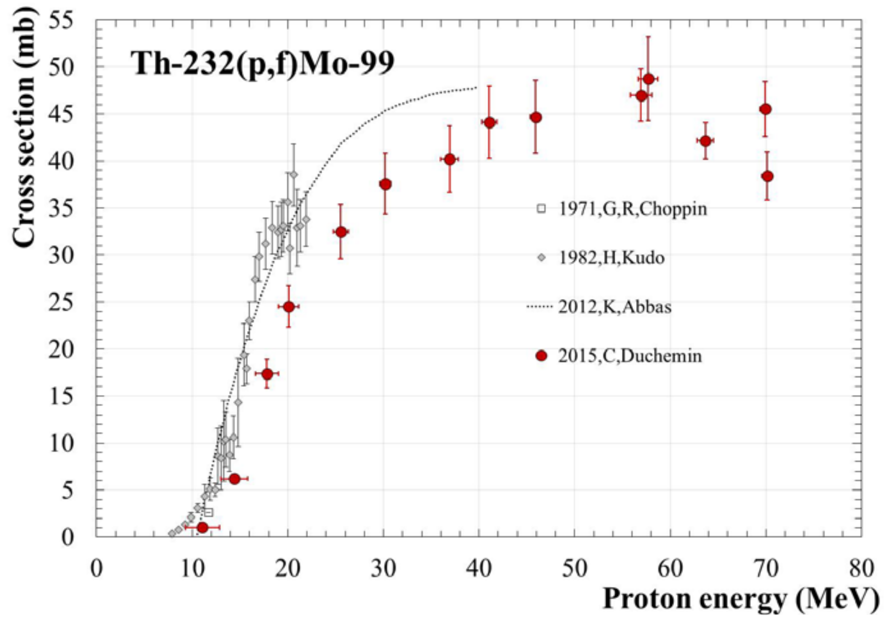
C. Duchemin et al., Appl. Radiat. Isot. 115 (2016) 113-124

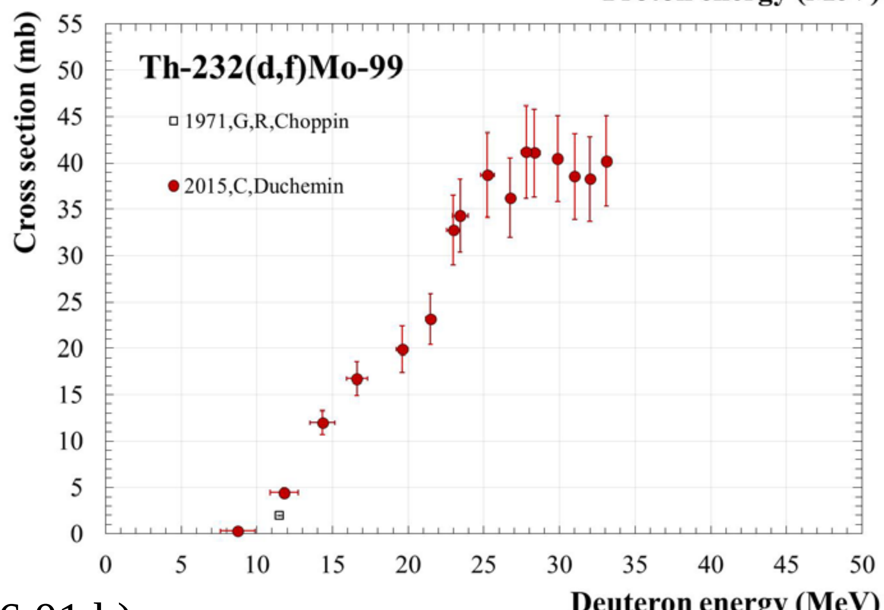
Novel therapeutic and imaging nuclide



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

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

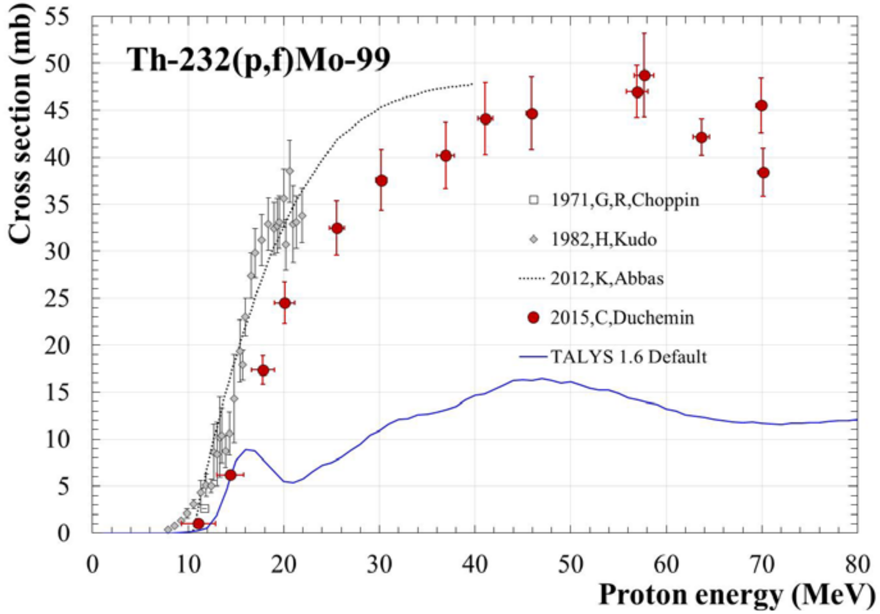


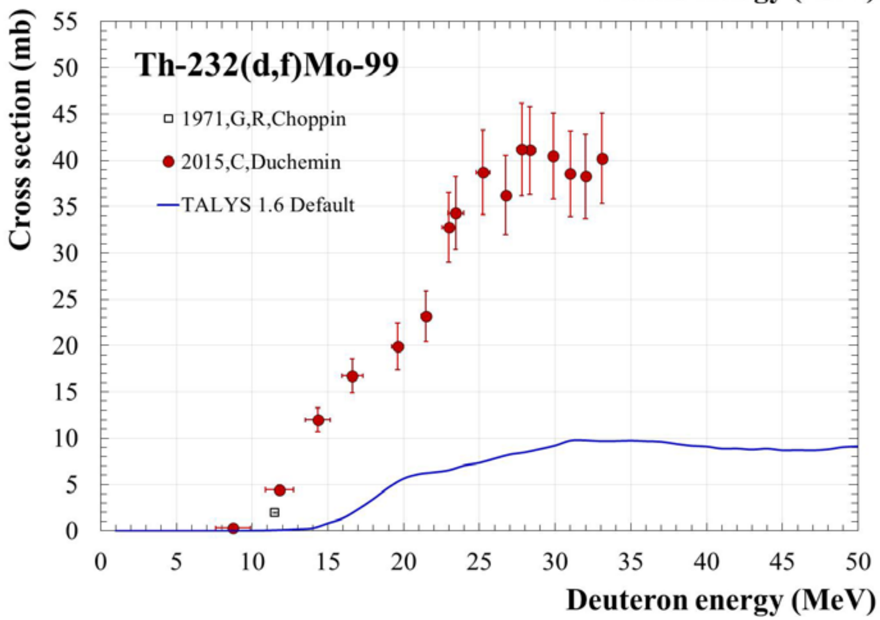


Mo-99(65.94 d)/Tc-99m(6.01 h)

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

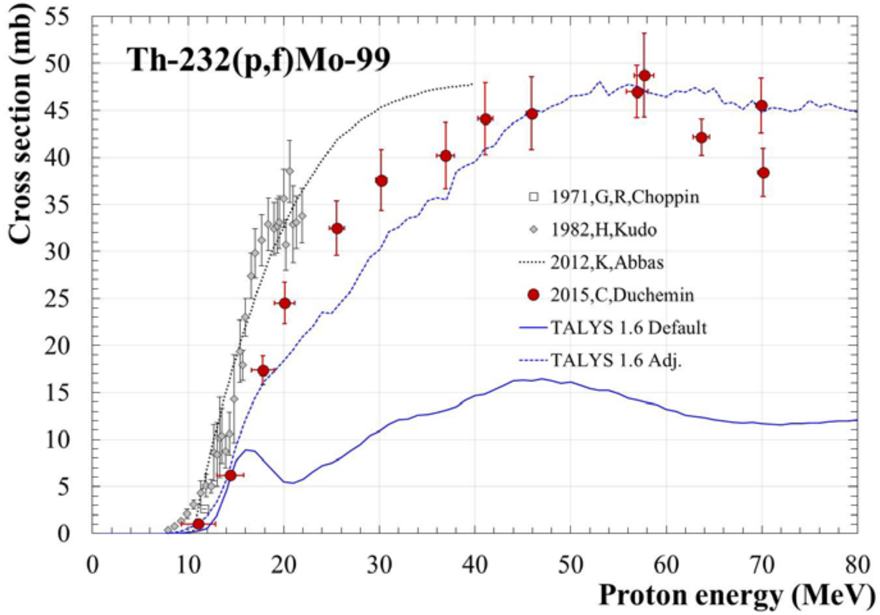
Potential method for production of $^{99}Mo/^{99m}Tc$

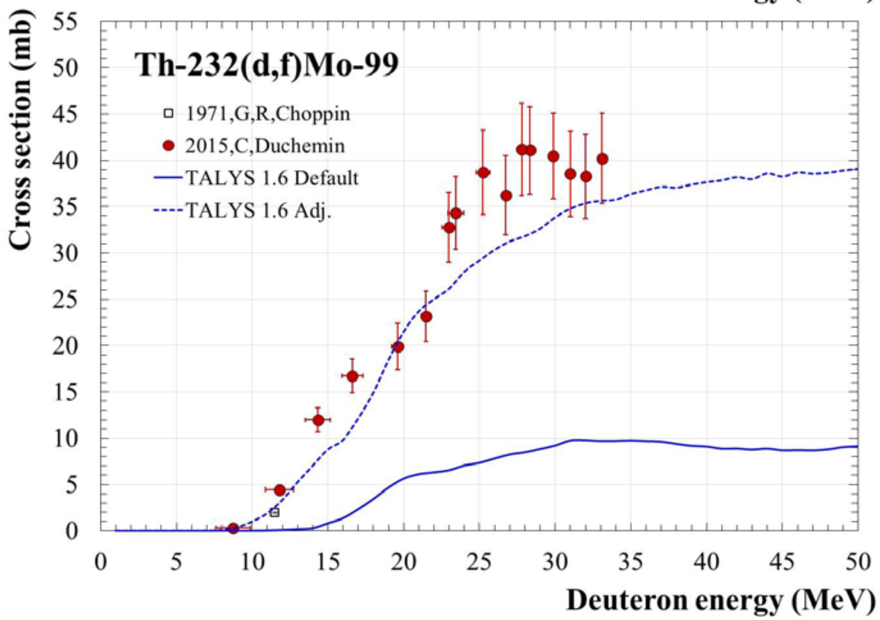




C. Duchemin et al., Phys. Med. Biol. 60 (2015) 931-946

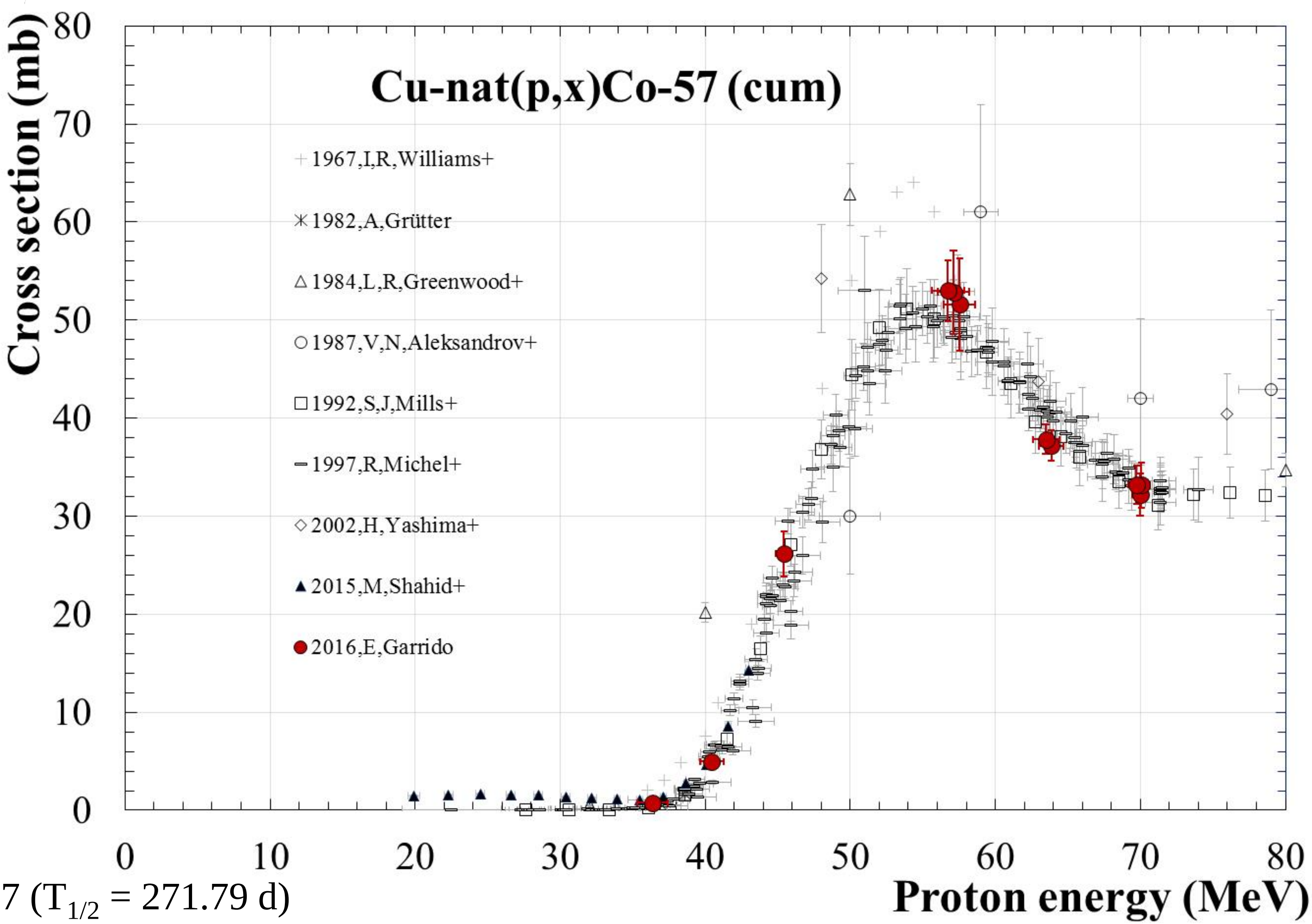
Potential method for production of $^{99}Mo/^{99m}Tc$





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

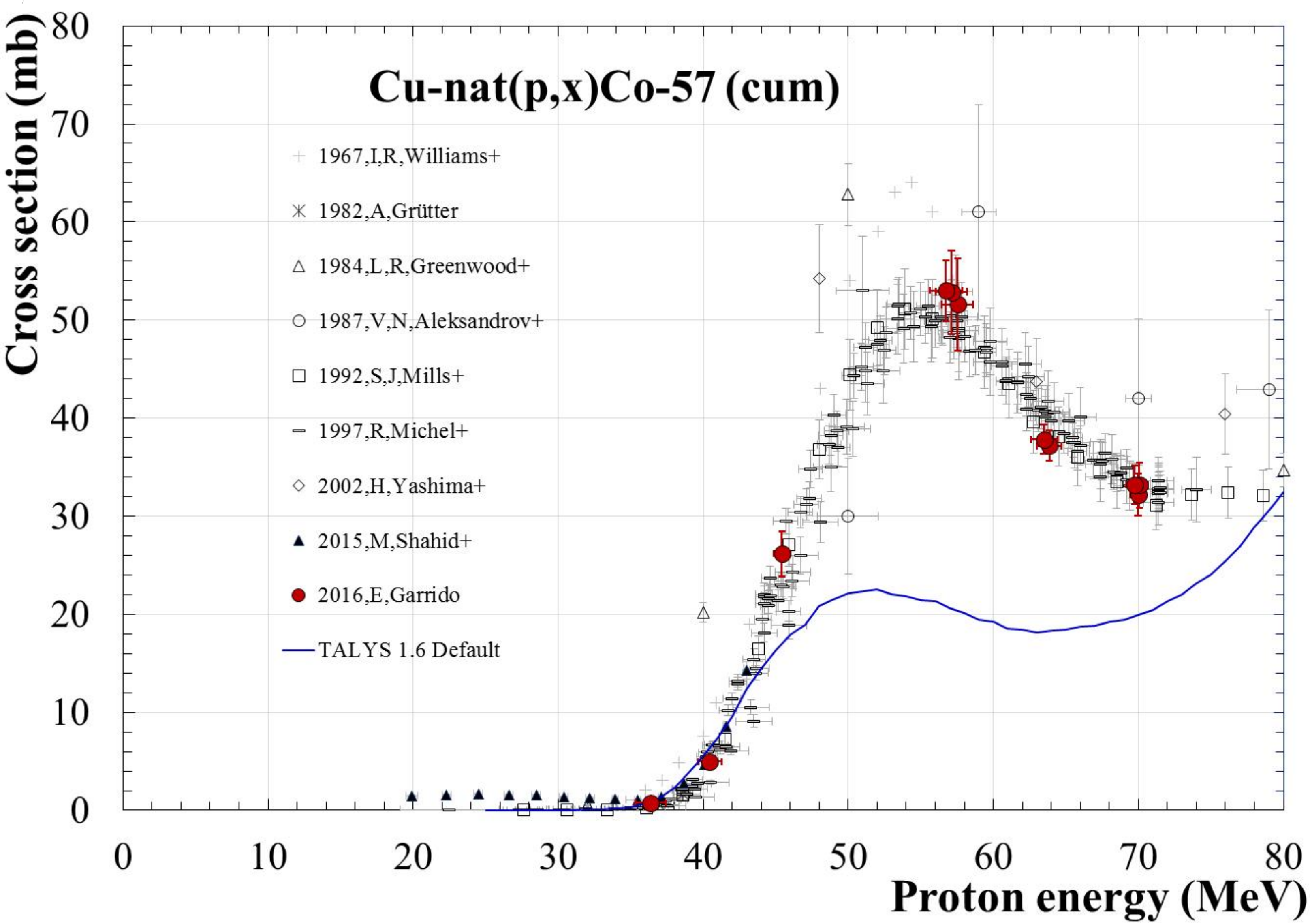
Monitor reaction



Co-57 ($T_{1/2}$ = 271.79 d)
EC process (100%) to stable Fe-57
Suitable for proton monitor reaction

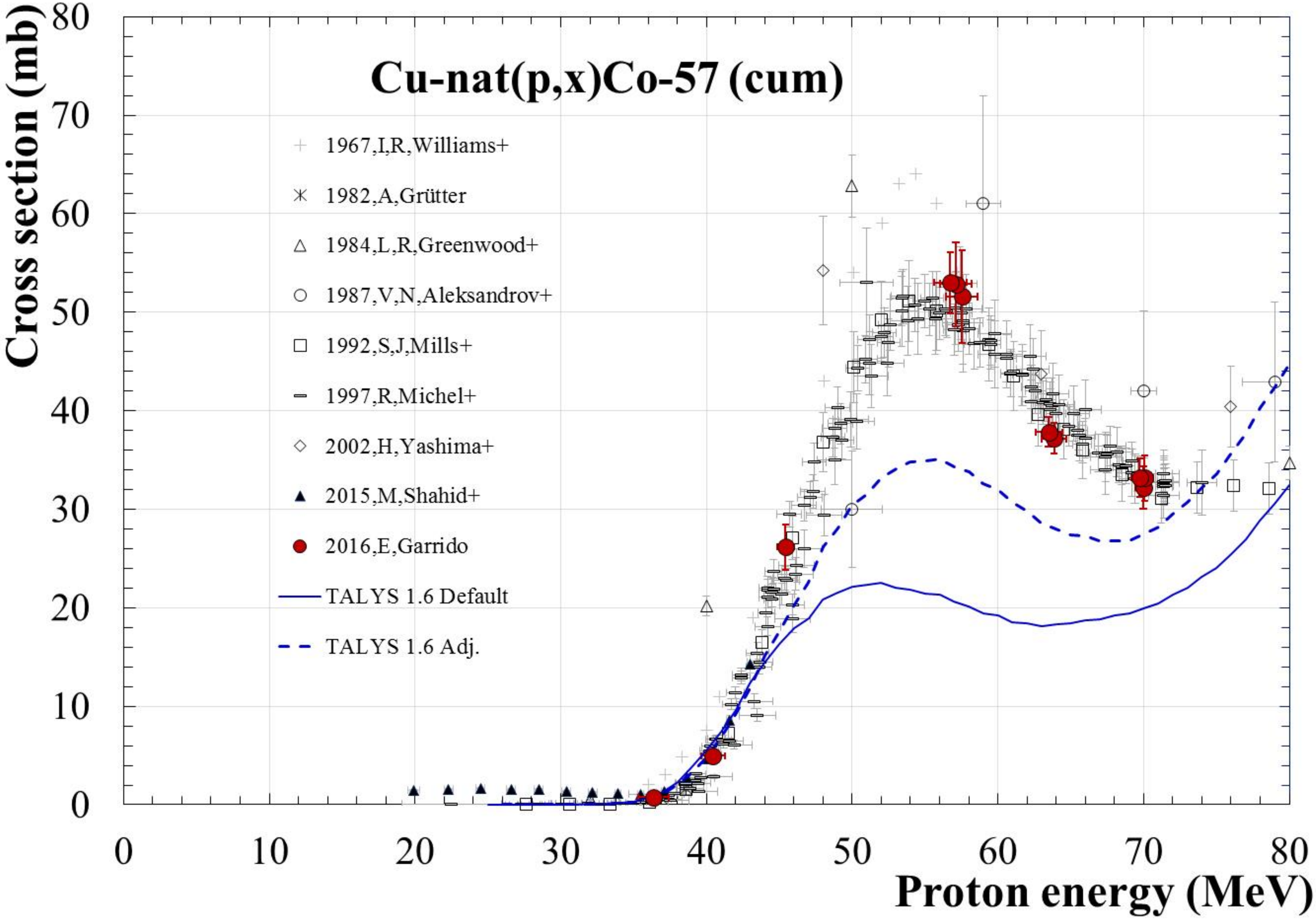
E. Garrido et al., Nucl. Instr. Meth. Phys. Res. B 383 (2016) 191-212

Monitor reaction



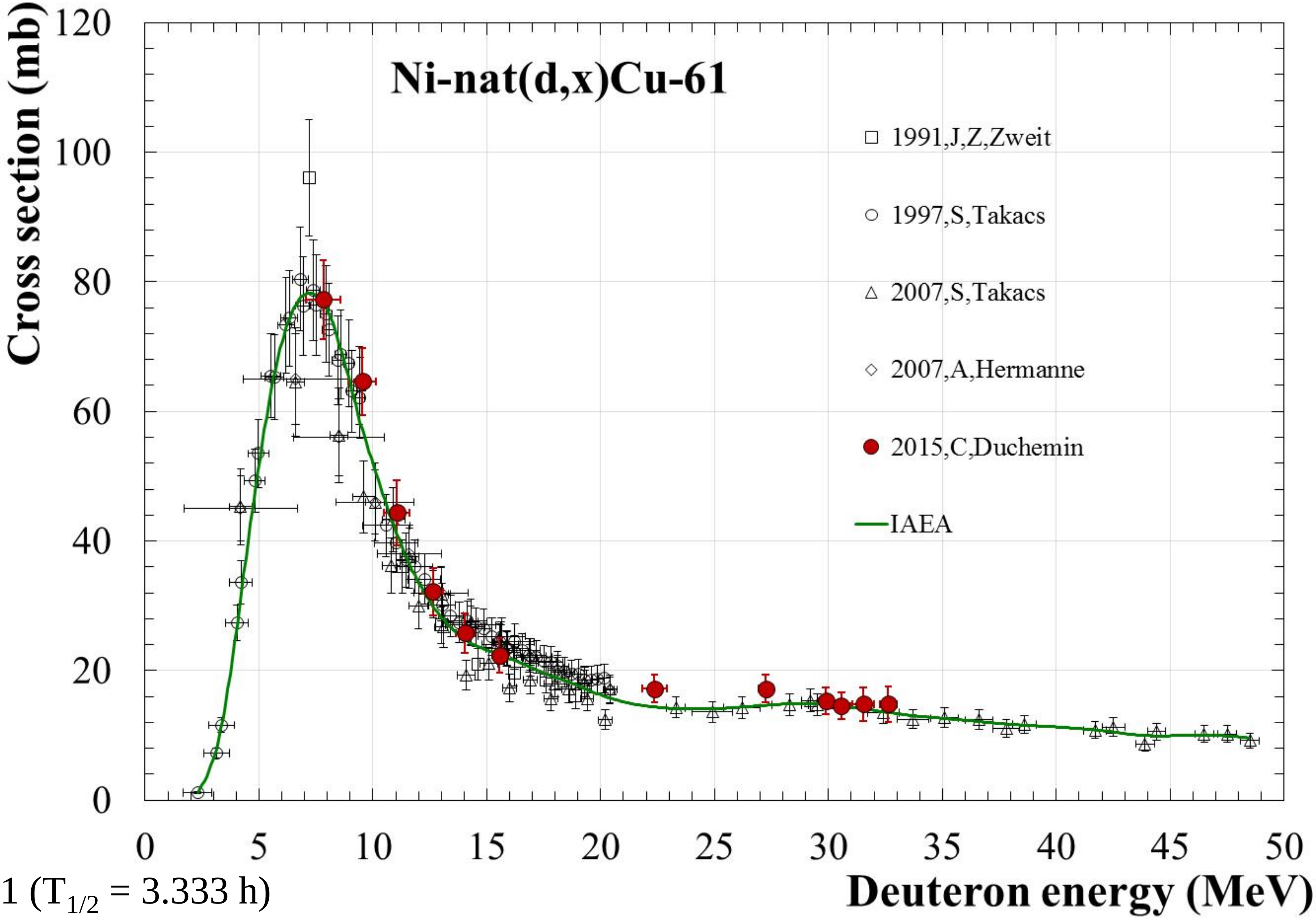
E. Garrido et al., Nucl. Instr. Meth. Phys. Res. B 383 (2016) 191-212

Monitor reaction



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

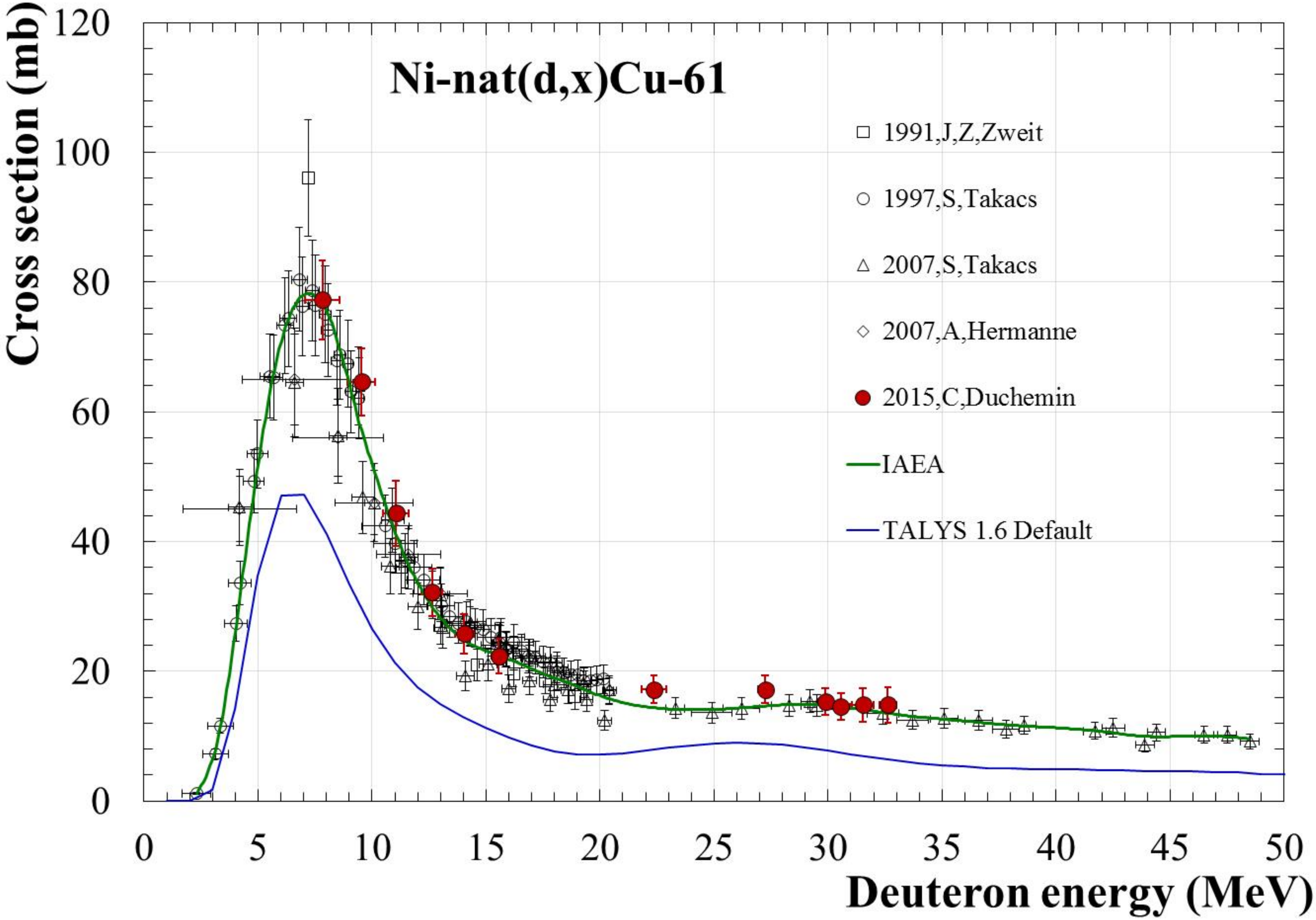
Monitor reaction



Cu-61 ($T_{1/2}$ = 3.333 h)
EC β^+ processes to stable Ni-61
Suitable for PET imaging

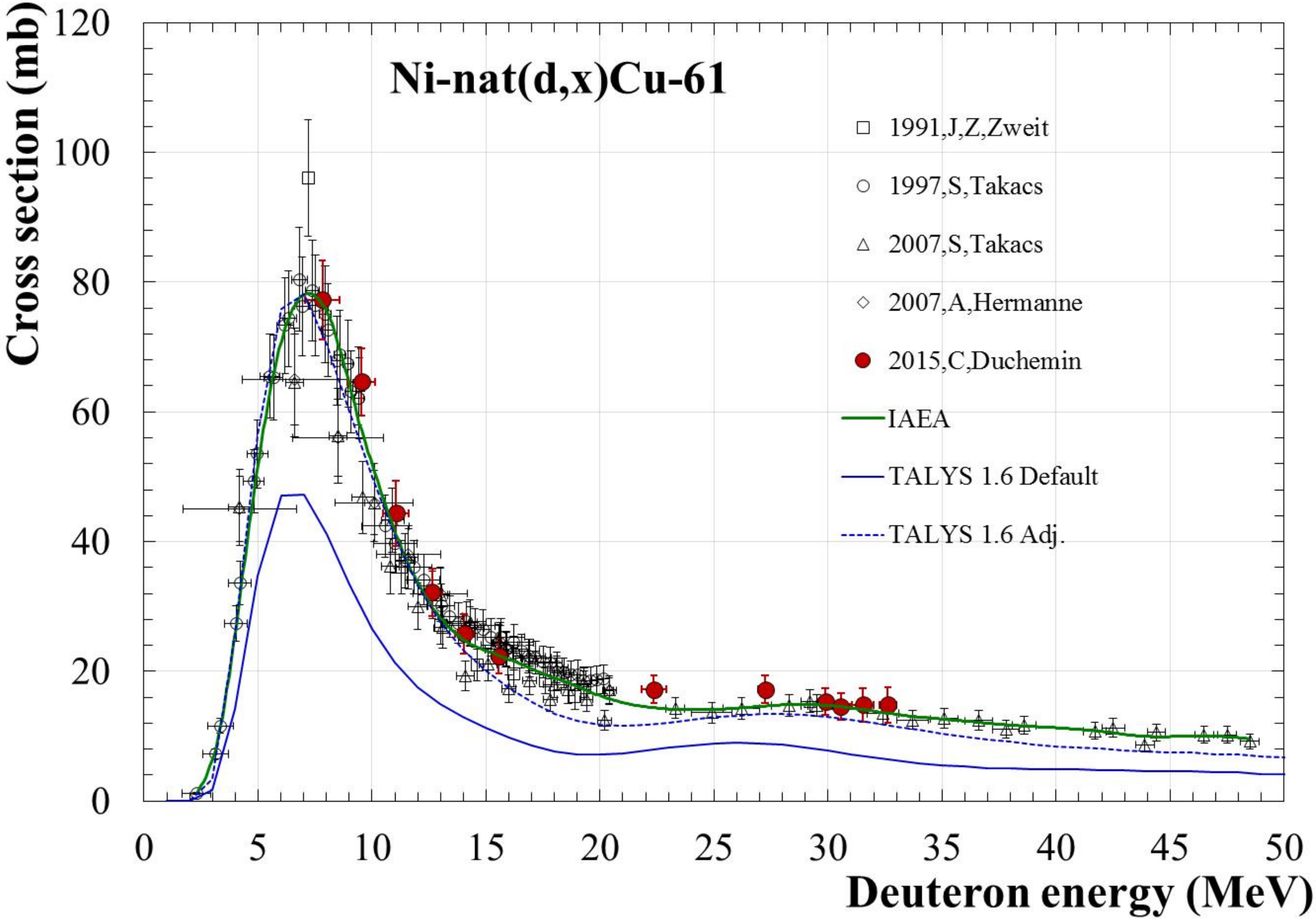
C. Duchemin et al., PhD thesis (2016)

Monitor reaction



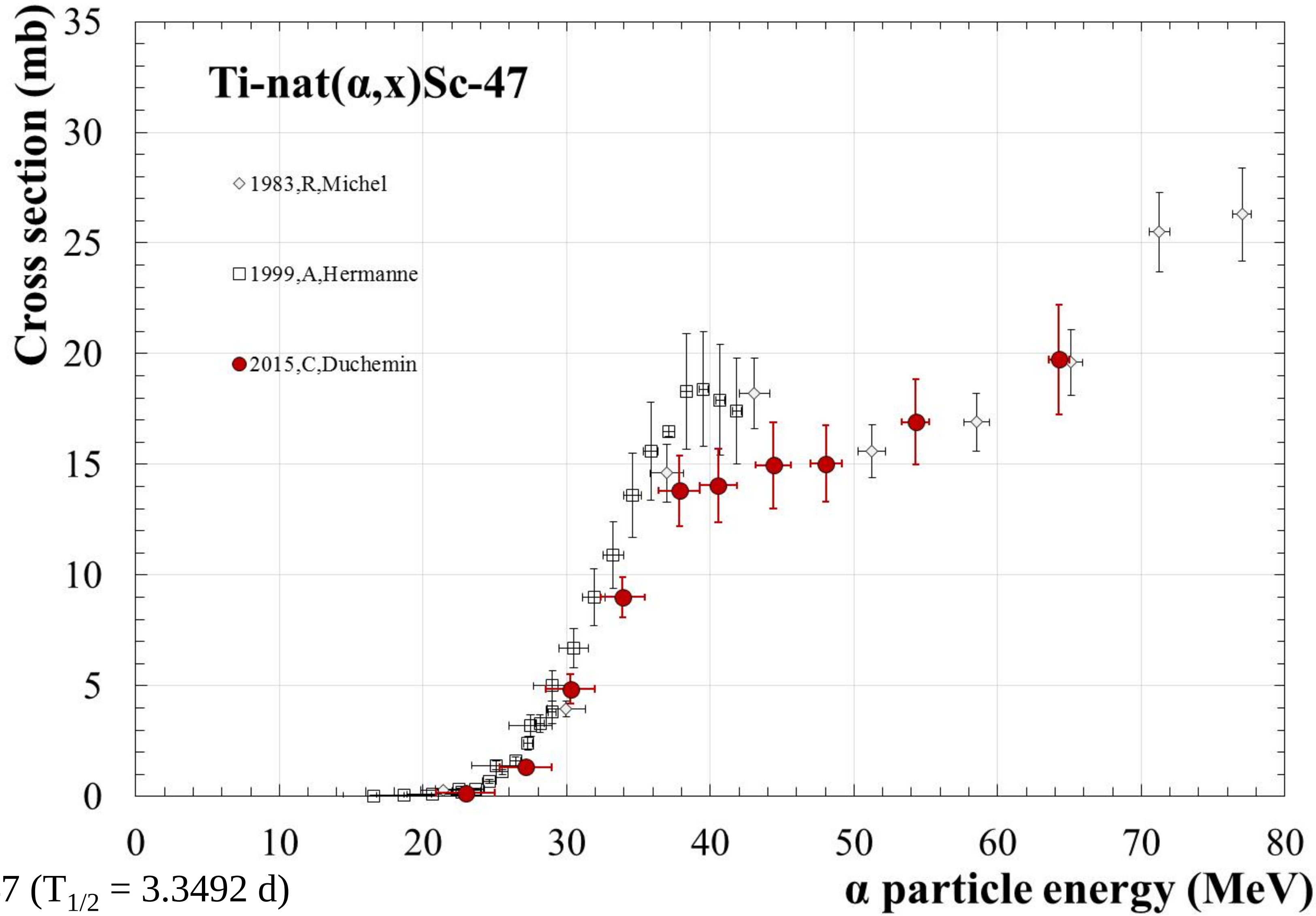
C. Duchemin et al., PhD thesis (2016)

Monitor reaction



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

Monitor reaction



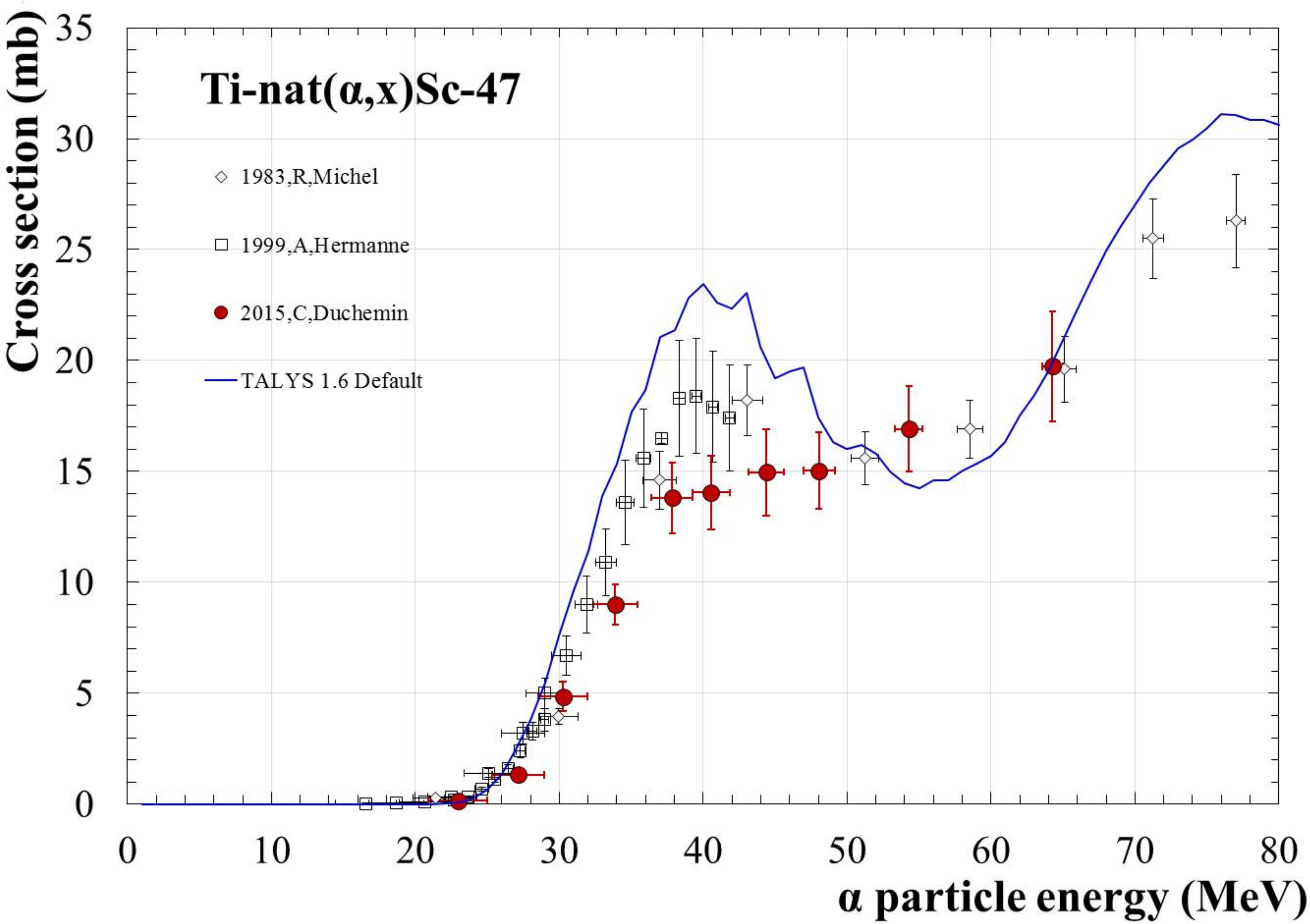
Sc-47 ($T_{1/2}$ = 3.3492 d)

β^- (100%) to stable Ti-47

Suitable for theranostic approach with the Sc-44

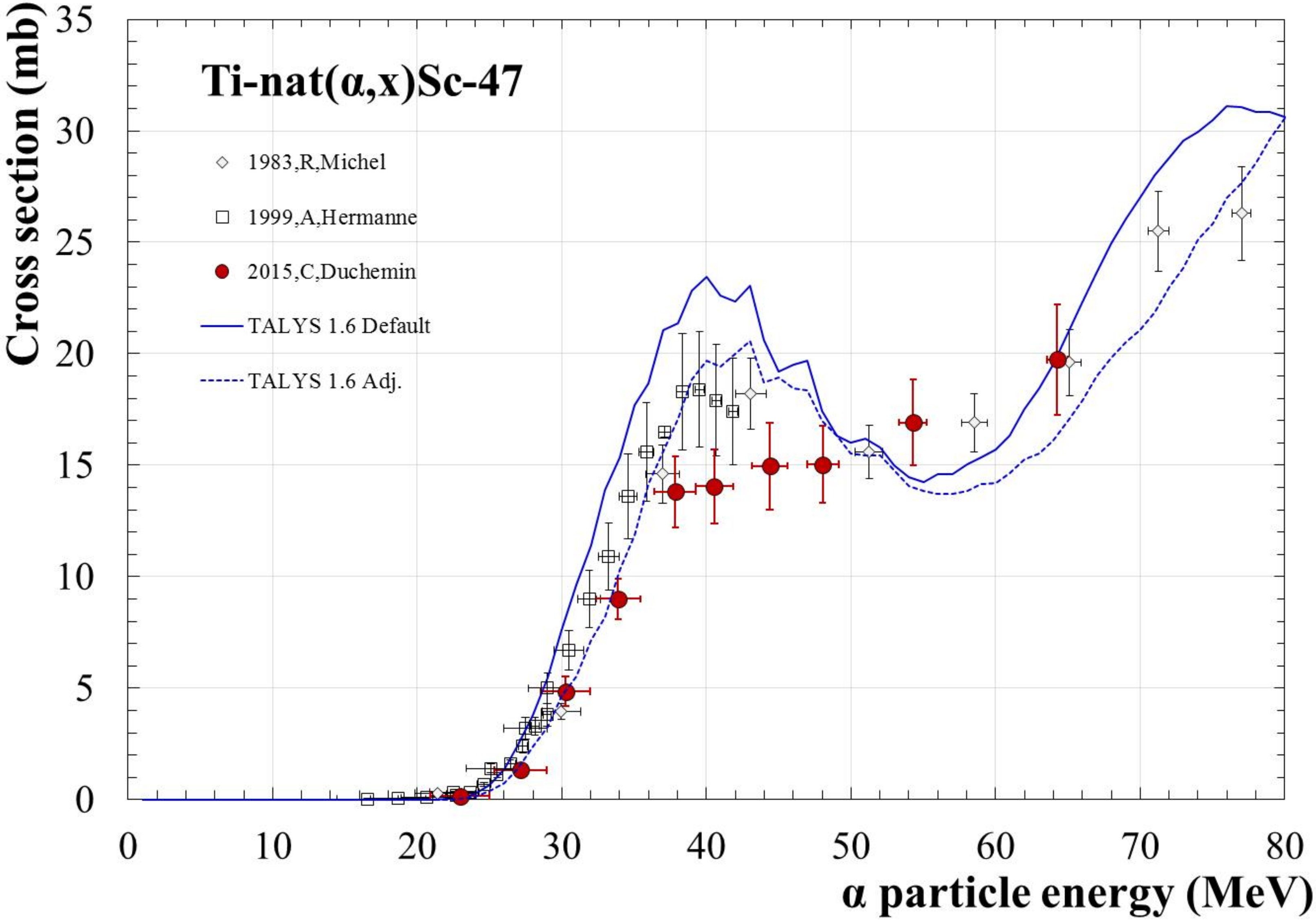
C. Duchemin et al., PhD thesis (2016)

Monitor reaction



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

Monitor reaction



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

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Conclusion and outlooks

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]

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