New nuclear data libraries for Pb and Bi

Arjan Koning
Marieke Duijvestijn
Robin Klein Meulekamp
Steven van der Marck
Alfred Hogenbirk

NRG Petten

CSEWG Meeting, Brookhaven National Laboratory, November 4-6, 2003

AN ECN KEMA COMPANY

Contents

- Nuclear data needs for Pb and Bi
- TALYS code
- TALYS vs. experiments
- New ENDF-6 formatted data libraries
- Integral benchmarks
- Conclusions

Nuclear data problems in lead cooled fast systems

Various independent checks of cross section sensitivity for lead/bismuth cooled ADS:

- IAEA ADS benchmark: reactivity burnup swing
- PDS-XADS
- MUSE
- Other ADS designs (CIEMAT, ANL (Palmiotti, Salvatores))

Result: (n,n'), (n,2n) and feeding from > 20 MeV yield considerable differences, among world data libraries, in source importance ϕ^* and up to 2000 pcm in k_{eff}, *on the basis of Pb cross sections only*.

Uncertainties in lead cross sections

Is this uncertainty really necessary?

Much experimental data from past 10 years not taken into account in data libraries.

Attacking the lead problem:

- Collect all relevant experimental data
- Compare nuclear model code with experiments and adjust nuclear model parameters, if needed.
- Produce all reaction channels with nuclear model code $\longrightarrow$ complete ENDF-6 datafile
- Do not stop at the usual 20 MeV but at 200 MeV. (HINDAS-project)

Why do we need a model code?

Low energies (E $<$ 1 MeV):

- Only a few open reaction channels: Elastic scattering, capture and fission.
- For important nuclei: (almost) Complete measurements can be made.

Higher energies (E $>$ 1 MeV):

- Many reaction channels open up and the existing experimental database can never cover what happens in reality: *Predictive nuclear models are essential*.
- A model code can reveal inconsistencies between uncorrelated experiments.

Important for nuclear applications:

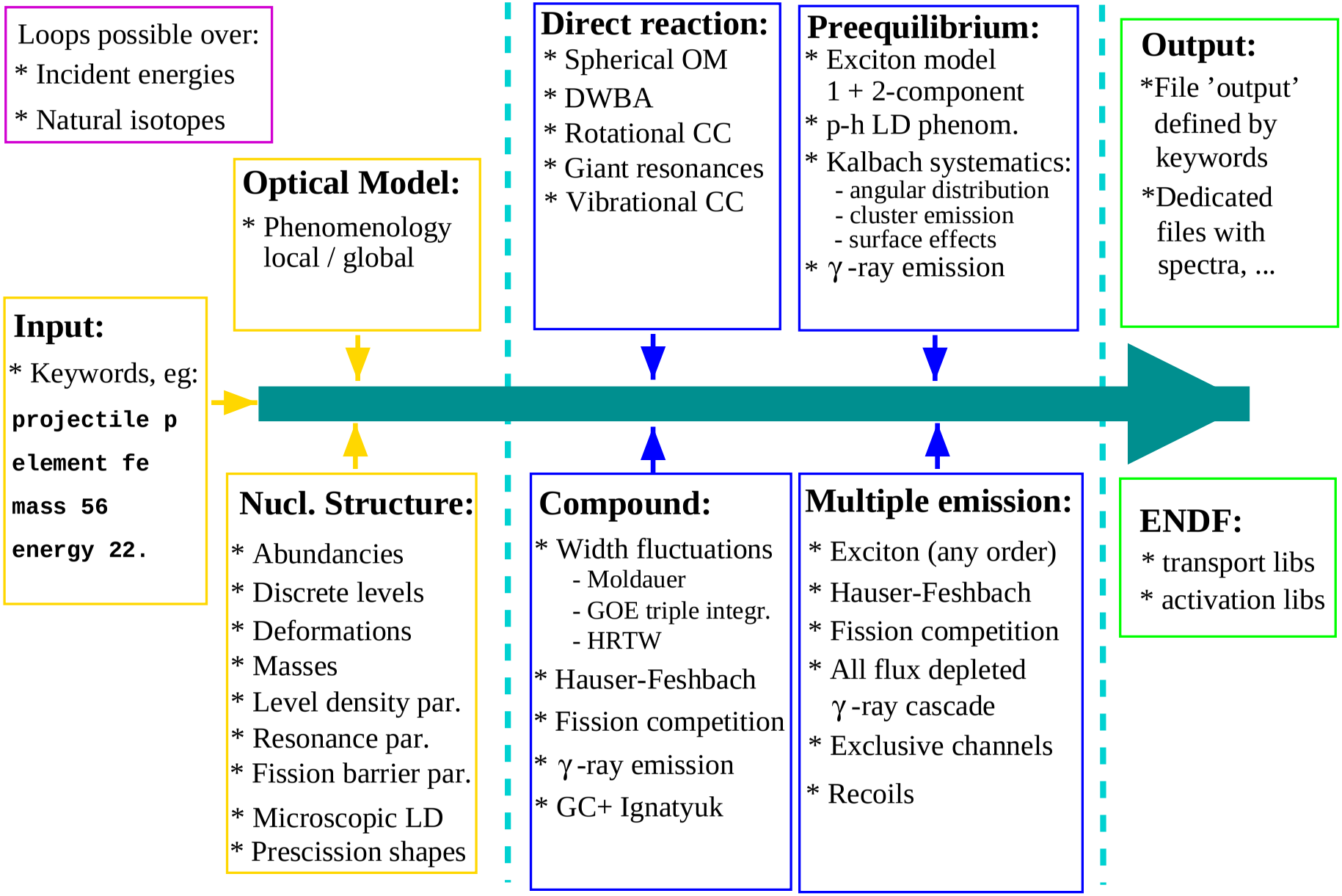
- Number of (expensive) experiments is very finite. A model code can produce nuclear data for *all* nuclides, energies and channels $\longrightarrow$ Required accuracy from applications is crucial information to decide on use of experiment or model code or both!

TALYS

TALYS: nuclear reaction software by NRG Petten (Koning, Duijvestijn) and CEA Bruyères-le-Châtel (Hilaire)

- Energy range 1 keV - 200 MeV.
- Neutrons, protons, deuterons, tritons, helions, alphas and photons.
- Many nuclear reaction models implemented.
- Continuous, smooth description over a wide energy and mass range.
- Automatic generation of data in ENDF-6 format.
- TALYS is not yet generally available.

TALYS: CALCULATIONAL SCHEME

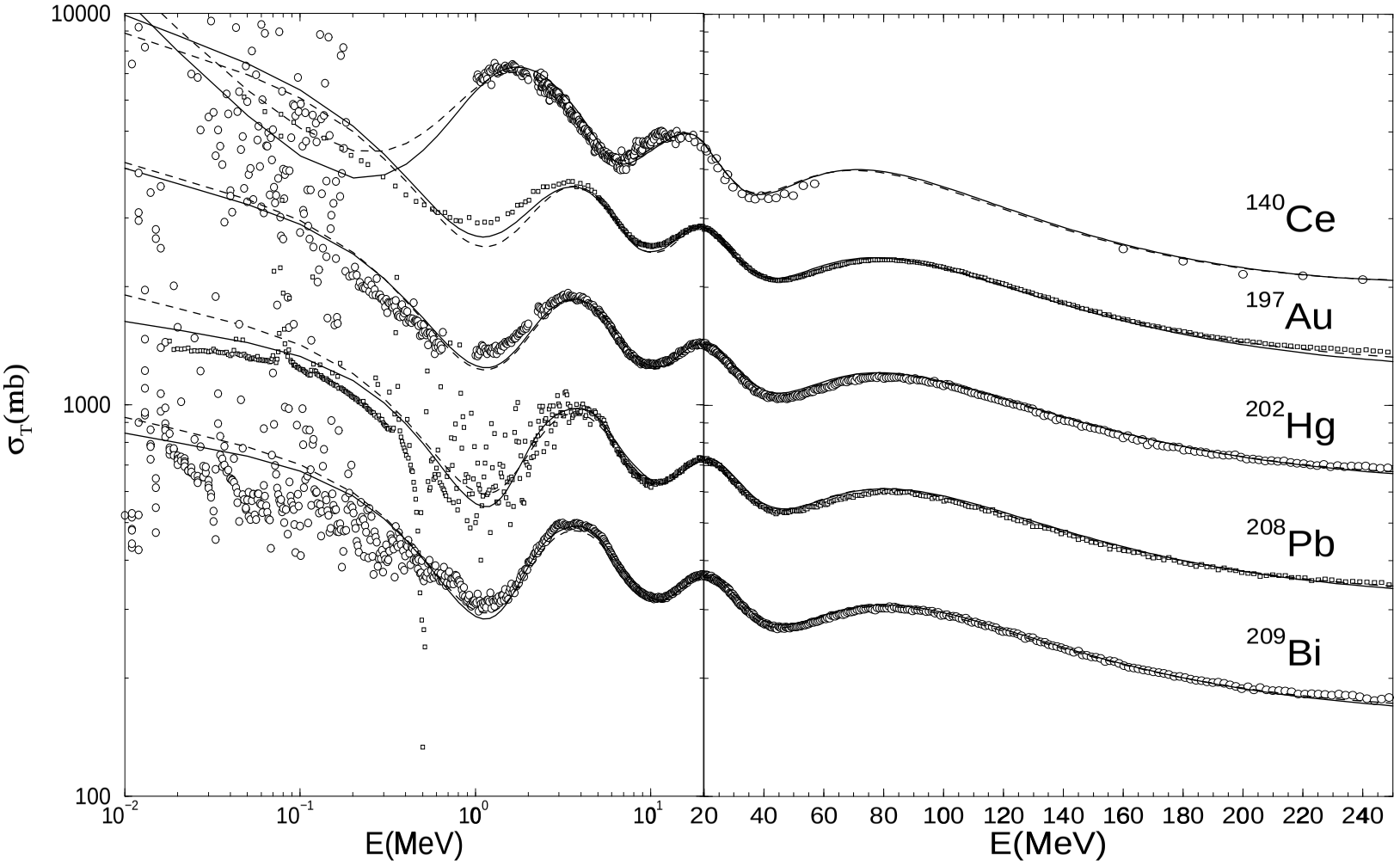


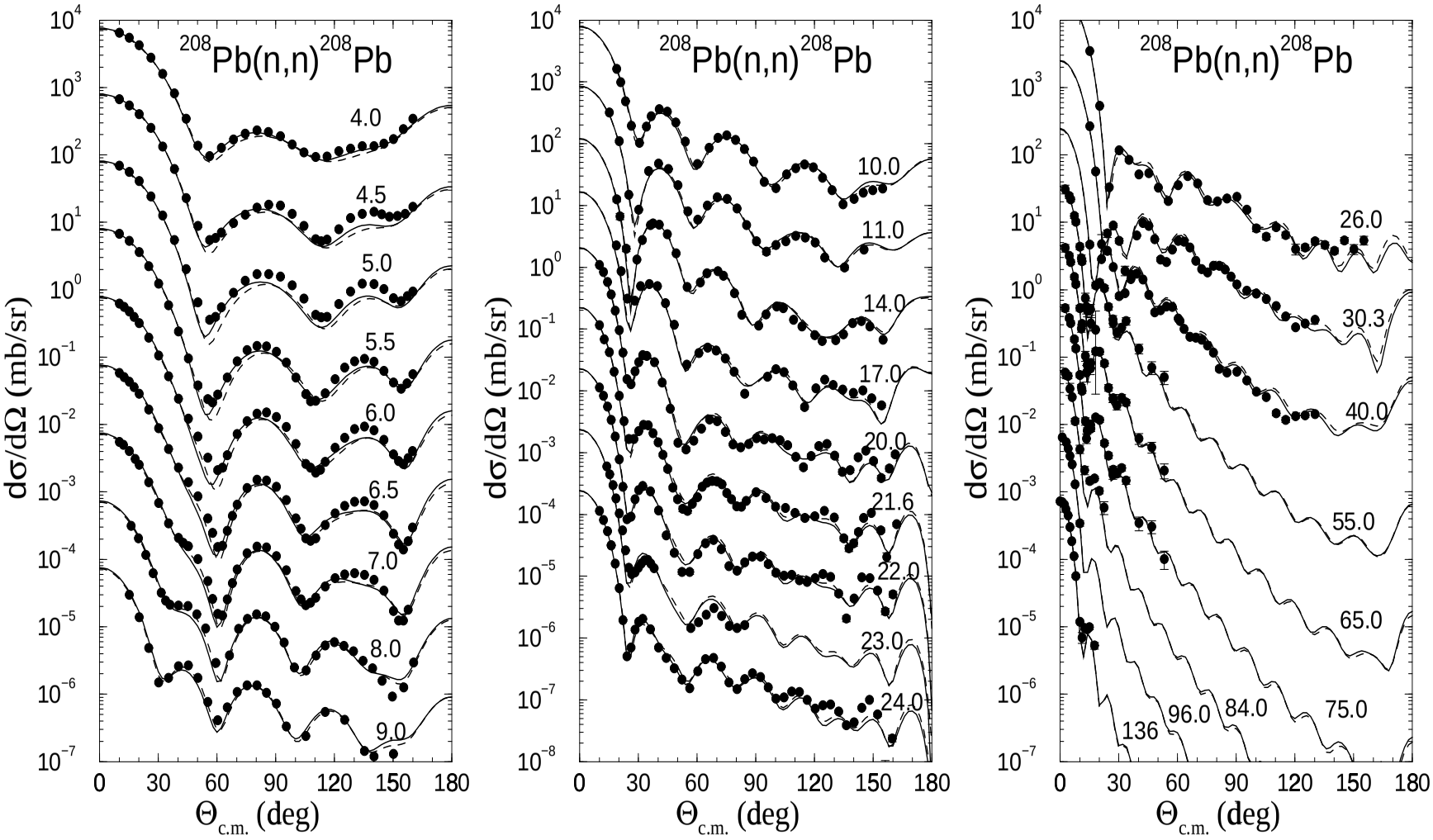
Data produced by TALYS

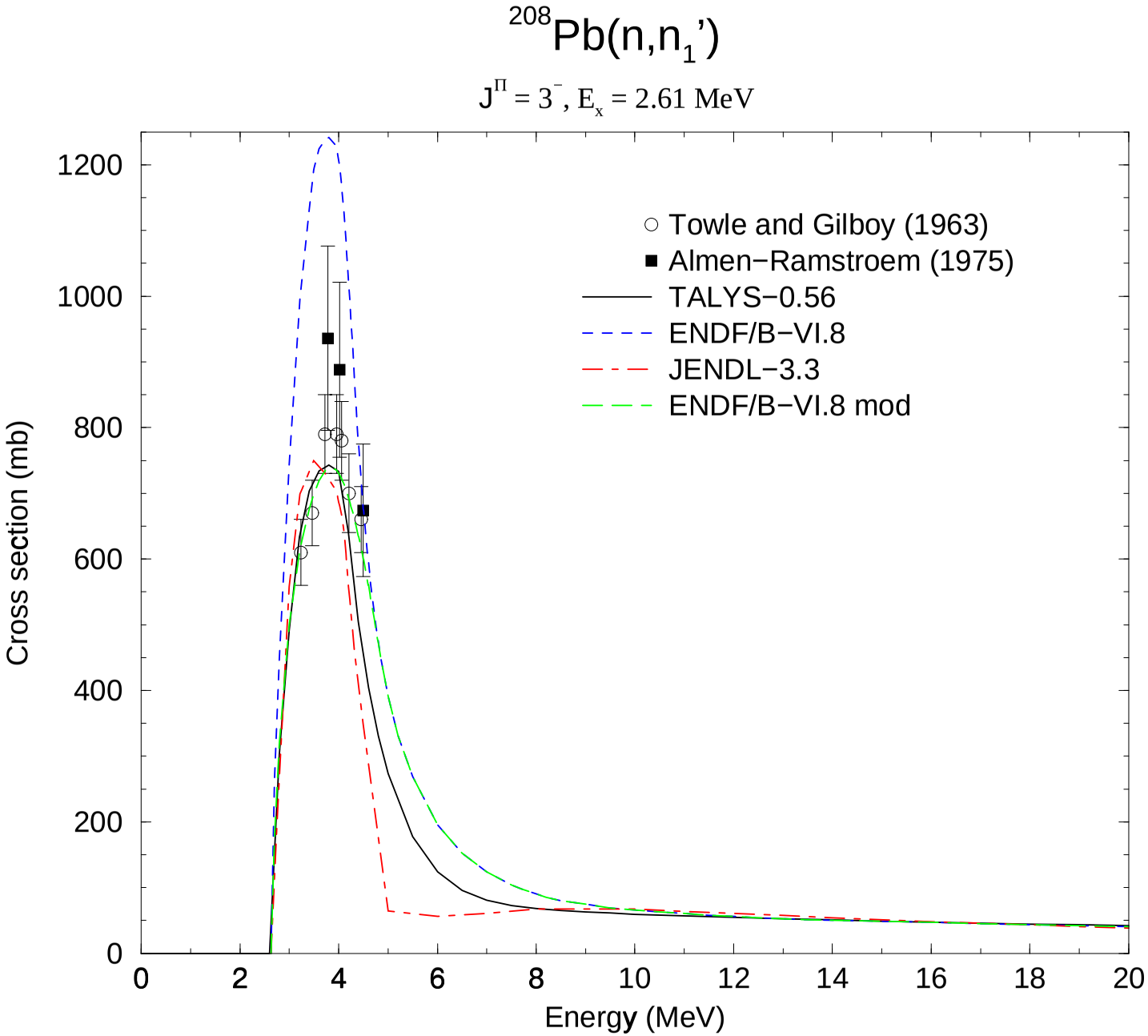
- Total, elastic and reaction cross sections.
- Inelastic cross sections.
- Elastic and inelastic angular distributions.
- All exclusive reaction channels (e.g. (n,p), (n,2n), (n,nd2a)).
- All exclusive double-differential spectra.
- All exclusive isomeric production cross sections.
- All exclusive discrete and continuum gamma ray production cross sections.
- Photonuclear reactions.
- Reactions on isomeric targets.

Data produced by TALYS

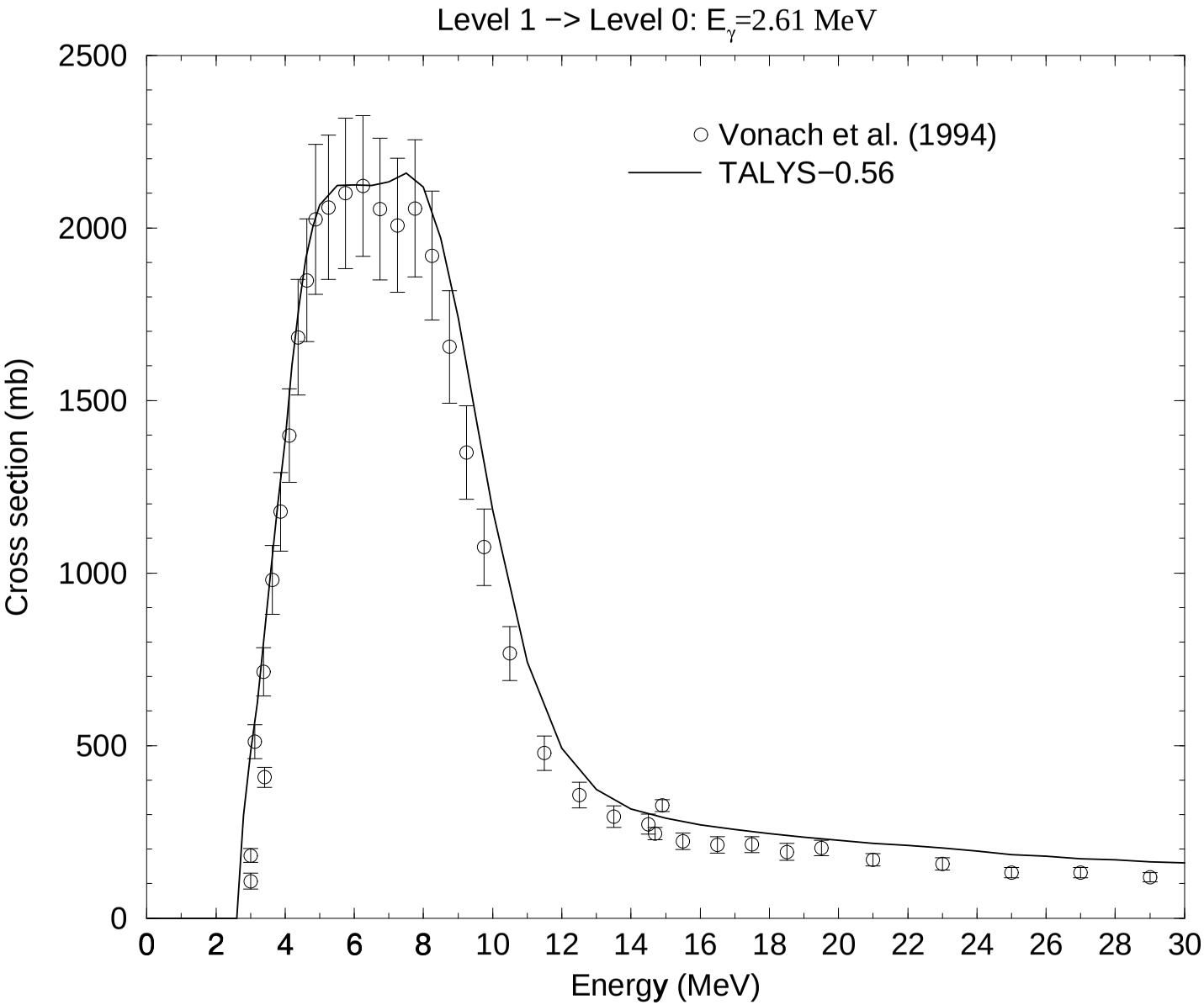
- Total particle production cross sections (n,xn).
- Total particle production double-differential spectra.
- Residual production cross sections (including isomers).
- Activation libraries in EAF and ENDF-6 format.
- Transport libraries in ENDF-6 format.


10000
1000
100
σT(mb)
10−2
10−1
100
101
E(MeV)
20
40
60
80
100
120
140
160
180
200
220
240
E(MeV)
140Ce
197Au
202Hg
208Pb
209Bi


^{208}Pb(n,n)^{208}Pb
4.0
4.5
5.0
5.5
6.0
6.5
7.0
8.0
9.0
dσ/dΩ (mb/sr)
Θc.m. (deg)
^{208}Pb(n,n)^{208}Pb
10.0
11.0
14.0
17.0
20.0
21.6
22.0
23.0
24.0
dσ/dΩ (mb/sr)
Θc.m. (deg)
^{208}Pb(n,n)^{208}Pb
26.0
30.3
40.0
55.0
65.0
75.0
84.0
96.0
136
dσ/dΩ (mb/sr)
Θc.m. (deg)

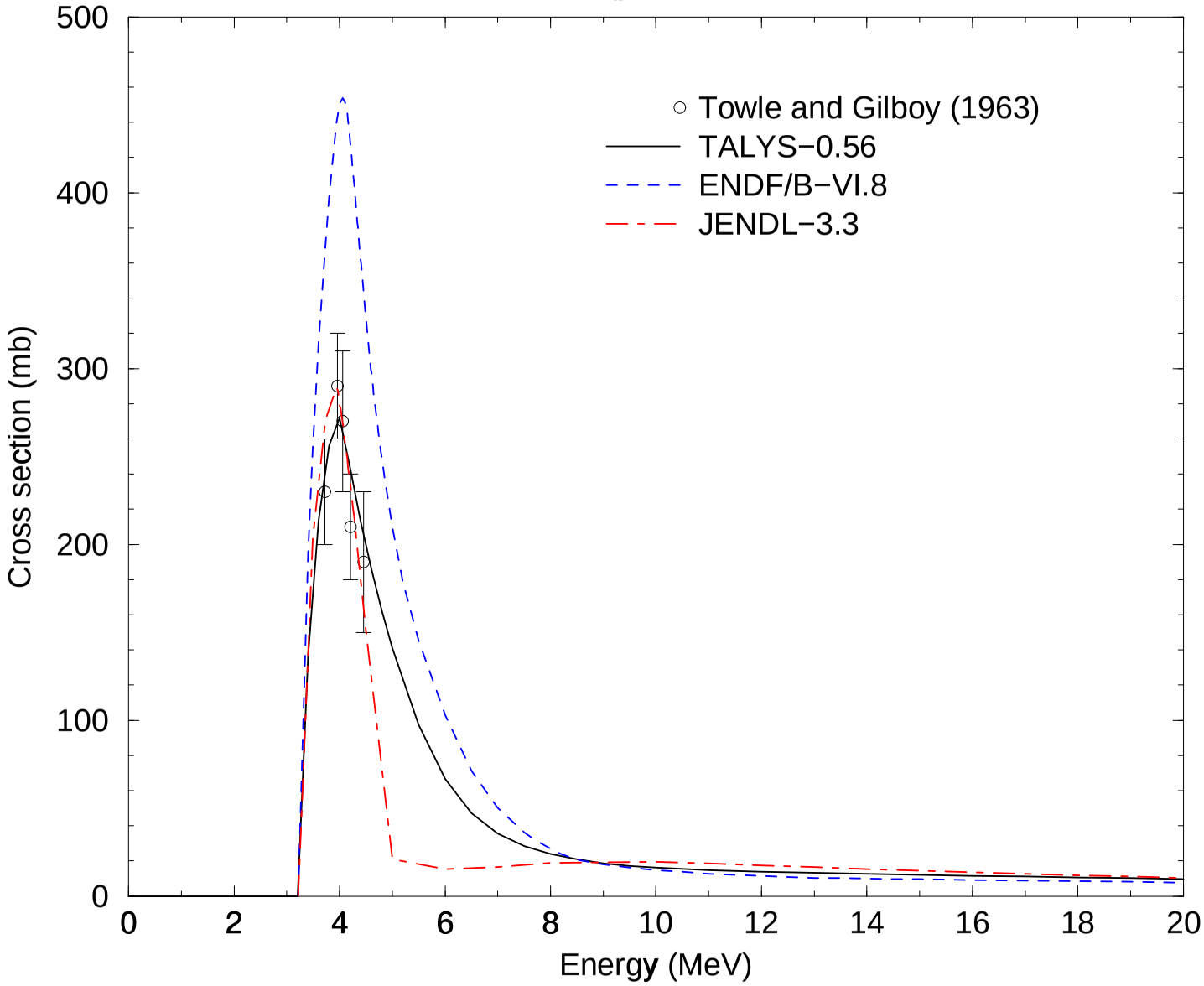

208Pb(n,n1')
Jπ = 3−, Ex = 2.61 MeV
Towle and Gilboy (1963)
Almen−Ramstroem (1975)
TALYS−0.56
ENDF/B−VI.8
JENDL−3.3
ENDF/B−VI.8 mod
Cross section (mb)
Energy (MeV)
0
200
400
600
800
1000
1200
0
2
4
6
8
10
12
14
16
18
20

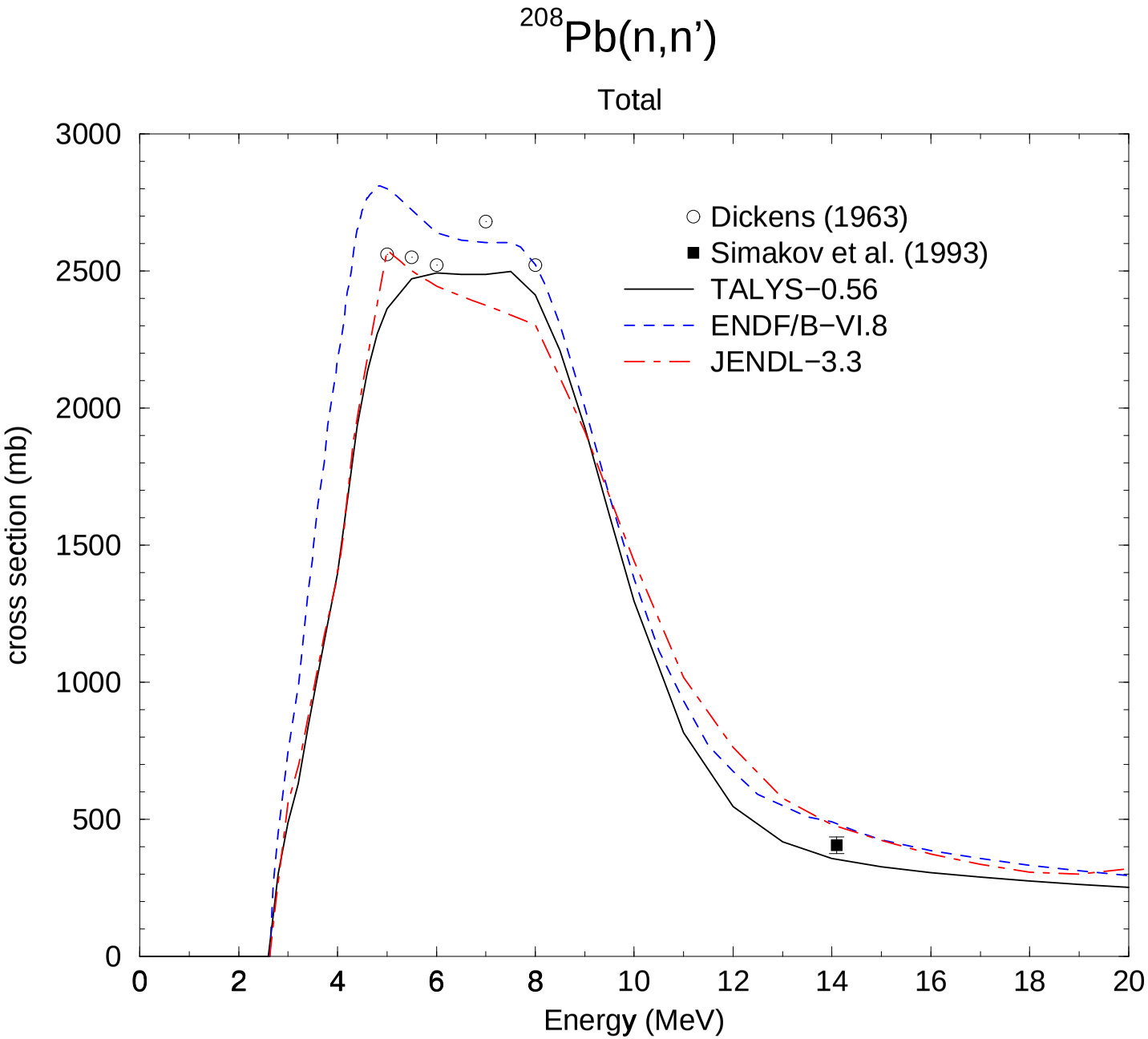
^{208}Pb(n,n'γ)


Level 1 −> Level 0: E$_\gamma$=2.61 MeV
Vonach et al. (1994)
TALYS−0.56
Cross section (mb)
Energy (MeV)
0
500
1000
1500
2000
2500
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30

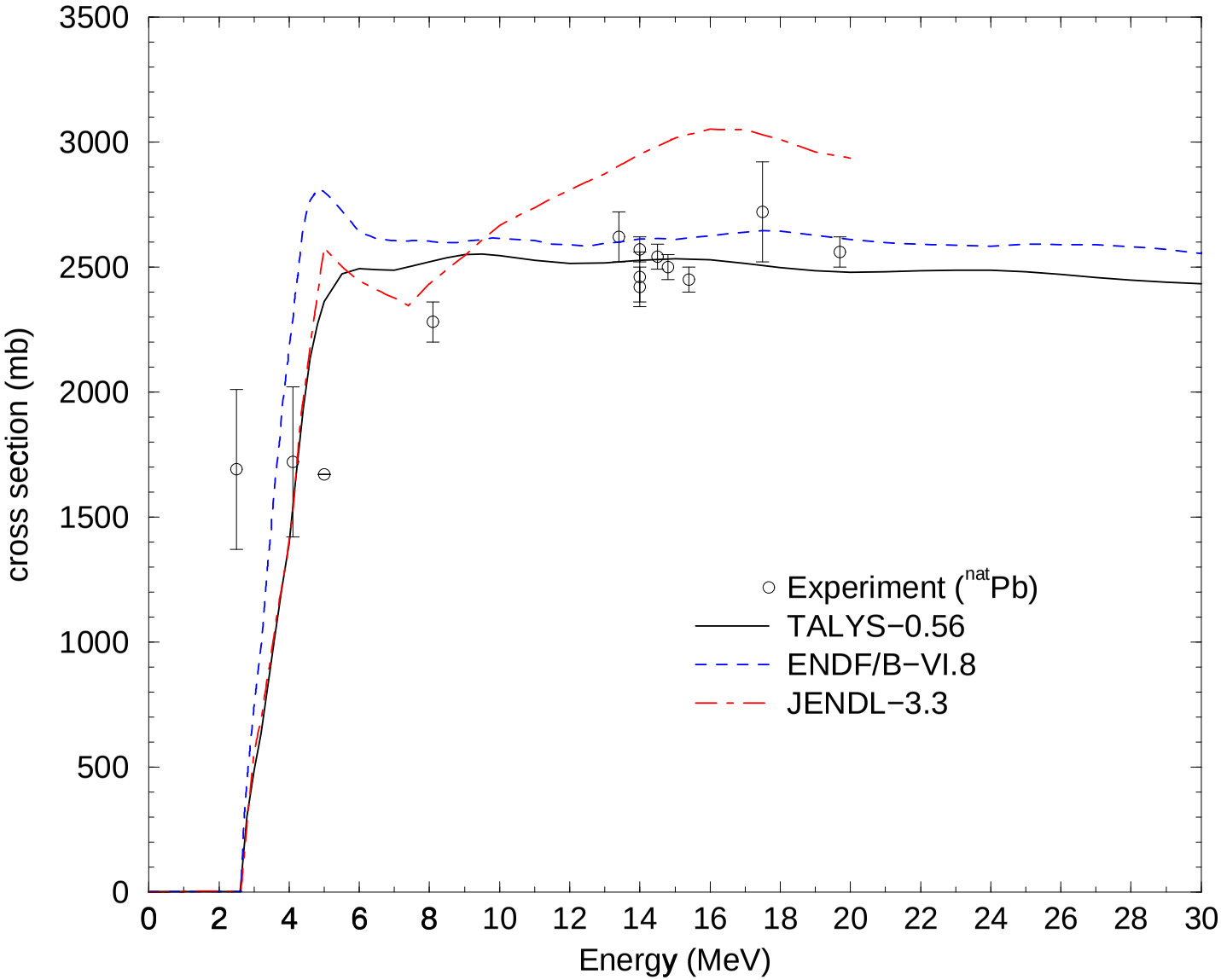
$^{208}Pb(n,n_2')$

$J^{\Pi} = 5^-$, E_x = 3.20 MeV

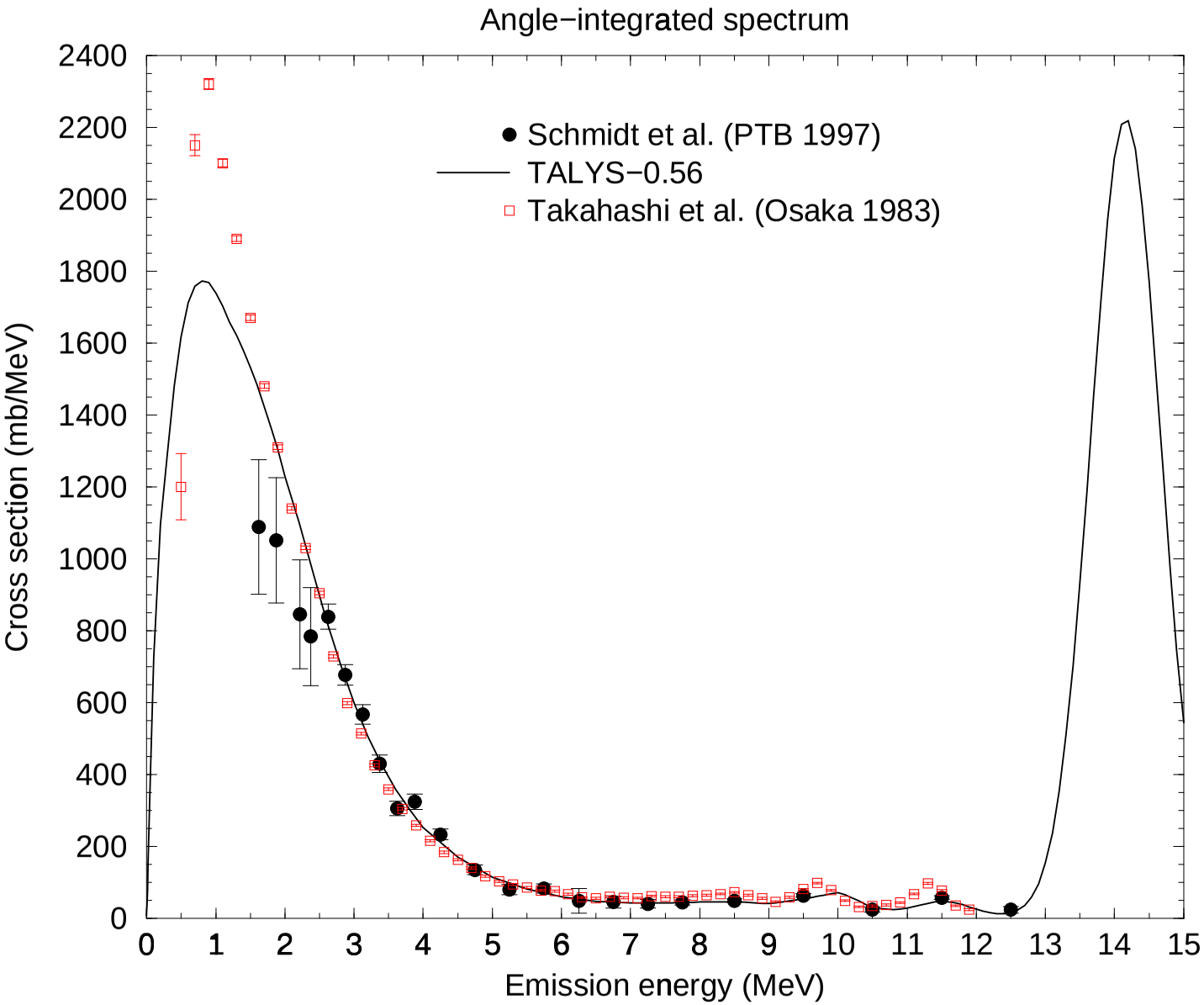



^{208}Pb(n,n')
Total
Dickens (1963)
Simakov et al. (1993)
TALYS−0.56
ENDF/B−VI.8
JENDL−3.3
cross section (mb)
Energy (MeV)
0
500
1000
1500
2000
2500
3000
0
2
4
6
8
10
12
14
16
18
20

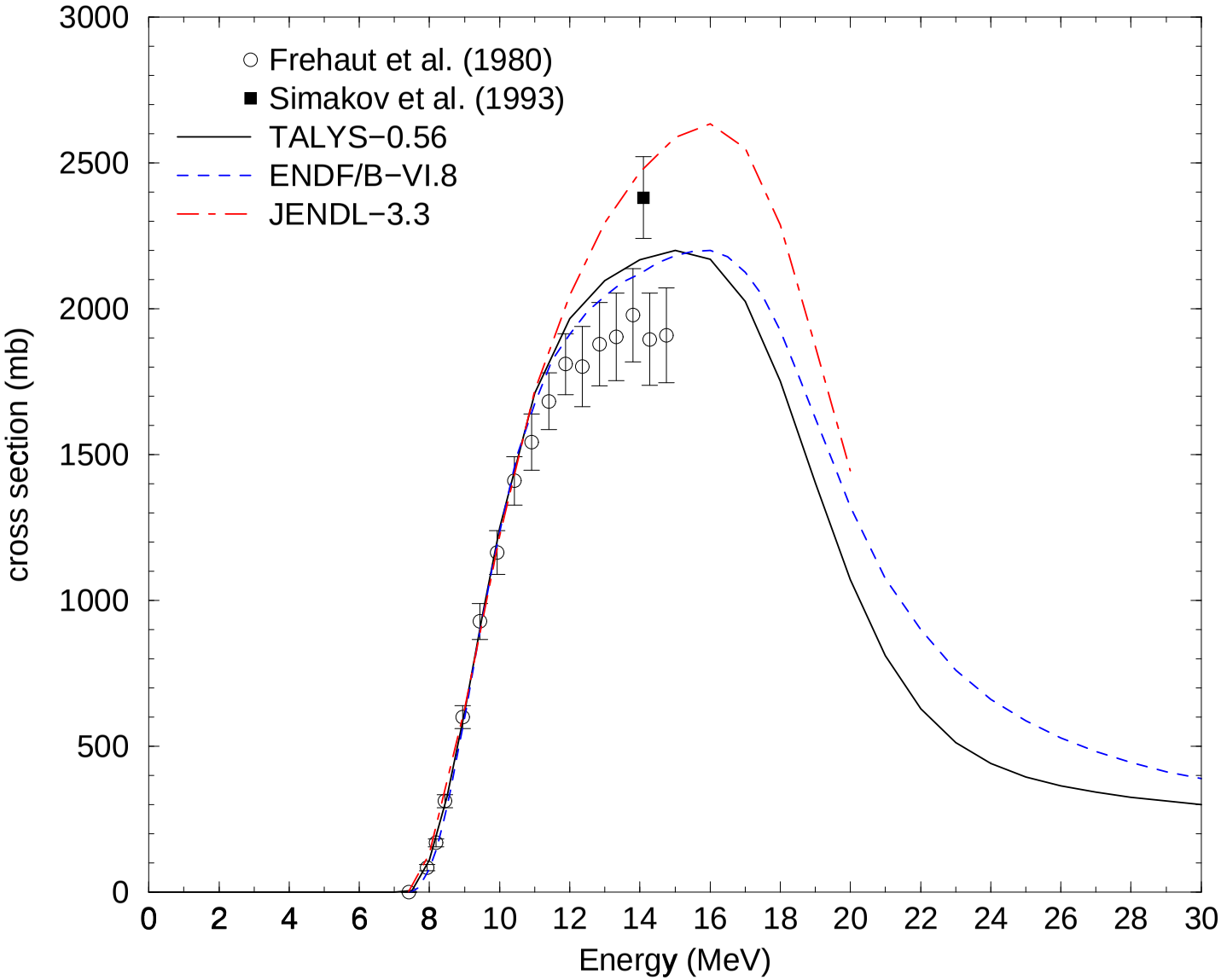
^{208}Pb(n,non)


cross section (mb)
Energy (MeV)
Experiment (^{nat}Pb)
TALYS−0.56
ENDF/B−VI.8
JENDL−3.3

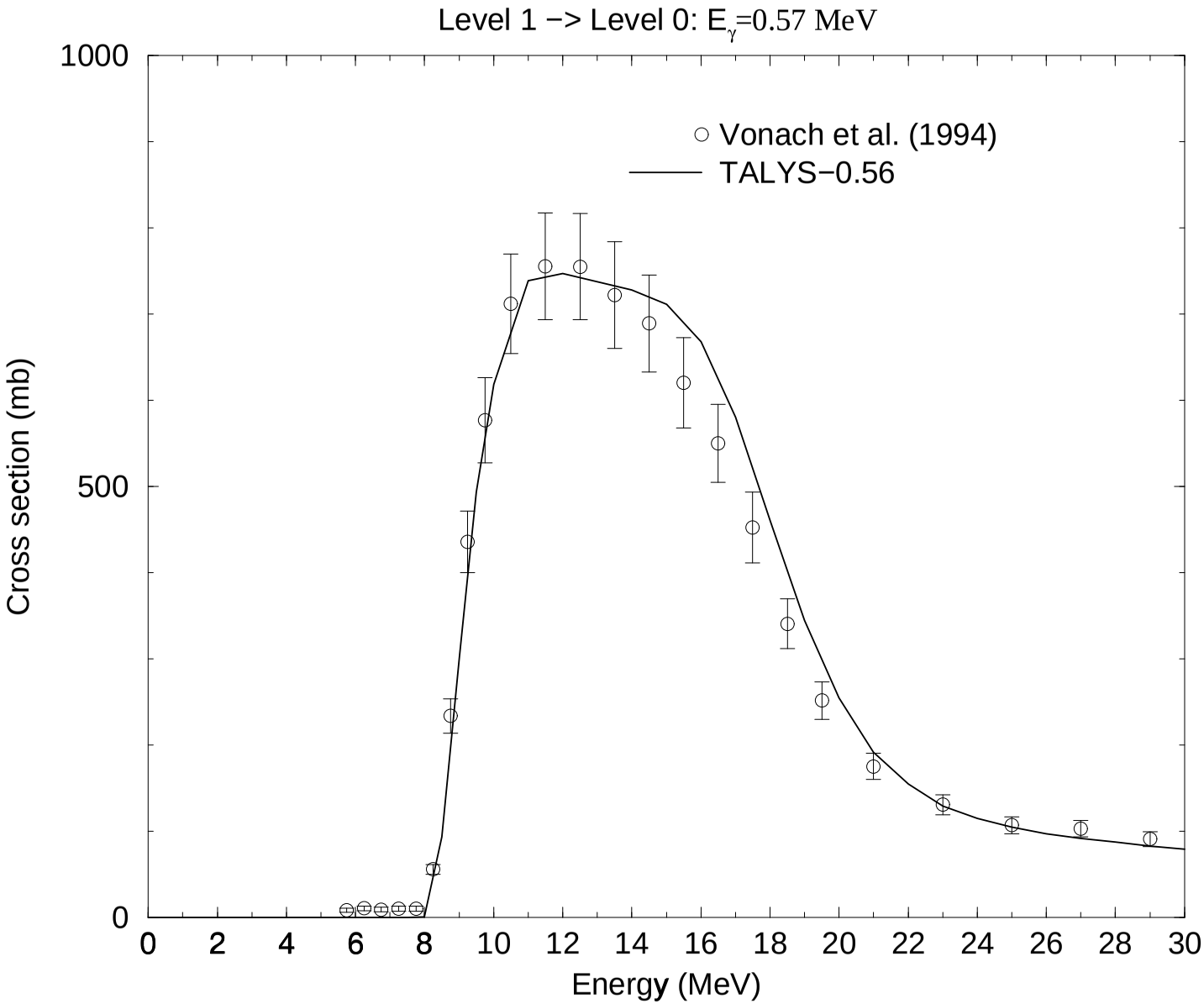
Pb(n,xn) at 14.23 MeV



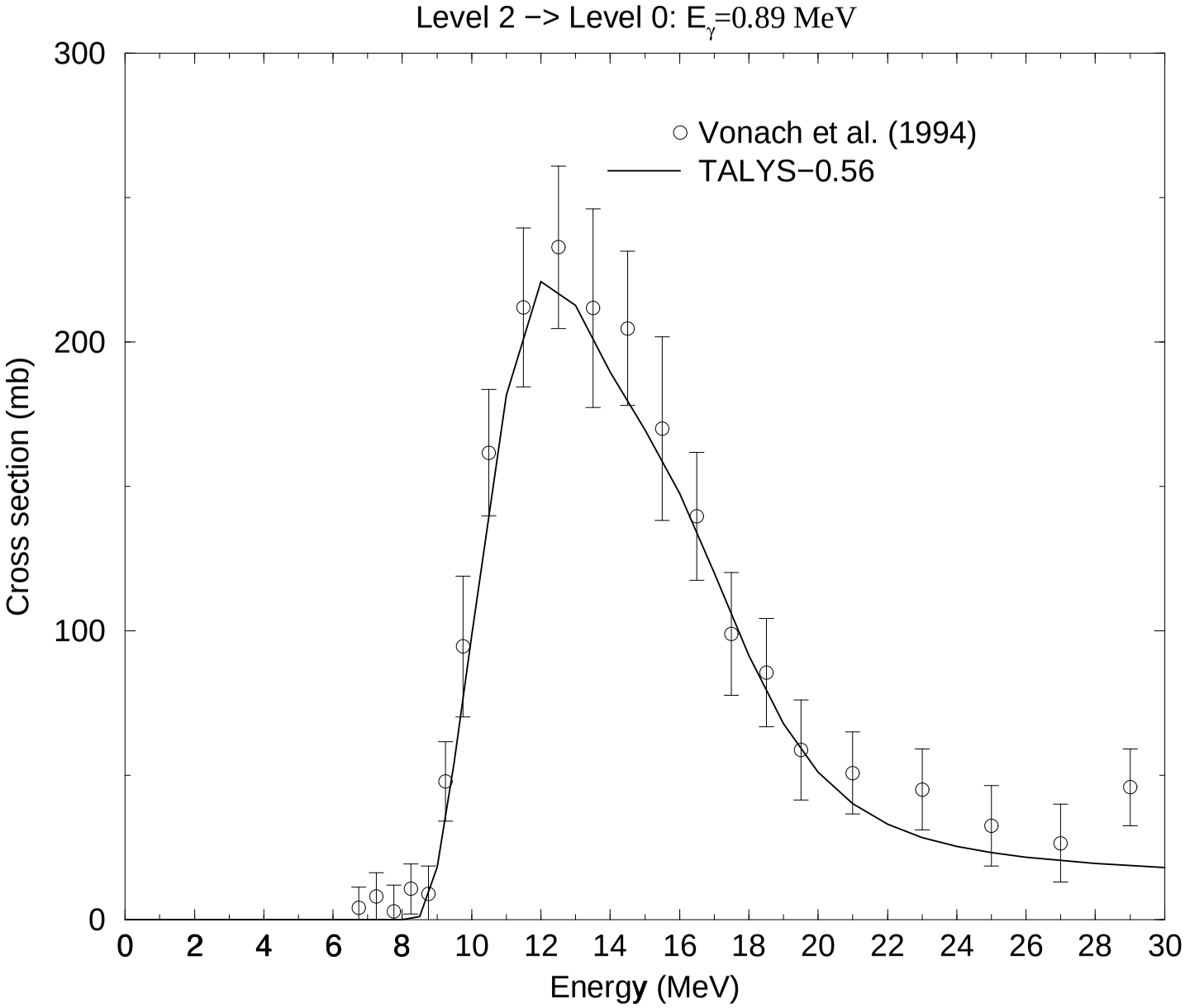
^{208}Pb(n,2n)^{207}Pb



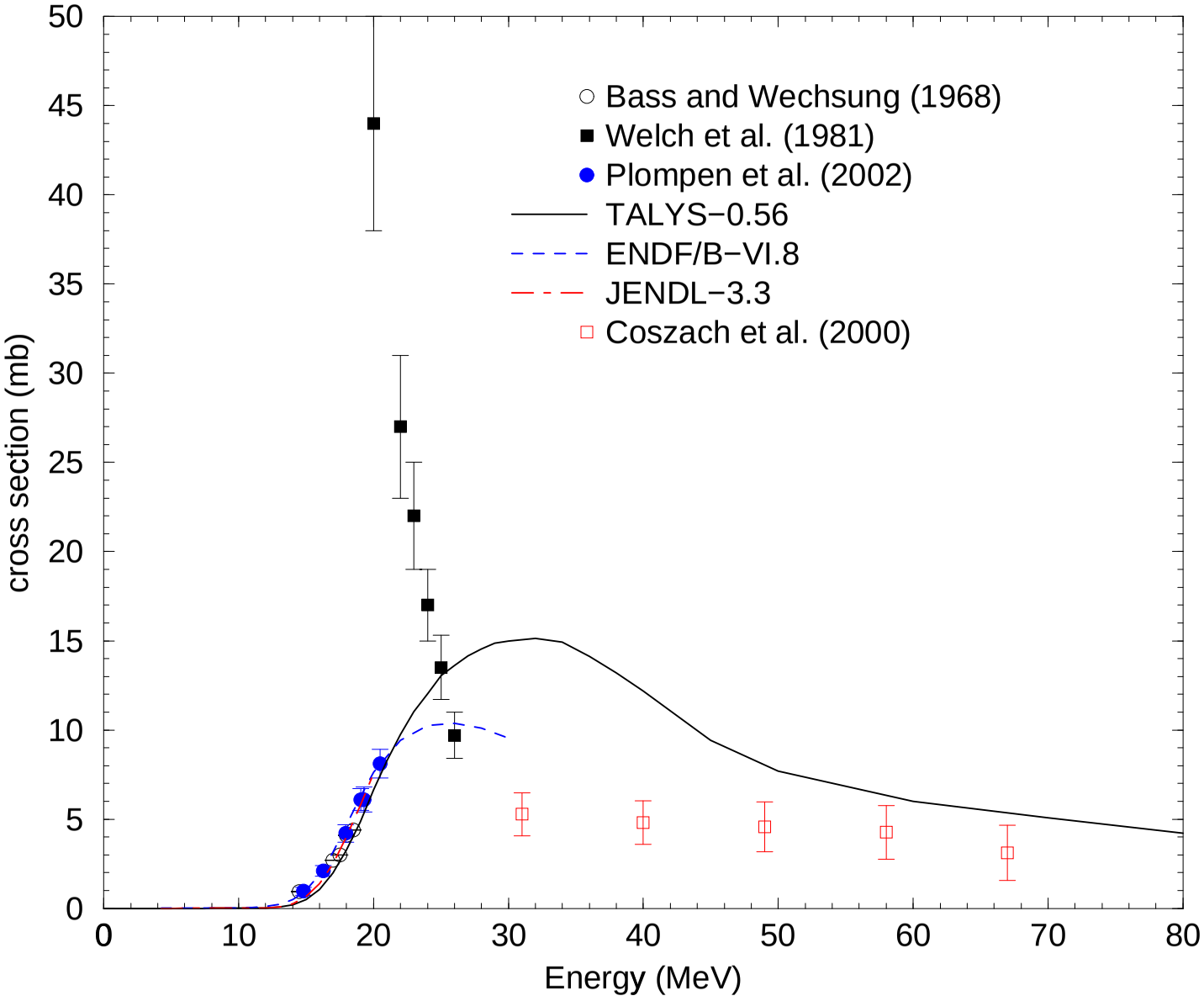
^{208}Pb(n,2nγ)

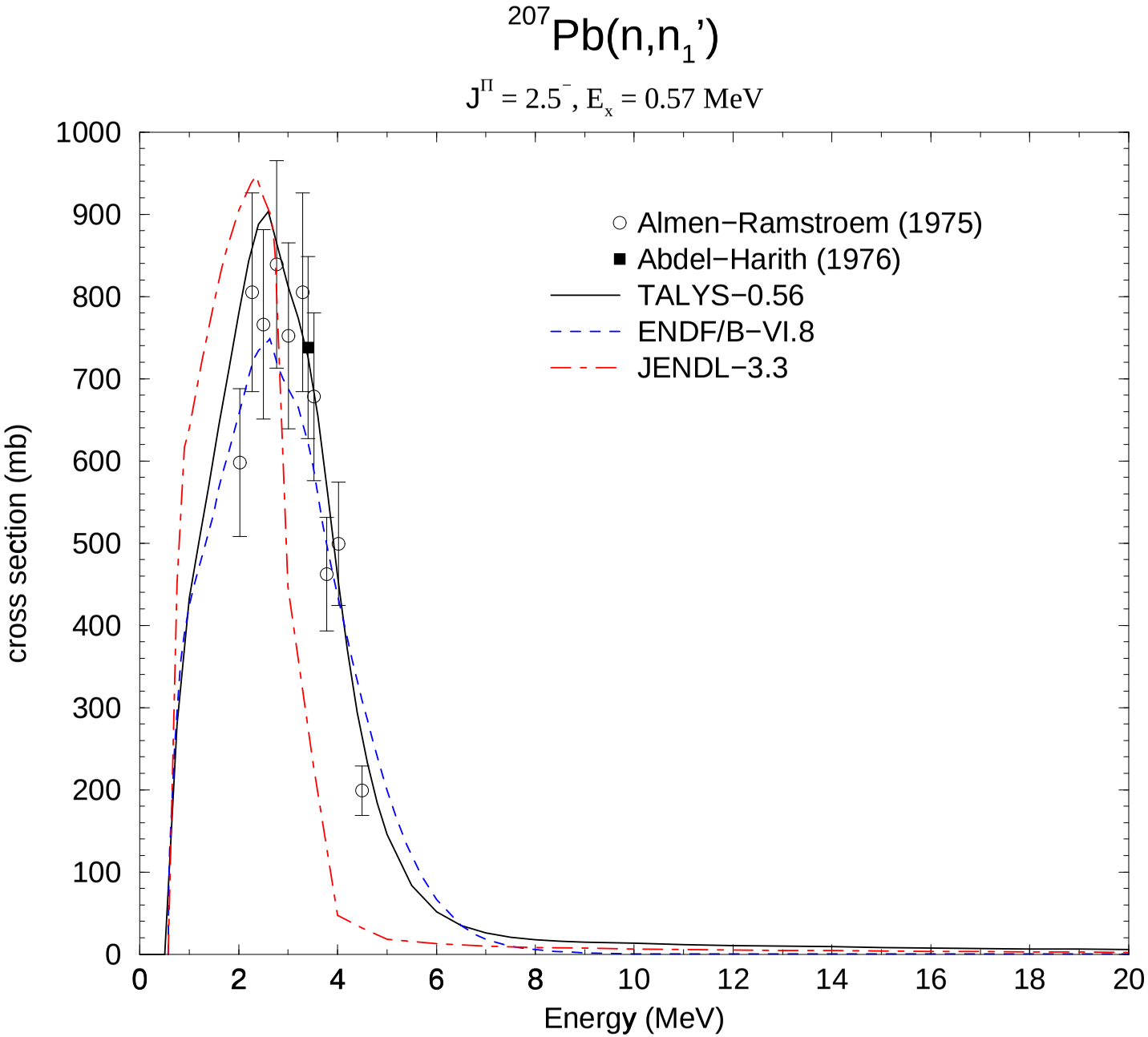

Level 1 −> Level 0: E$_\gamma$=0.57 MeV
Vonach et al. (1994)
TALYS−0.56
Cross section (mb)
Energy (MeV)
0
500
1000
0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30

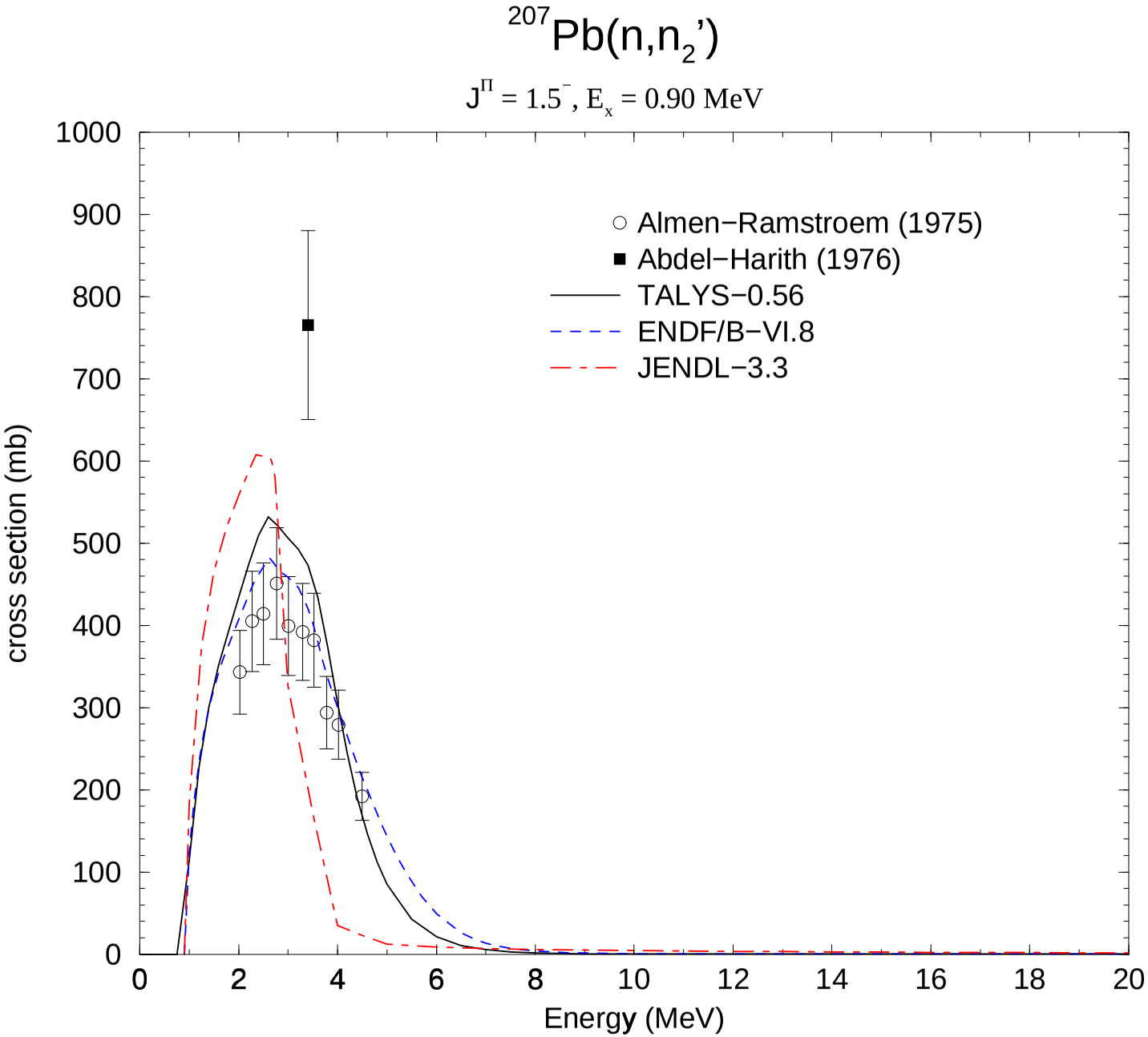
^{208}Pb(n,2nγ)


Level 2 -> Level 0: E$_\gamma$=0.89 MeV
Vonach et al. (1994)
TALYS−0.56
Cross section (mb)
Energy (MeV)
0
100
200
300
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30

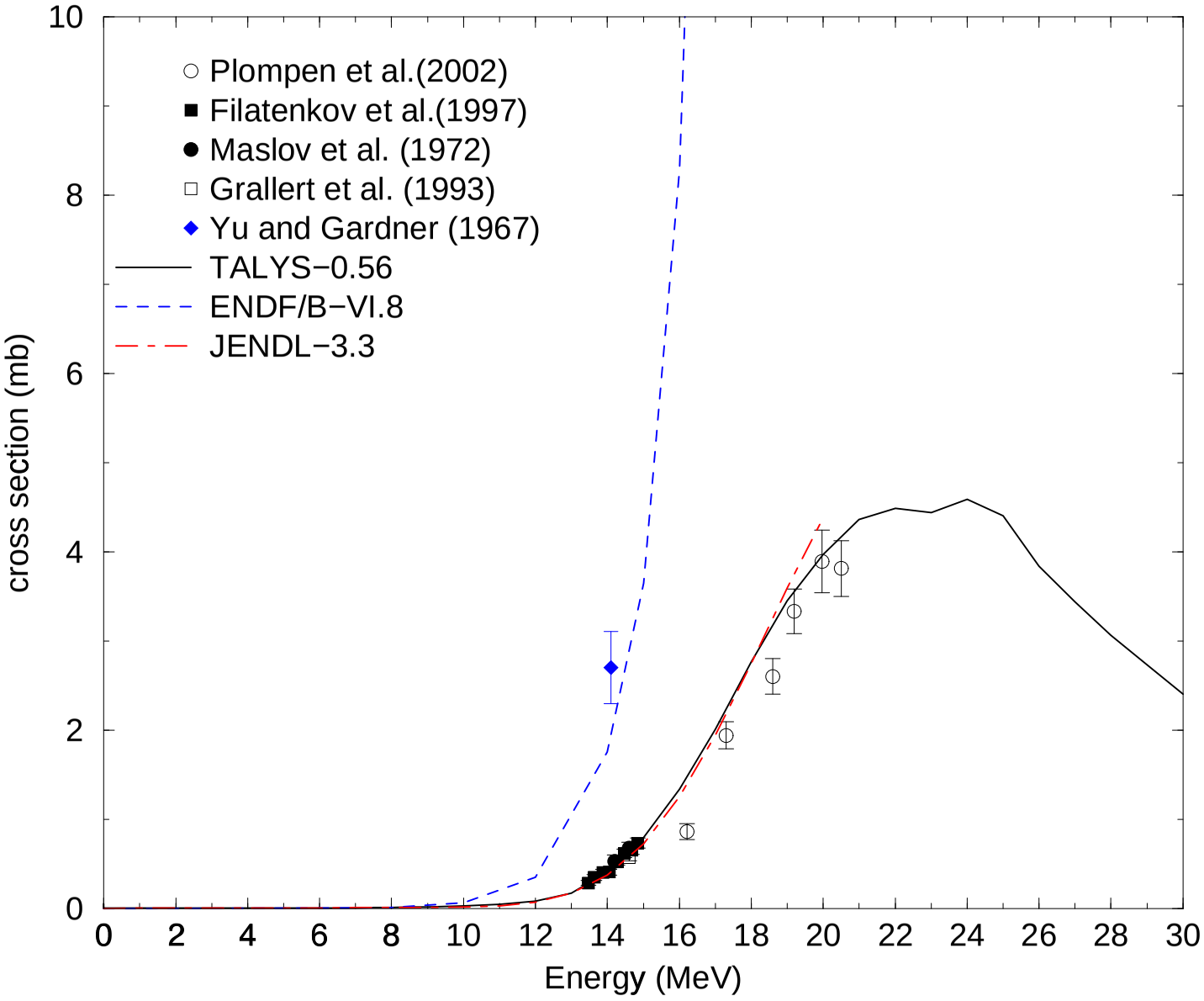
$^{208}Pb(n,p)^{208}Tl$




^{207}Pb(n,n$_1$')
J$^{\Pi}$ = 2.5$^-$, E$_x$ = 0.57 MeV
Almen−Ramstroem (1975)
Abdel−Harith (1976)
TALYS−0.56
ENDF/B−VI.8
JENDL−3.3
cross section (mb)
Energy (MeV)
0
100
200
300
400
500
600
700
800
900
1000
0
2
4
6
8
10
12
14
16
18
20

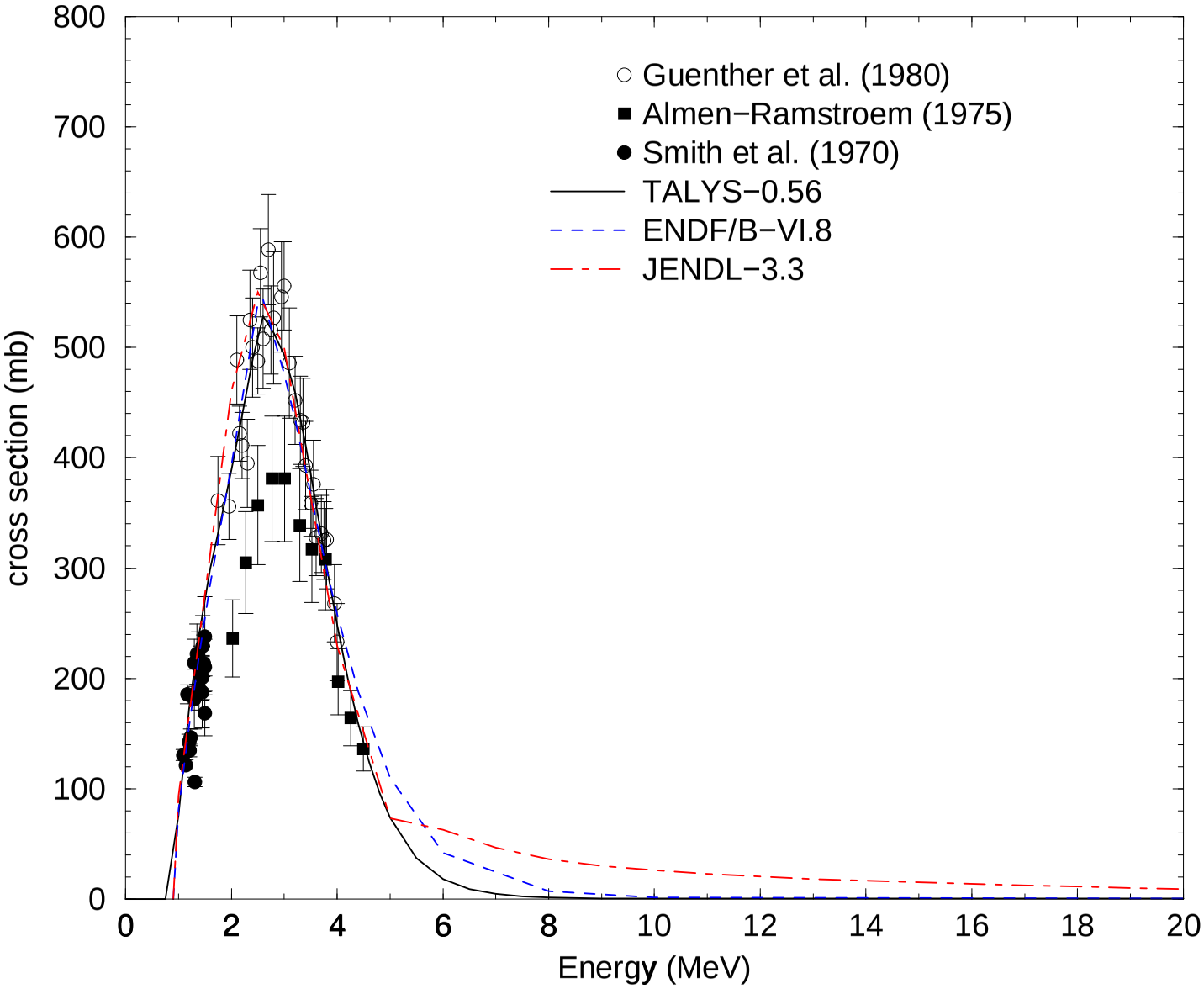

^{207}Pb(n,n$_2$')
J$^{\Pi}$ = 1.5$^-$, E$_x$ = 0.90 MeV
Almen−Ramstroem (1975)
Abdel−Harith (1976)
TALYS−0.56
ENDF/B−VI.8
JENDL−3.3
cross section (mb)
Energy (MeV)
0
100
200
300
400
500
600
700
800
900
1000
0
2
4
6
8
10
12
14
16
18
20

^{206}Pb(n,α)

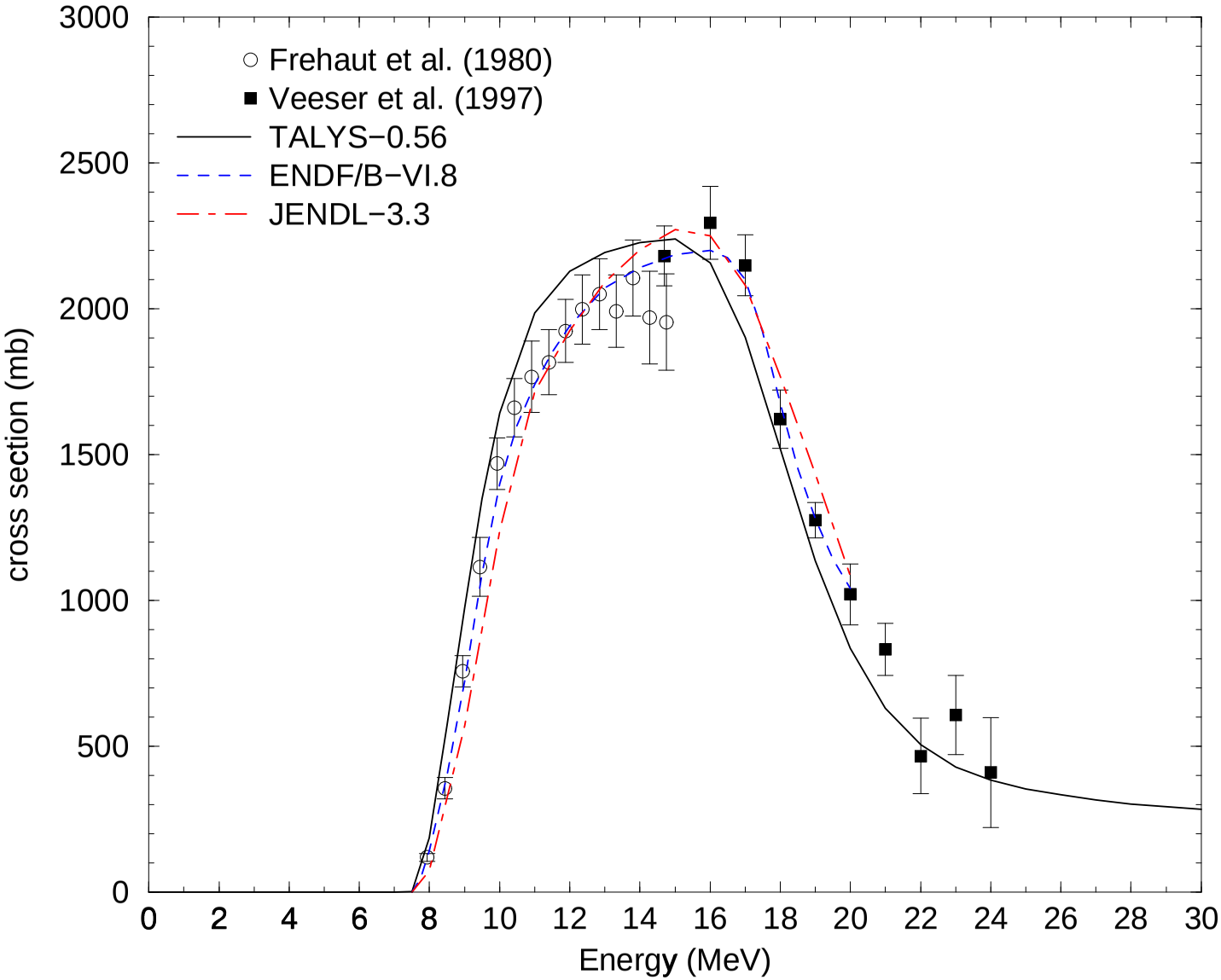

Plompen et al.(2002)
Filatenkov et al.(1997)
Maslov et al. (1972)
Grallert et al. (1993)
Yu and Gardner (1967)
TALYS−0.56
ENDF/B−VI.8
JENDL−3.3
cross section (mb)
Energy (MeV)
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30

$^{209}Bi(n,n_1')$

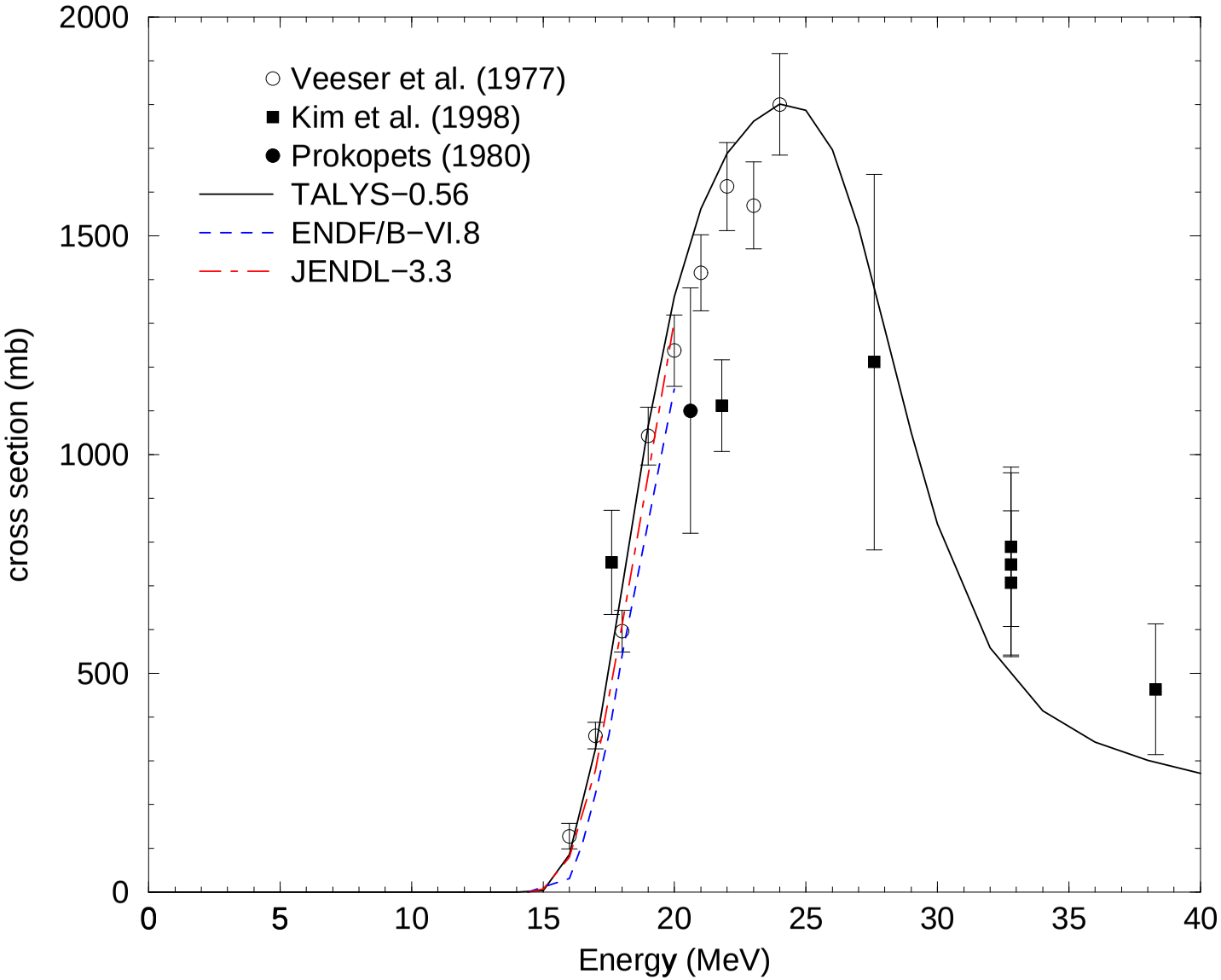
$J^{\Pi} = 3.5^{-}$, $E_x = 0.90$



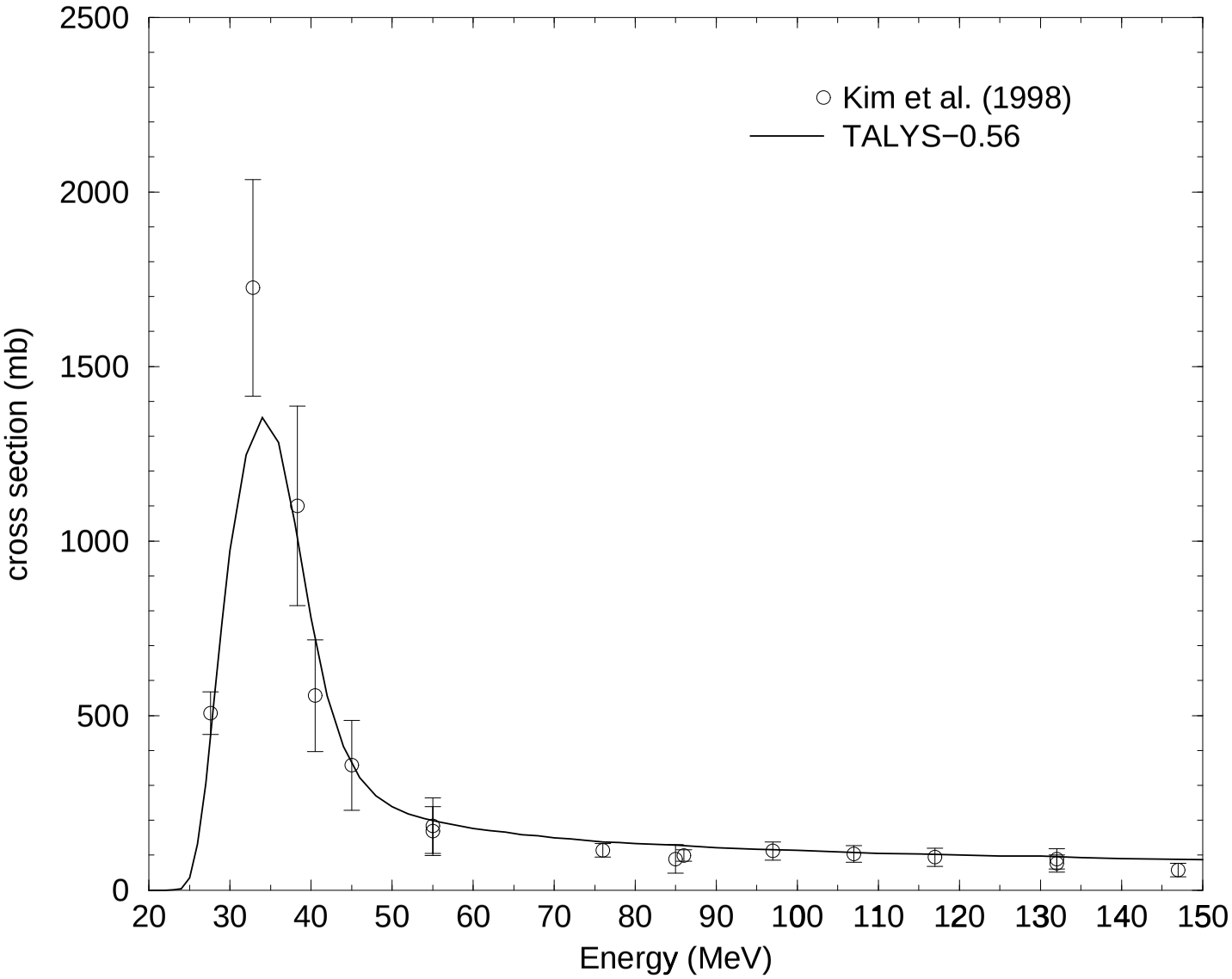
$^{209}Bi(n,2n)^{208}Bi$



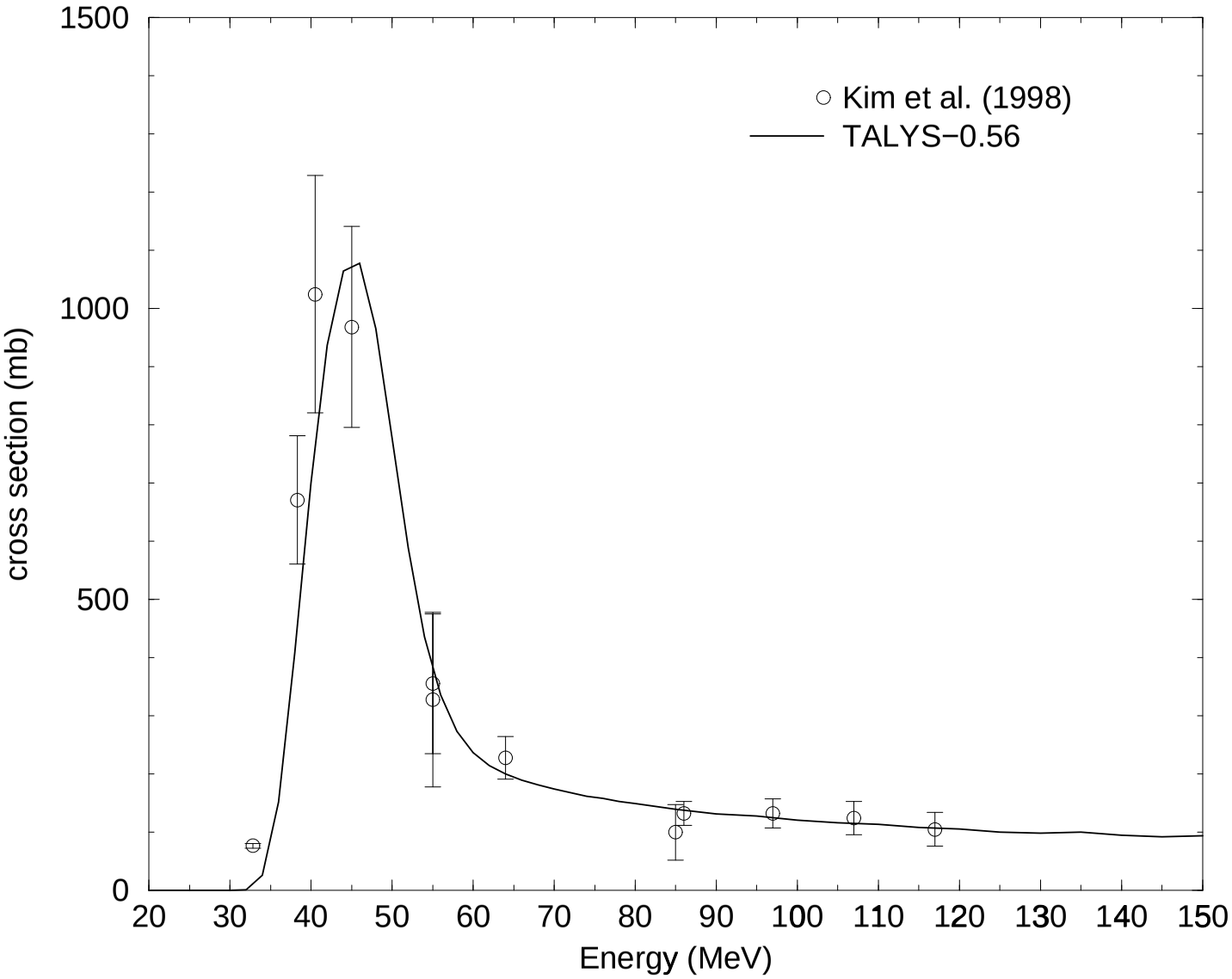
$^{209}Bi(n,3n)^{207}Bi$



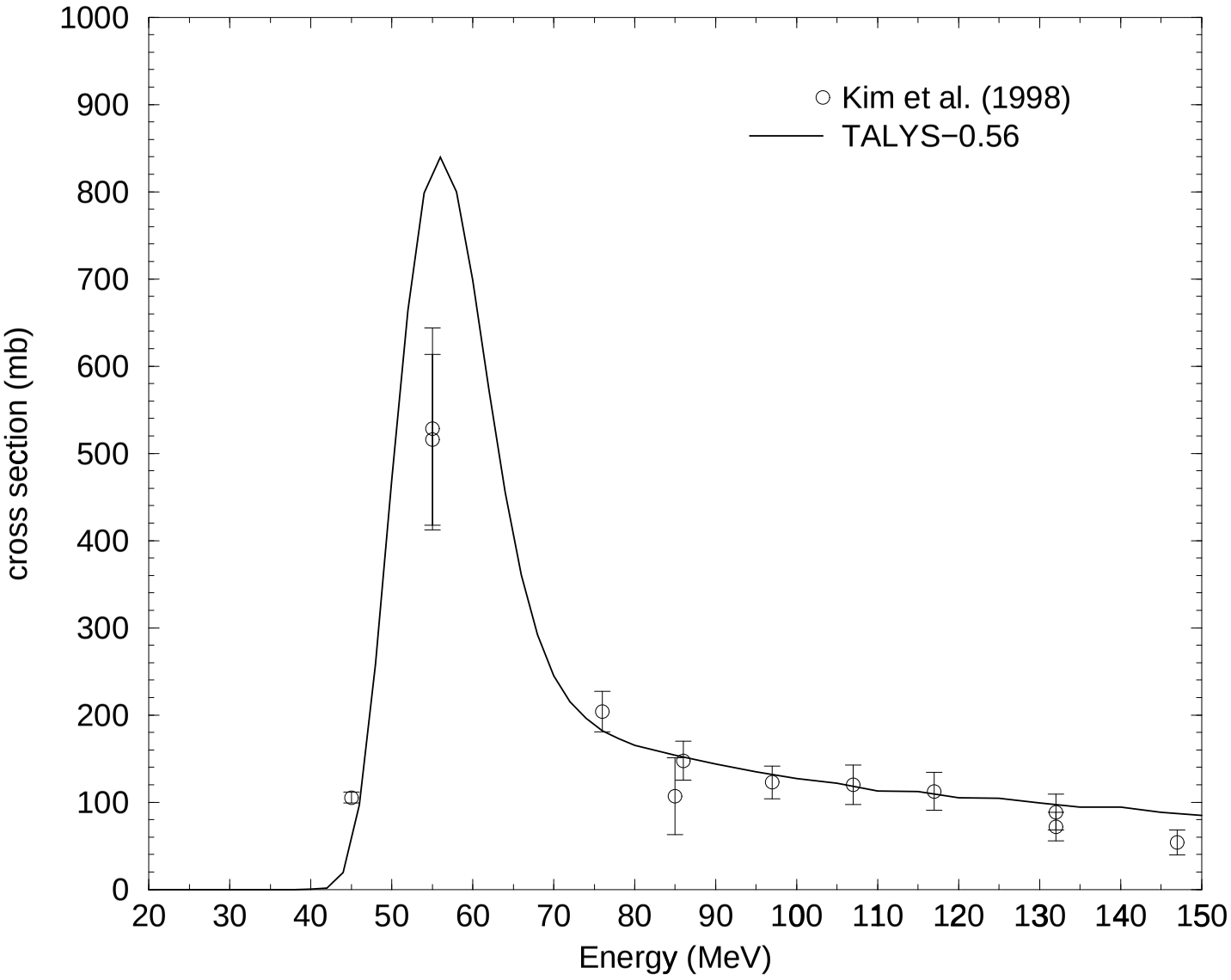
$^{209}Bi(n,4n)^{206}Bi$



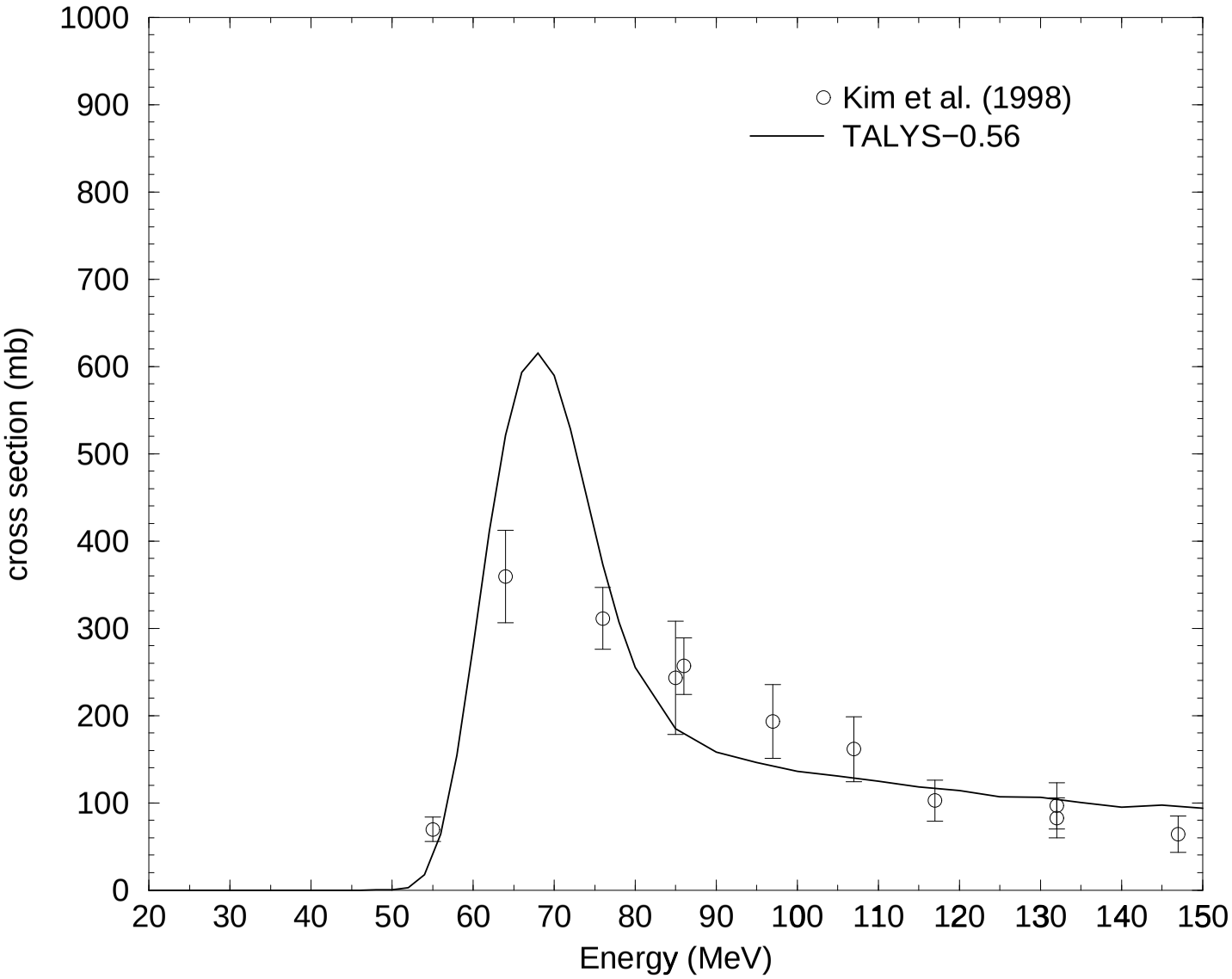
$^{209}Bi(n,5n)^{205}Bi$



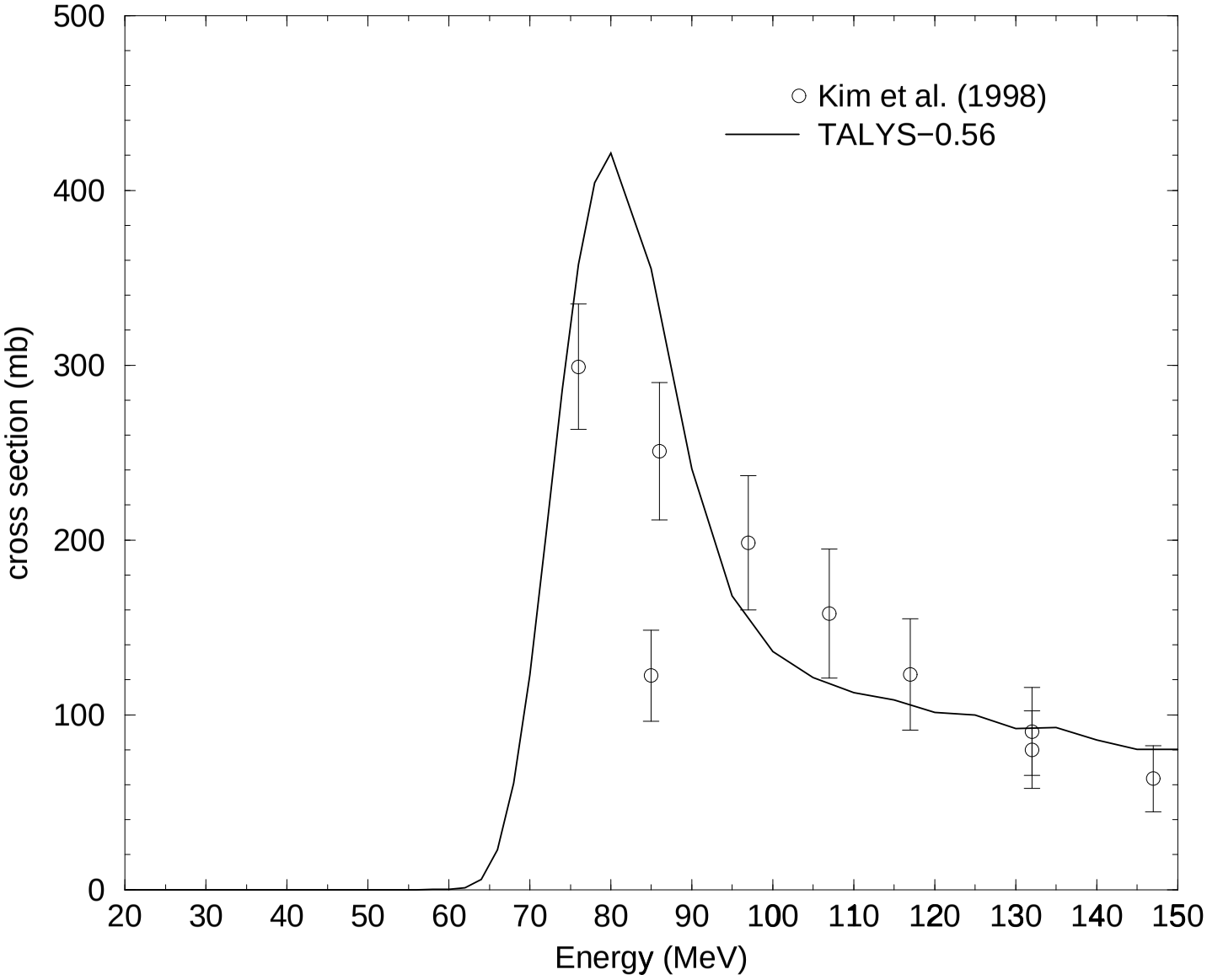
$^{209}Bi(n,6n)^{204}Bi$



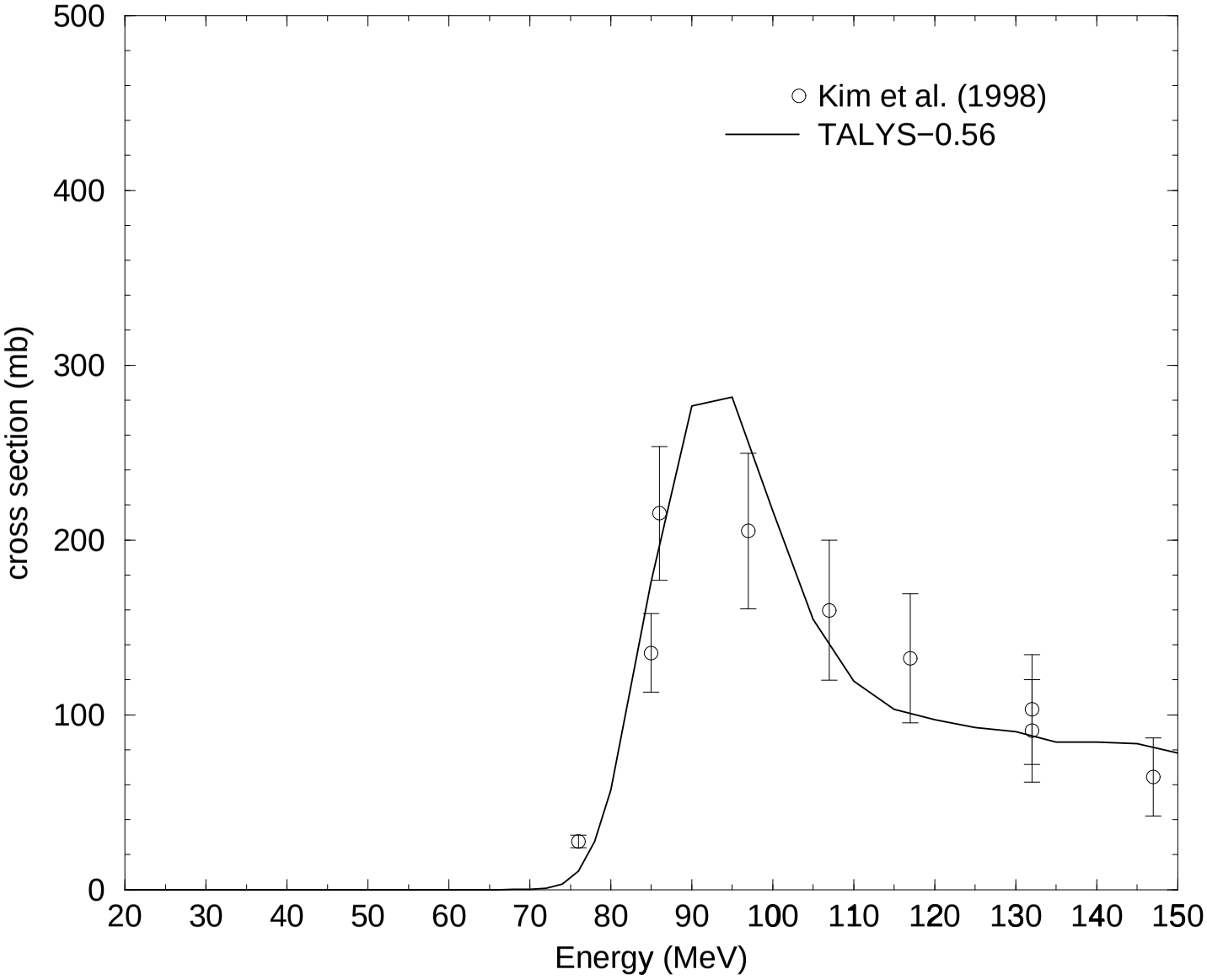
$^{209}Bi(n,7n)^{203}Bi$



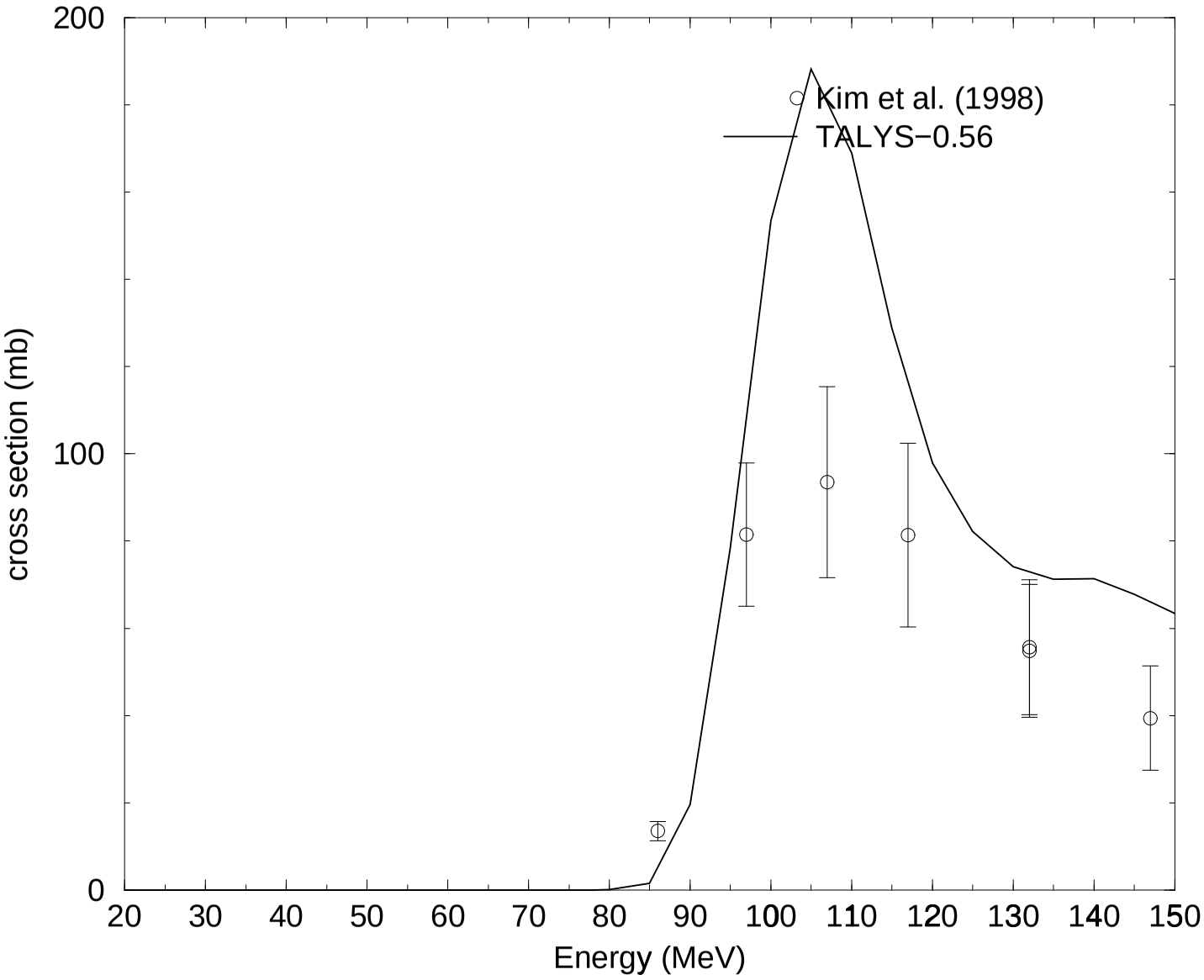
$^{209}Bi(n,8n)^{202}Bi$



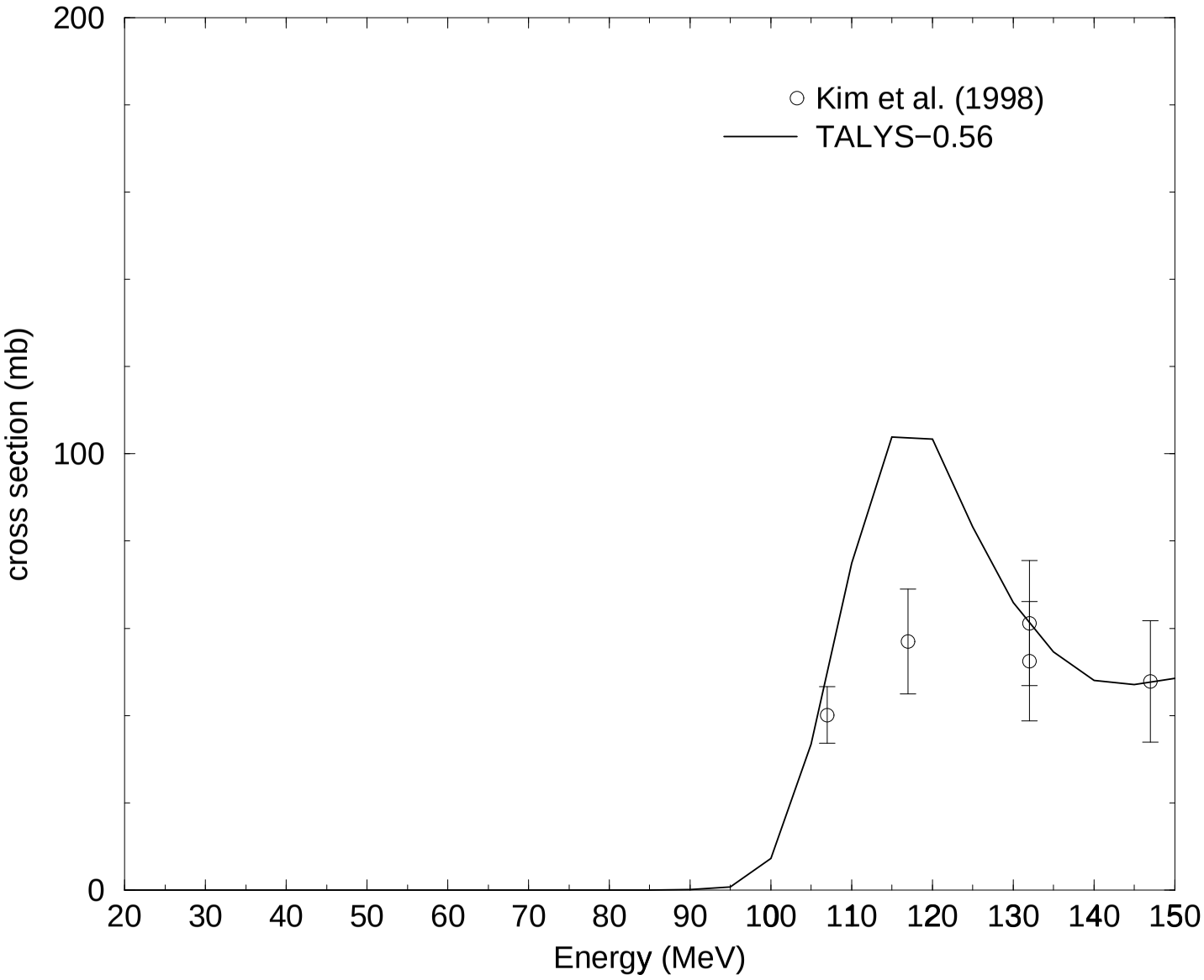
$^{209}Bi(n,9n)^{201}Bi$



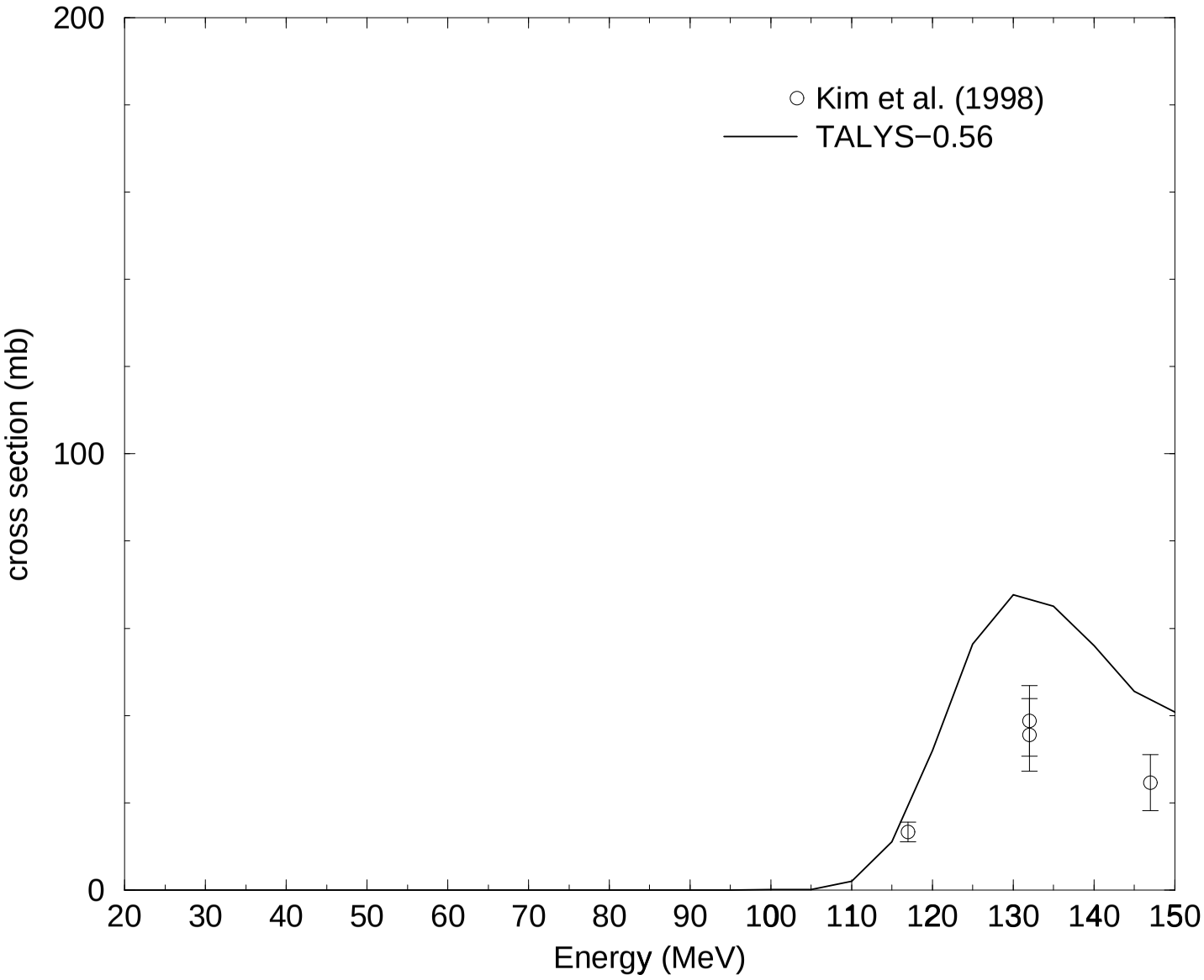
$^{209}Bi(n,10n)^{200}Bi$



$^{209}Bi(n,11n)^{199}Bi$



$^{209}Bi(n,12n)^{198}Bi$



Pb-204

Library	Resonance range	URR - 20 MeV	> 20 MeV
ENDF/B-VI.8	—	—	—
JENDL-3.3	Mizumoto (1987)	Mizumoto(1987), Shibata(2001)	—
JEFF-3.0	Mizumoto (1987)	Mizumoto(1987)	—
BROND-2.2	Blokhin (1984)	Blokhin(1990)	—
NRG	Larson (1989)	Koning(2003)	Koning(2003)

Pb-206

Library	Resonance range	URR - 20 MeV	> 20 MeV
ENDF/B-VI.8	Larson (1989)	Fu (1989)	Chadwick (1996)
JENDL-3.3	Mizumoto (1987)	Mizumoto(1987), Shibata(2001)	—
JEFF-3.0	Mizumoto (1987)	Mizumoto(1987)	—
BROND-2.2	Blokhin (1984)	Blokhin(1990)	—
NRG	Larson (1989)	Koning(2003)	Koning(2003)

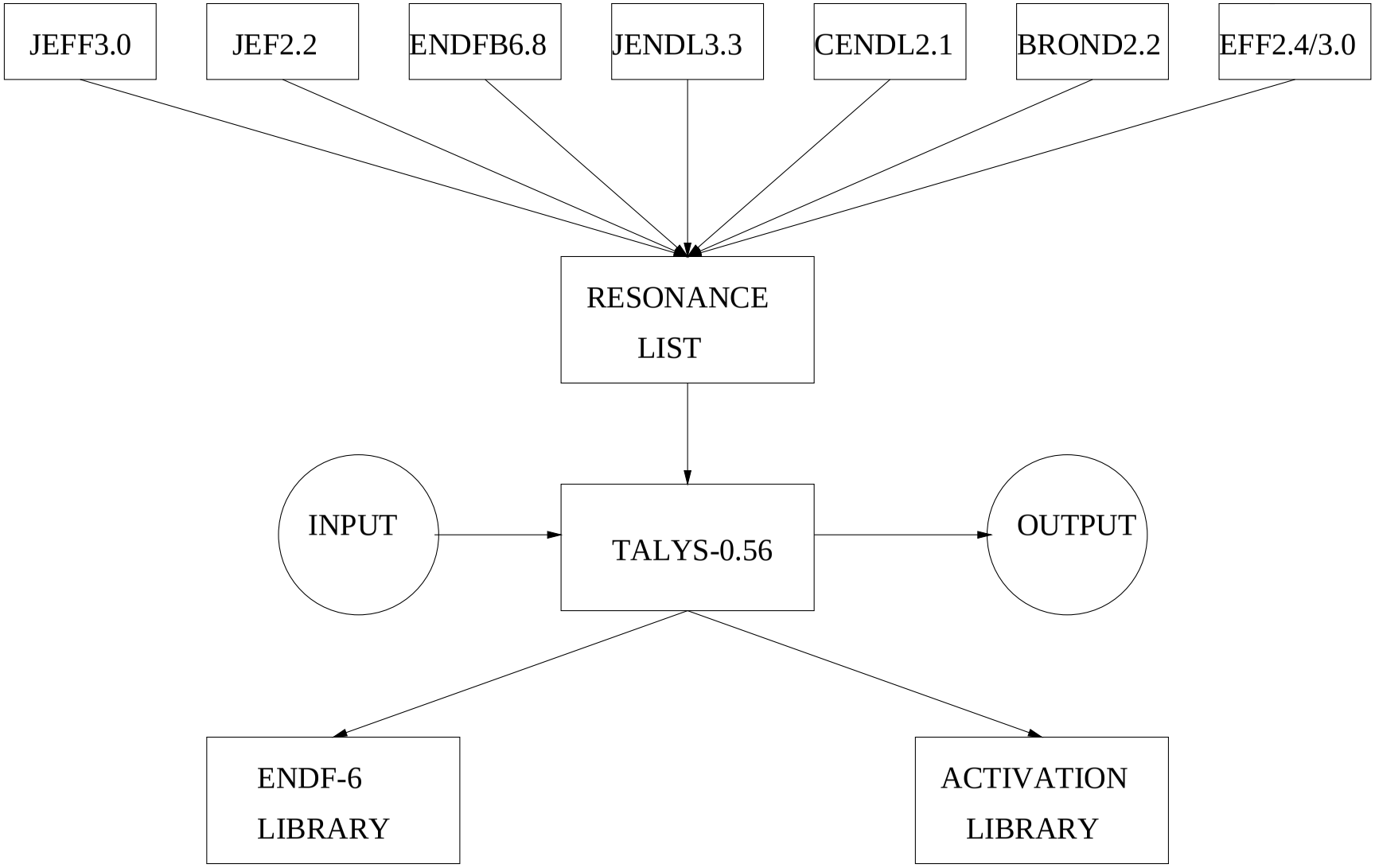
	Library	Resonance range	URR - 20 MeV	> 20 MeV
Pb-207	ENDF/B-VI.8	Larson (1989)	Fu (1989)	Chadwick (1996)
	JENDL-3.3	Mizumoto (1987)	Mizumoto(1987), Shibata(2001)	—
	JEFF-3.0	Mizumoto (1987)	Mizumoto(1987)	—
	BROND-2.2	Blokhin (1984)	Blokhin(1990)	—
	NRG	Larson (1989)	Koning(2003)	Koning(2003)

	Library	Resonance range	URR - 20 MeV	> 20 MeV
Pb-208	ENDF/B-VI.8	Larson (1989)	Young (1996)	Chadwick (1996)
	JENDL-3.3	Larson (1989)	Mizumoto(1987), Shibata(2001)	—
	JEFF-3.0	Larson (1989)	Mizumoto(1987)	—
	BROND-2.2	Blokhin (1984)	Blokhin(1990)	—
	NRG	Larson (1989)	Koning(2003)	Koning(2003)

	Library	Resonance range	URR - 20 MeV	> 20 MeV
Bi-209	ENDF/B-VI.8	Smith (1989)	Smith (1989)	Chadwick (1998)
	JENDL-3.3	Yamamuro (1989)	Yamamuro(1989), Shibata(2001)	—
	JEFF-3.0	Smith (1989)	Smith (1989)	—
	BROND-2.2	Blokhin (1984)	Blokhin(1990)	—
	NRG	Yamamuro (1989)	Koning(2003)	Koning(2003)

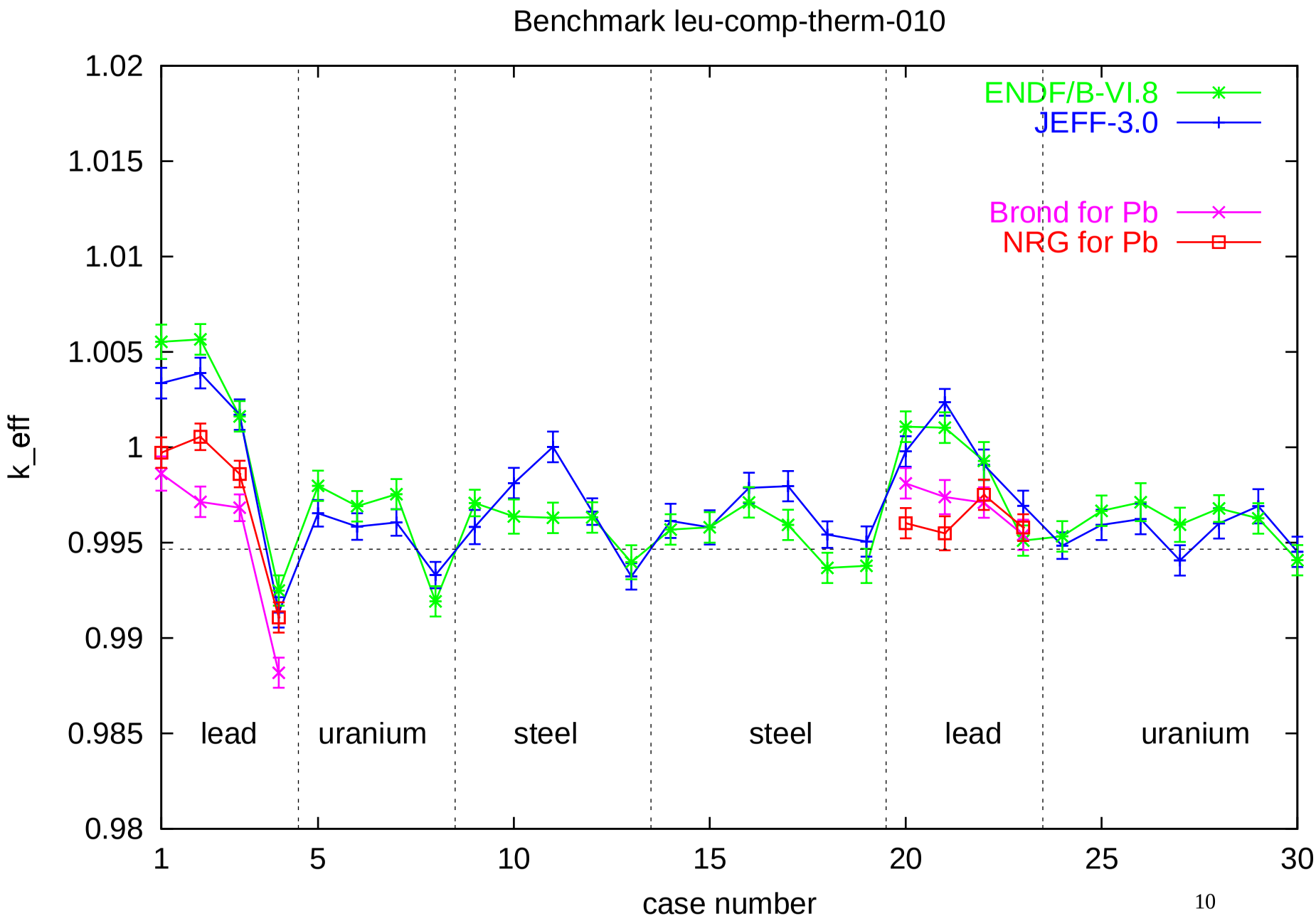
ENDF-6 philosophy used by TALYS

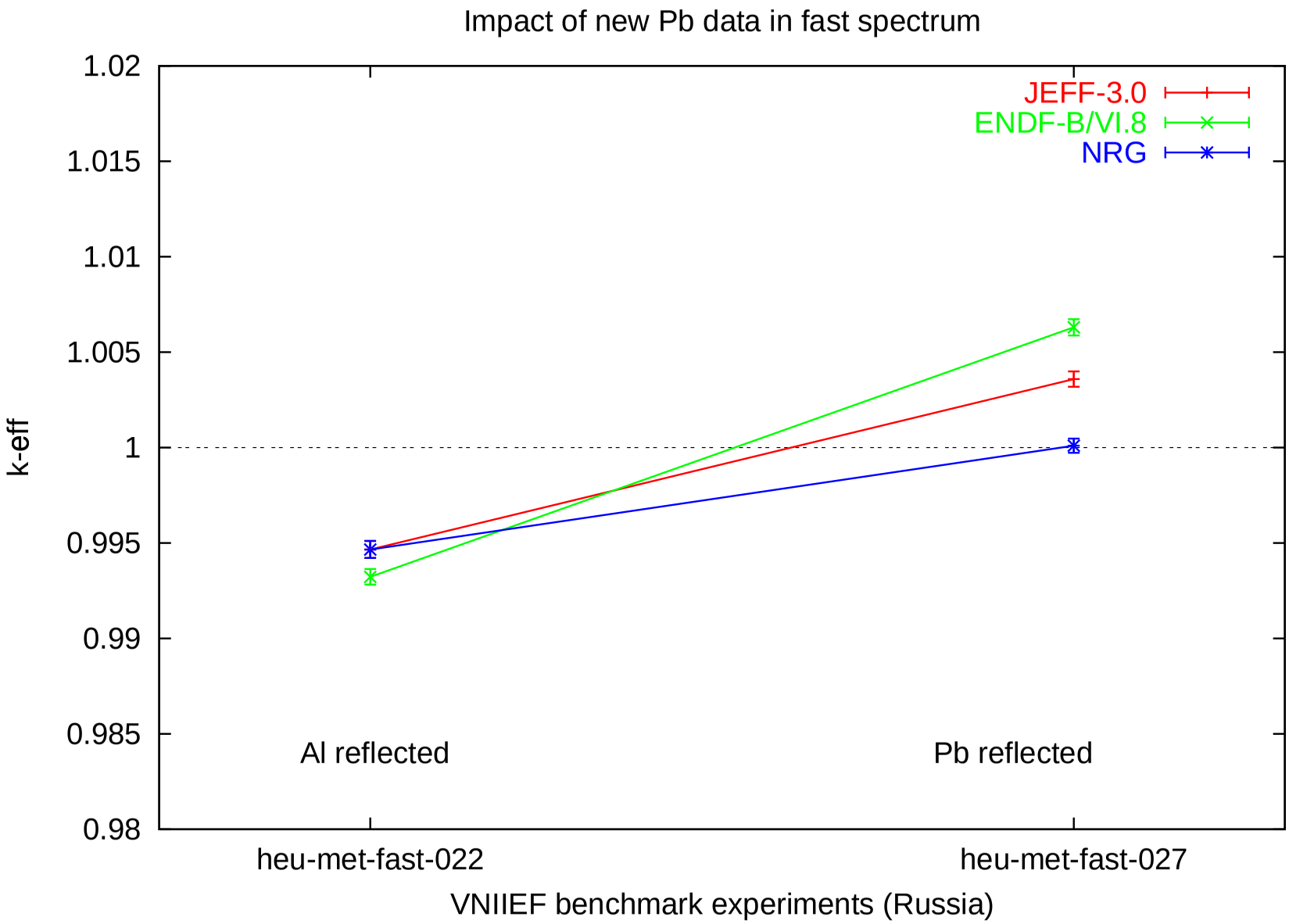
- Data file should be complete, i.e. should include all required transport and activation data.
- Use one and the same file structure for every isotope:
  - MF1
  - MF2: use best possible resonance data from other library.
  - MF3: include all open reaction channels. Use MT5 for $(n, \gamma n)$ and re action cross section above 20 MeV.
  - MF4: Use only for elastic scattering angular distribution (MT2).


JEFF3.0
JEF2.2
ENDFB6.8
JENDL3.3
CENDL2.1
BROND2.2
EFF2.4/3.0
RESONANCE LIST
INPUT
TALYS-0.56
OUTPUT
ENDF-6 LIBRARY
ACTIVATION LIBRARY

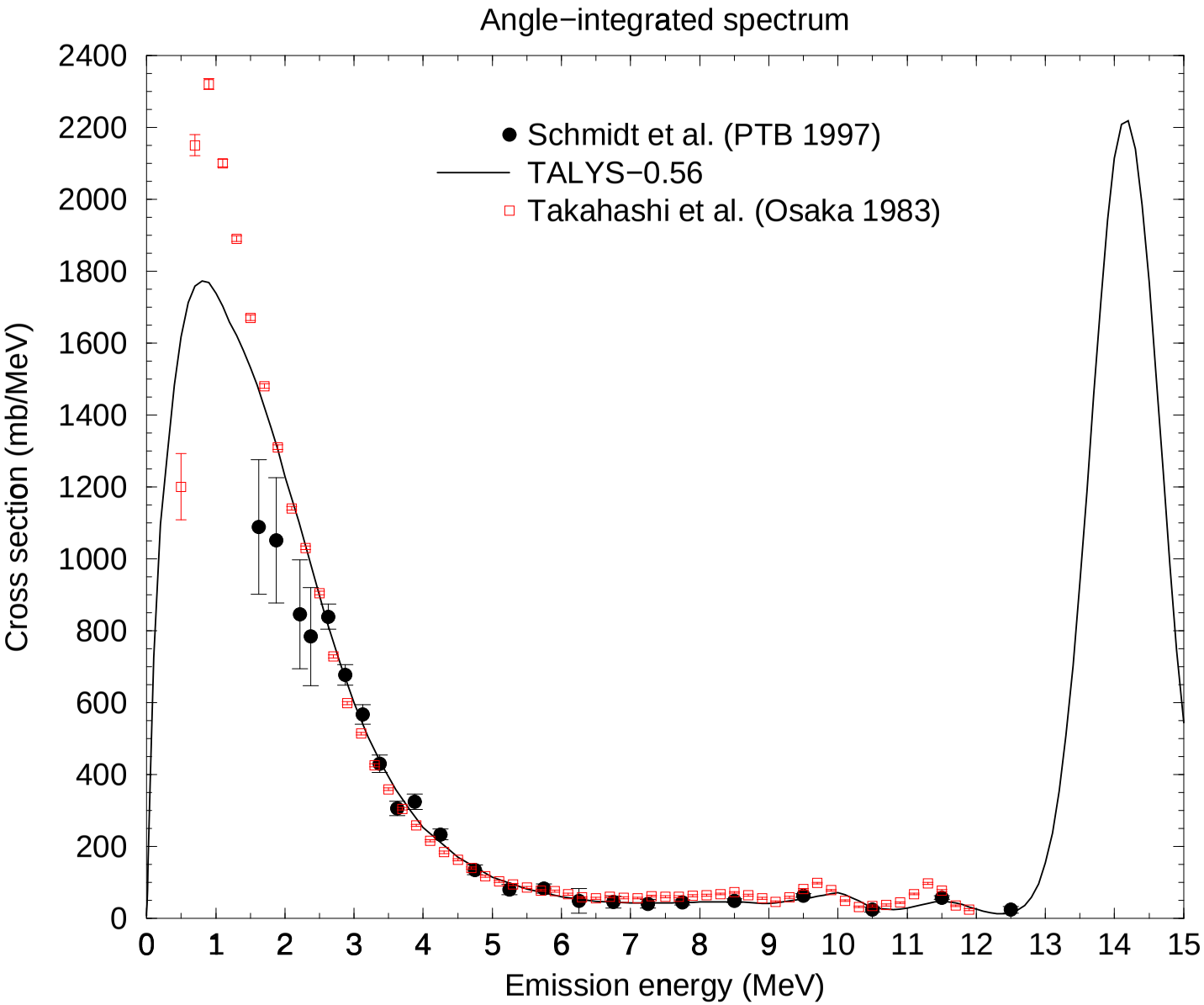
ENDF-6 philosophy used by TALYS

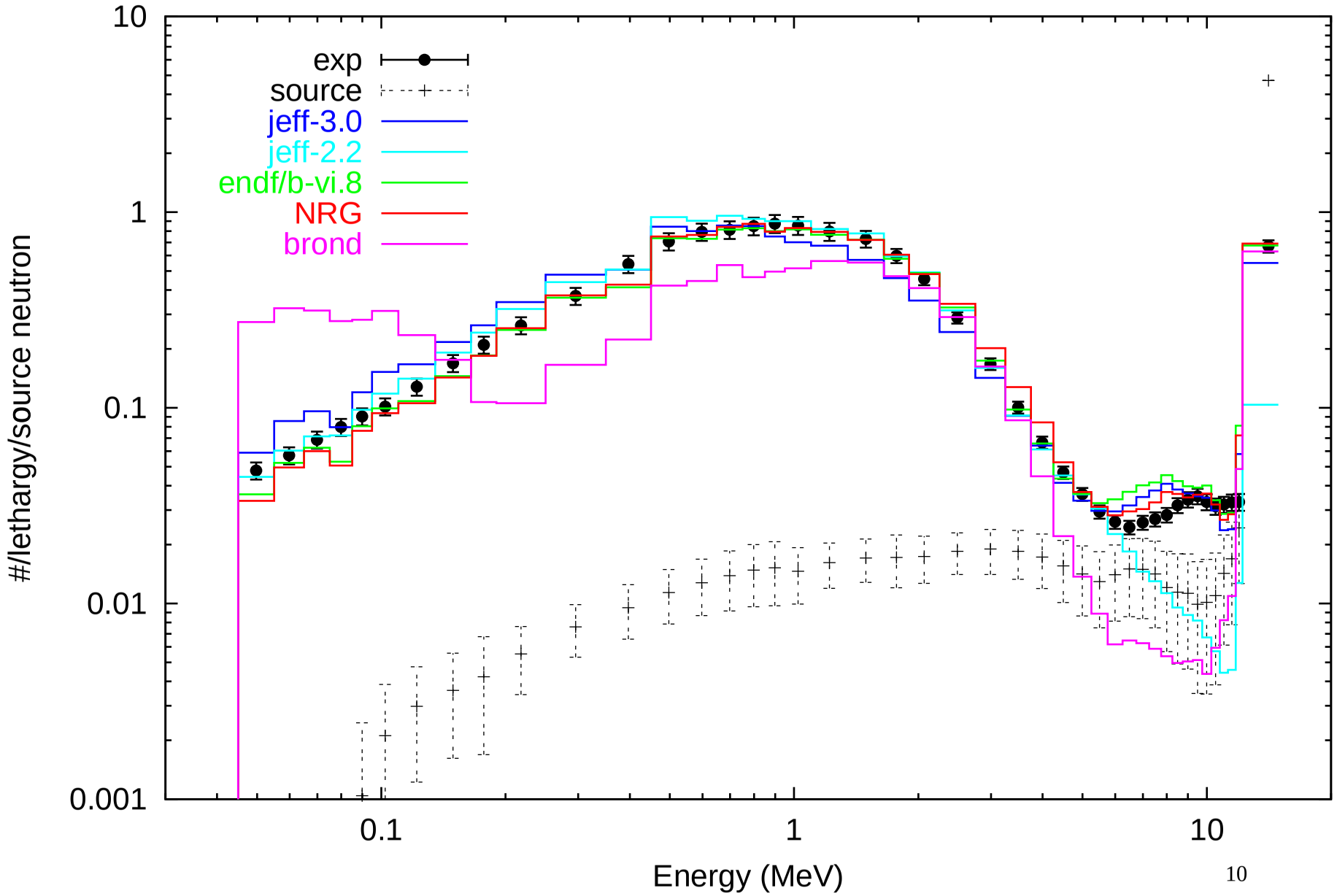
- MF6: Use for all discrete angular distributions (apart from elastic), all double -differential exclusive spectra, all discrete and continuum photon production and recoils. Above 20 MeV, use MF6/MT5 for particle production cross sections and spectra and residual production cross sections.
- MF8: In case of isomers: point to MF (6, 9 or 10) where info exists.
- MF9: Isomeric cross sections for non-threshold reactions.
- MF10: Isomeric cross sections for threshold reactions.

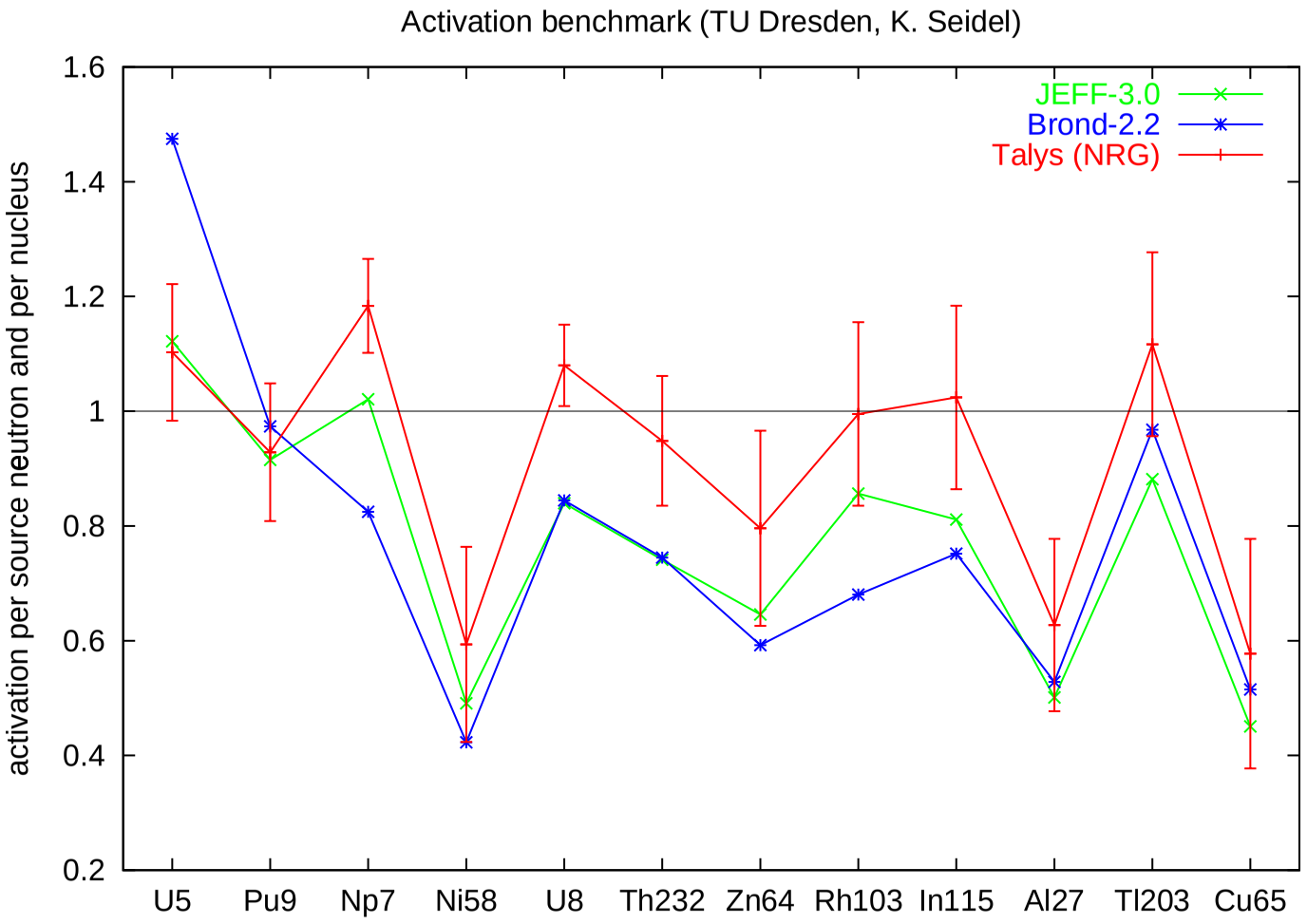

Benchmark leu-comp-therm-010
ENDF/B-VI.8
JEFF-3.0
Brond for Pb
NRG for Pb
k_eff
1.02
1.015
1.01
1.005
1
0.995
0.99
0.985
0.98
lead
uranium
steel
steel
lead
uranium
1
5
10
15
20
25
30
case number

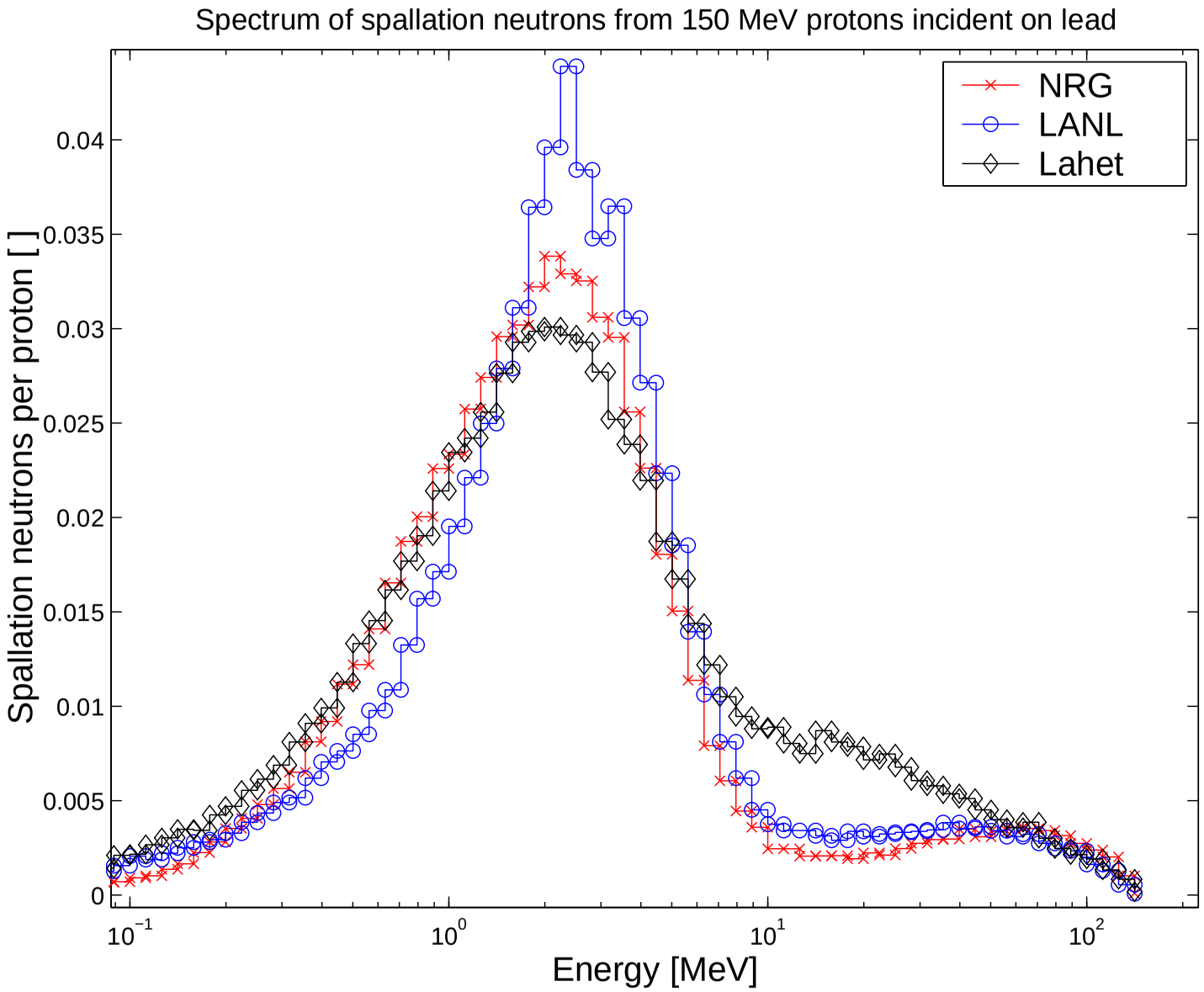

Impact of new Pb data in fast spectrum
1.02
1.015
1.01
1.005
1
0.995
0.99
0.985
0.98
k-eff
JEFF-3.0
ENDF-B/VI.8
NRG
Al reflected
Pb reflected
heu-met-fast-022
heu-met-fast-027
VNIIEF benchmark experiments (Russia)

Pb(n,xn) at 14.23 MeV



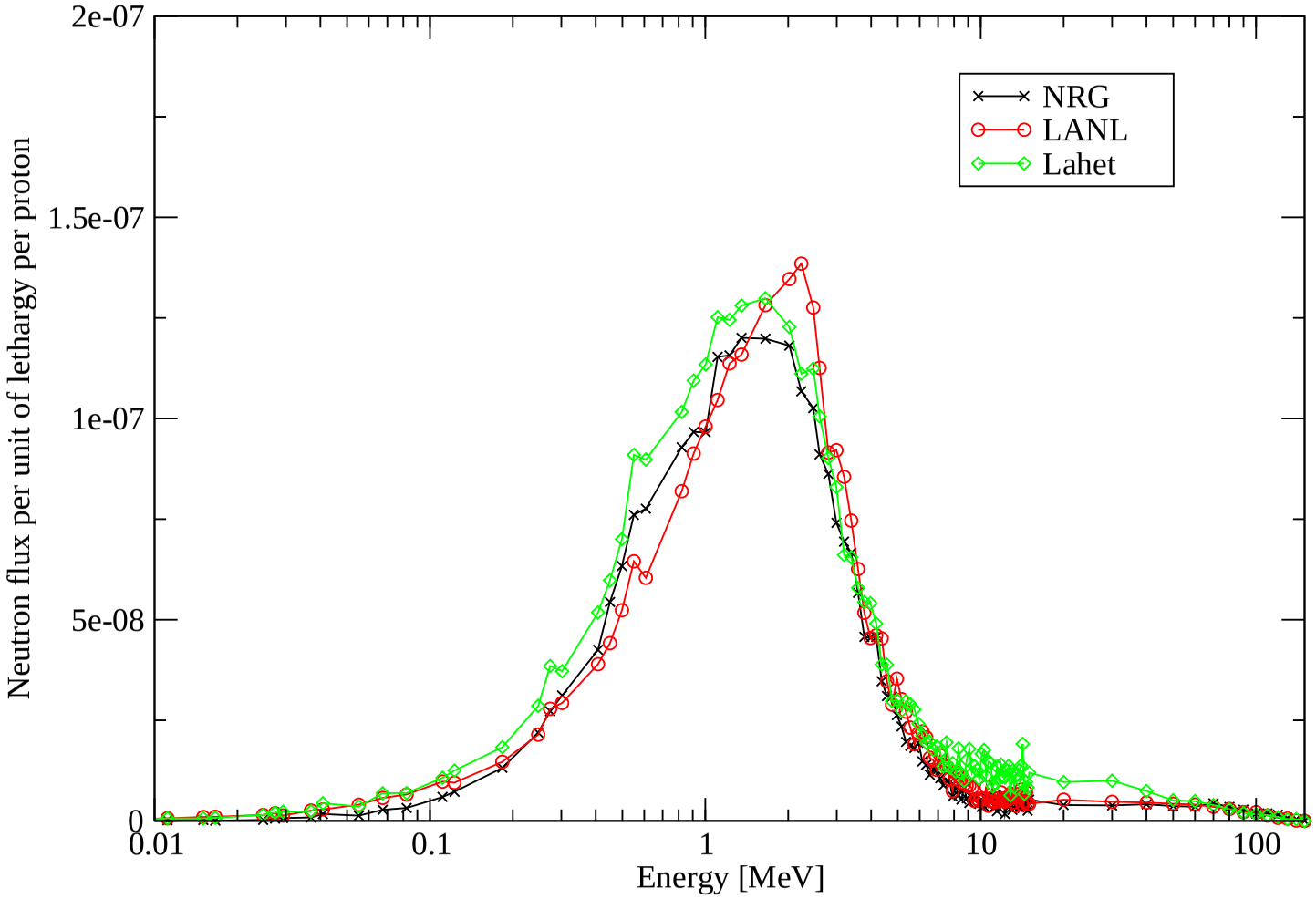

TU-Dresden (Seidel) benchmark: leakage through Pb-sphere (2.5 - 25cm)
exp
source
jeff-3.0
jeff-2.2
endf/b-vi.8
NRG
brond
#/lethargy/source neutron
Energy (MeV)
10
1
0.1
0.01
0.001
0.1
1
10


Activation benchmark (TU Dresden, K. Seidel)
activation per source neutron and per nucleus
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
JEFF-3.0
Brond-2.2
Talys (NRG)
U5
Pu9
Np7
Ni58
U8
Th232
Zn64
Rh103
In115
Al27
Tl203
Cu65


Spectrum of spallation neutrons from 150 MeV protons incident on lead
NRG
LANL
Lahet
Spallation neutrons per proton [ ]
0
0.005
0.01
0.015
0.02
0.025
0.03
0.035
0.04
10^{-1}
10^{0}
10^{1}
10^{2}
Energy [MeV]

Neutron spectrum outside lead buffer

for incident protons of 150 MeV



Conclusions

New ENDF-6 formatted data libraries for ^{204,206,207,208}Pb and ^{209}Bi

- Older Pb and Bi evaluations from the major world libraries (ENDF/B-VI, JENDL and JEFF) all suffer from problems below 20 MeV, at the microscopic and benchmark level.
- Existing experimental data < 20 MeV consistently analyzed with TALYS code.
- New experiments/model developments used for 20 < E < 200 MeV.
- Evaluated datafiles improved from unresolved resonance range up to 200 MeV with TALYS:
  - Consistent good description of many reaction channels.
  - Complete ENDF-6 datafile including all secondary distributions, photons, etc.
  - Successful NJOY processing.

Conclusions

- Data files will be proposed for the next version of JEFF.
- First integral benchmarks with MCNP(X) encouraging.
- Sensitivy studies and further integral benchmarks should reveal whether good is good enough.
- TALYS is ready to give other nuclides the same treatment (All Ca, Sc, Fe, and Ge isotopes under construction).