Neutron-induced reactions on U and Th – a new approach via AMS

in collaboration with:

neutrons

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- IRMM / Geel: A. Plompen, A. Krasa
- IKI Budapest: T. Belgya, L. Szentmiklosi
- TSL Uppsala: A. Prokofiev et al.
- VERA (Vienna): P. Steier, S. Winkler, C. Lederer
- ANU (Canberra): L.K. Fifield, S. Tims, M. Srncik
- ETH Zurich: M. Christl, J. Lachner
- ANSTO: M. Hotchkis
- Univ. Heidelberg: J. Lippold
- IAEA: R. Capote , V. Semkova

A. Wallner

The Australian National University (ANU)

AMS

Australian National University

European

European Commission
Joint Research Centre
Institute for Reference Materials and Measurements

EFNUDAT
European Facilities for Nuclear Data Measurements

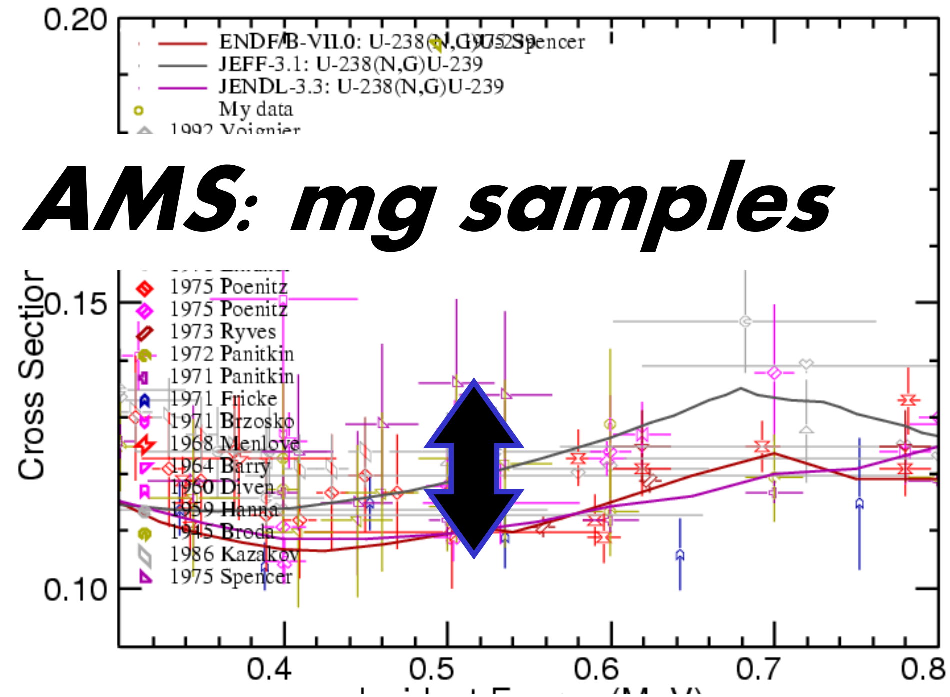
AMS at VERA / ANU / ETH / ANSTO – neutron activations at IRMM / KIT / BRR / PTB

- **Neutron capture cross section of ^{235}U by activation and detection of ^{236}U by AMS**
- **Thermal neutron capture cross section of ^{235}U by activation and detection of ^{236}U by AMS** (EURATOM I3 project; collaboration IKI Budapest, Hungary / VERA, Univ. of
- **Definition of a standard neutron field with the ^{7}Li(p,n)^{7}Be reaction at E_p=1912 keV**
- **Cross section measurements of ^{204}Pb(n,3n)^{202g}Pb with the AMS technique** (EU
- **Cross section measurements of ^{232}Th(n,2n)^{231}Th, ^{232}Th(n,3n)^{230}Th and ^{238}U(n,3n)^{236}U with the AMS technique** (EU programme – EU-project EUFRAT, IRMM Geel, Belgium / VERA, Univ. of Vienna) 2010 - …
- **Neutron capture in the low MeV energy range on ^{238}U and ^{232}Th measured with AMS** (EU programme – EU-project EUFRAT, IRMM Geel, Belgium / VERA, Univ. of Vienna and ANSTO, Australia) 2011 - …

Australian National University

Motivation: $^{238}U(n,\gamma)/^{232}Th(n,\gamma)$: E_n > thermal

- ratio of capture to fission of the fissile isotopes
- existing data via TOF and detections of prompt γ-rays;
- multiple scattering corrections due to large sample size in previous exp.? (e.g. 2 gram)
- HPRL (NEA) listed = highest priority
- large discrepancies above thermal E_n
- k_{eff} in reactor technology: even 5-10% discrepancy in cross section too much
- very few exp. data above thermal energies
- activation + AMS is an independent method; no influence from fission channel



AMS: mg samples

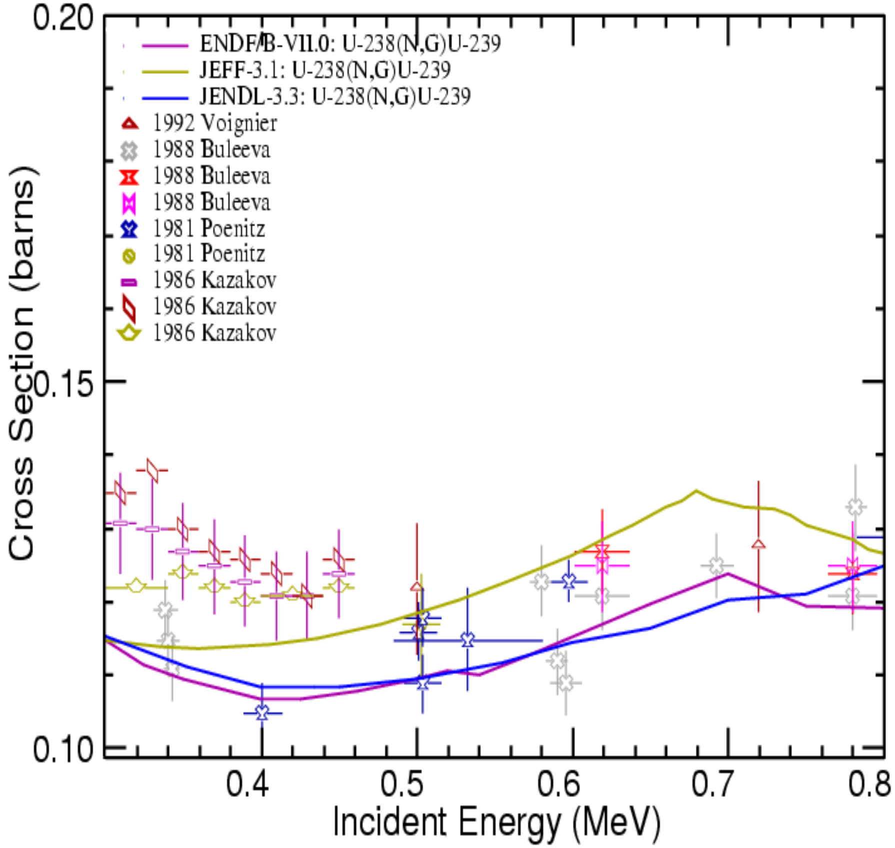
AMS: atom counting

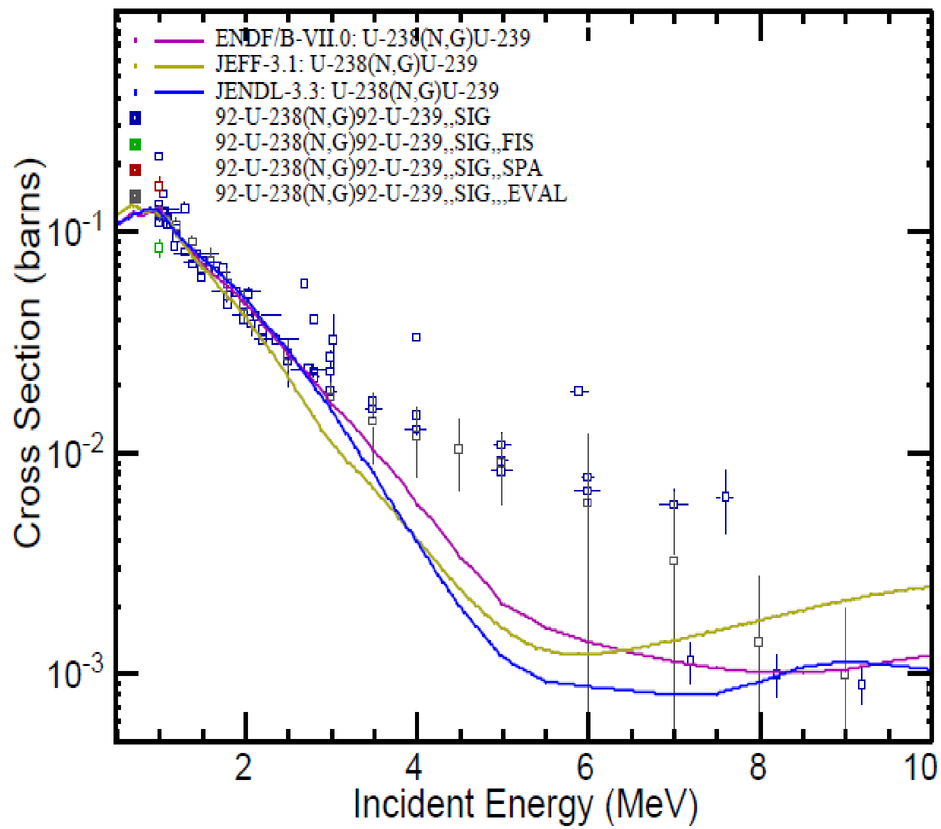
- *new method* → *does not rely on previous drawbacks*
  → *simple direct method!*

$^{238}U(n,\gamma)^{239}U$

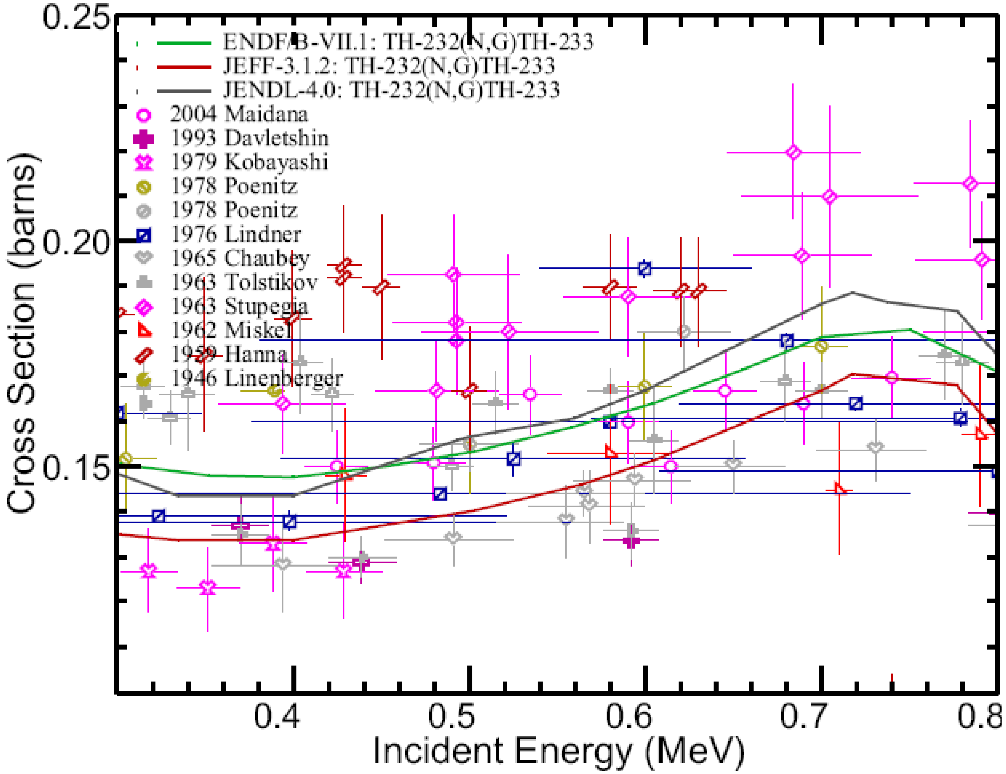
EXFOR database

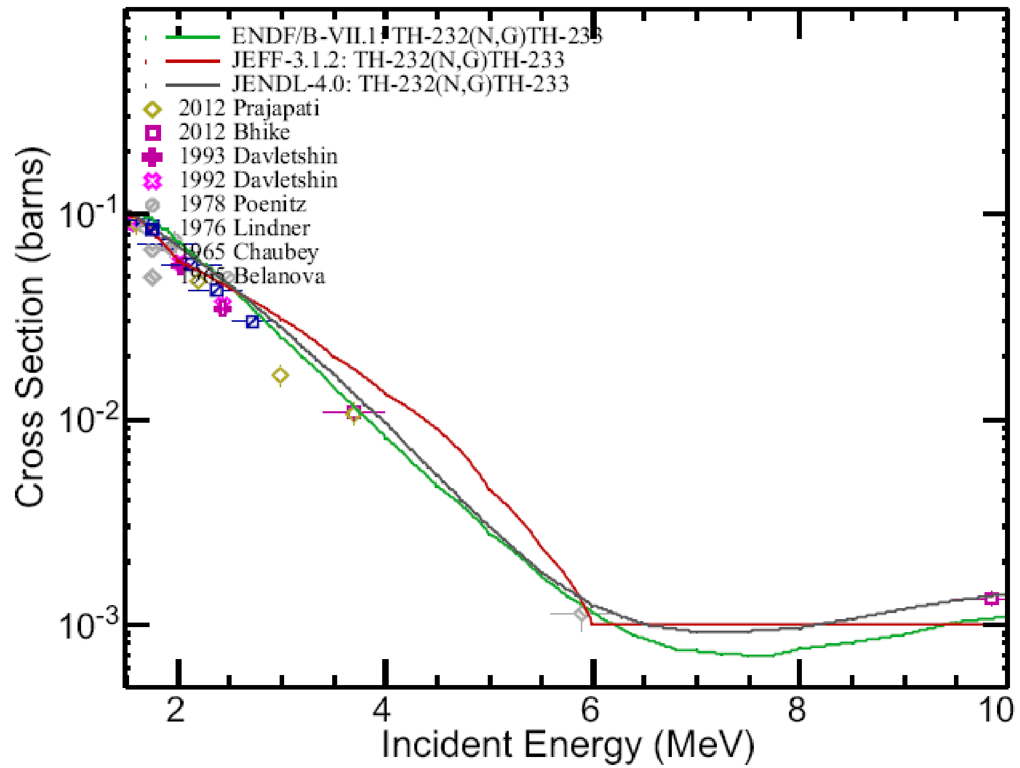
0.3 – 10 MeV





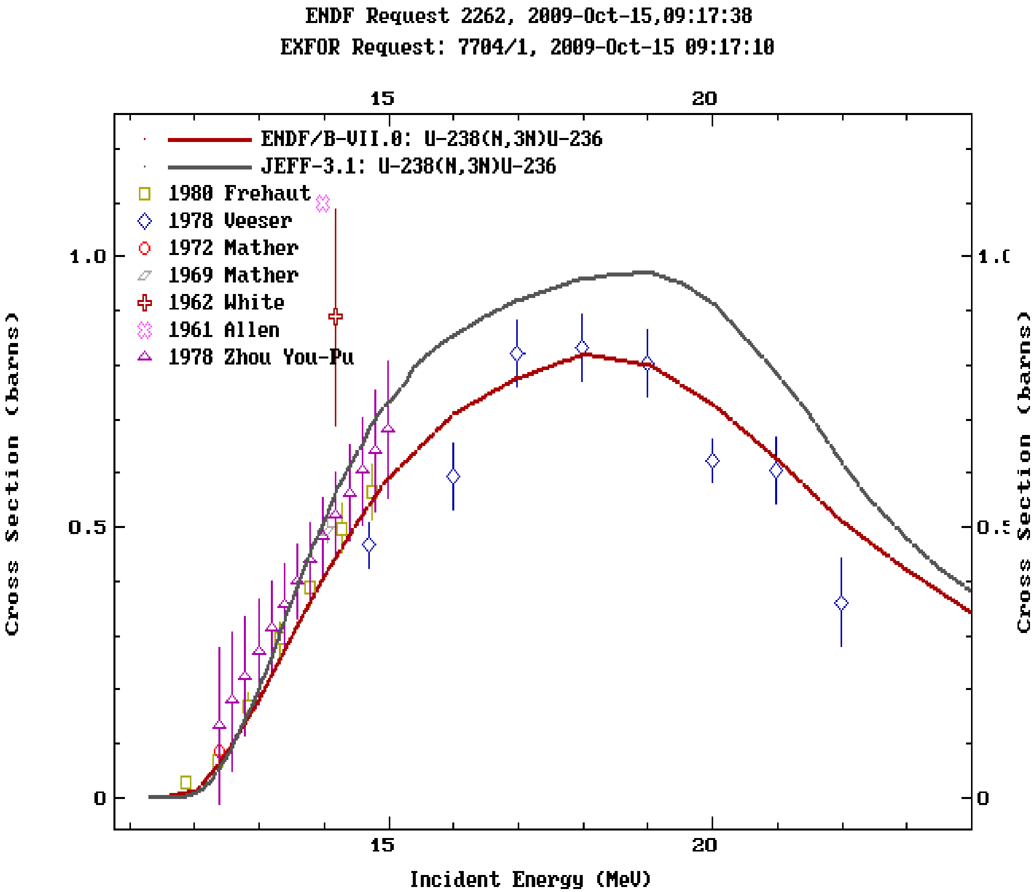
$^{232}Th(n,\gamma)^{233}Th$

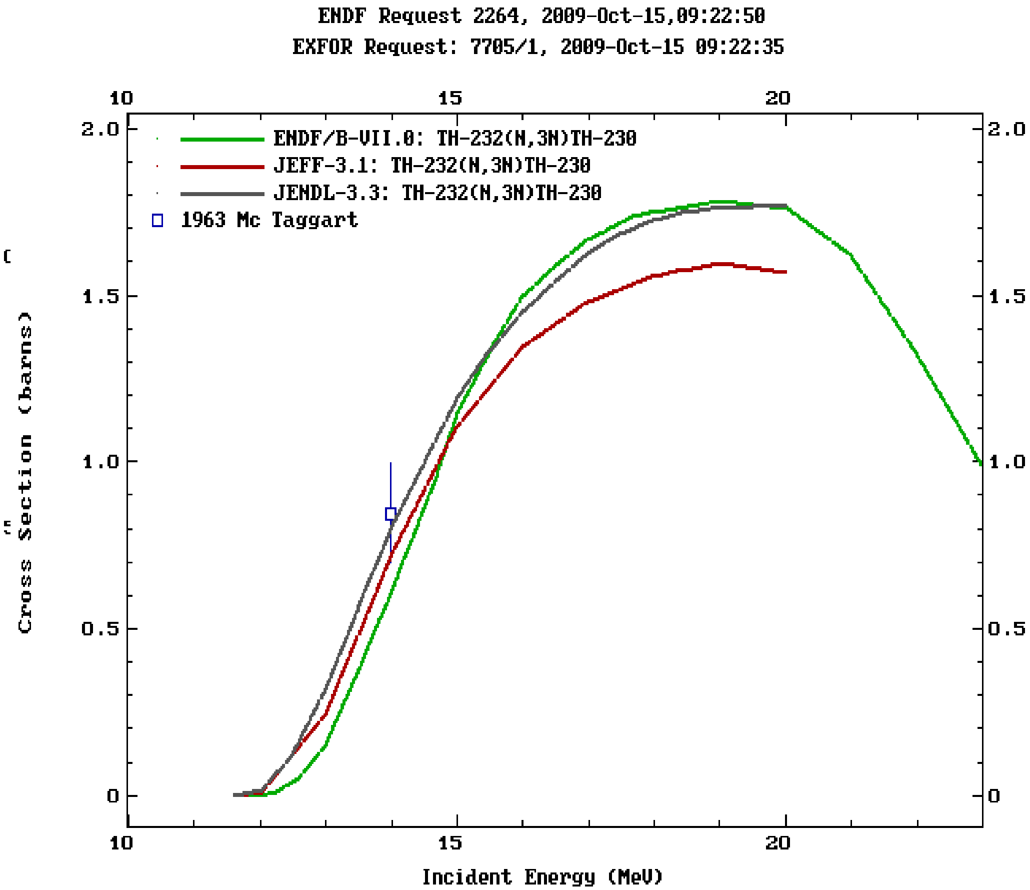


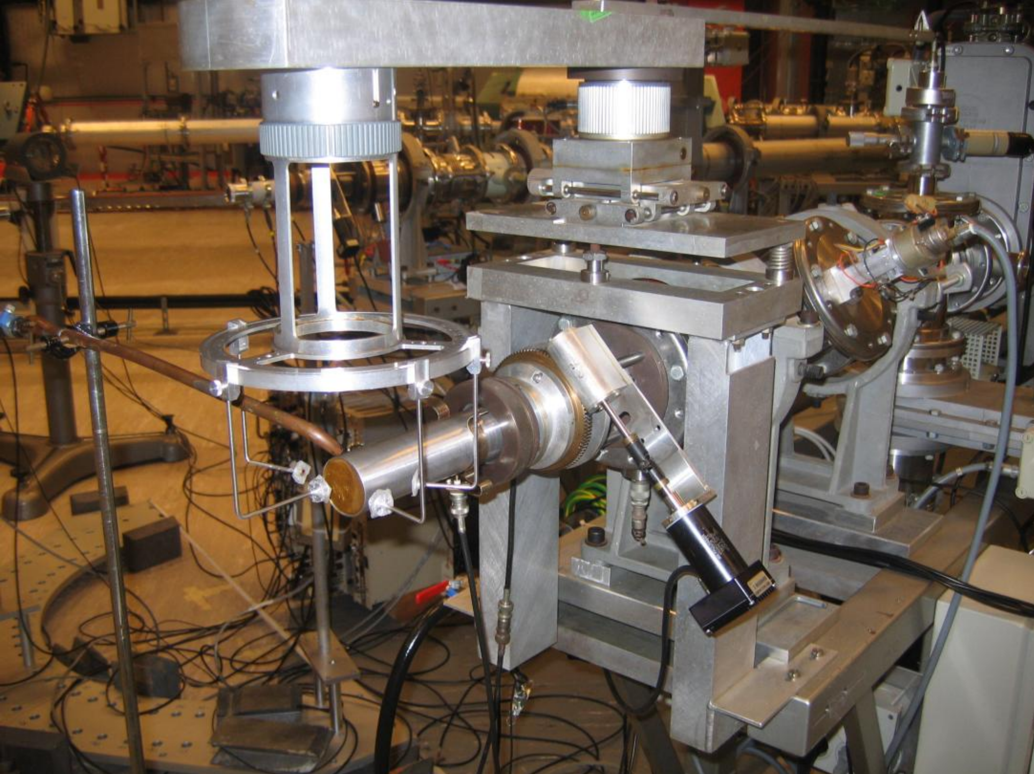


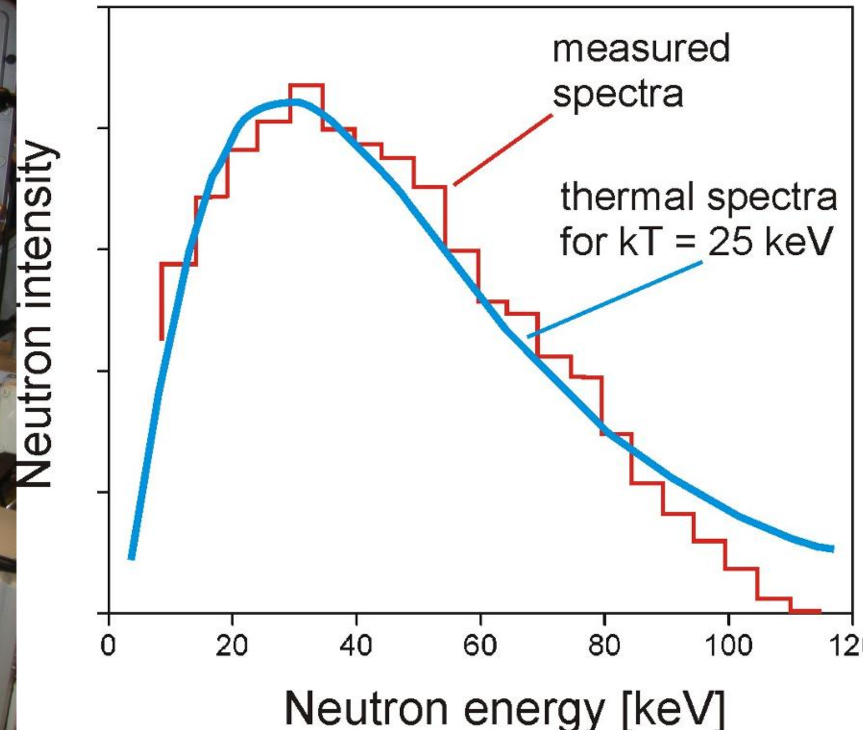
0.3 – 0.8 MeV **1.5 – 10 MeV**

(n,3n) for ^{238}U and ^{232}Th





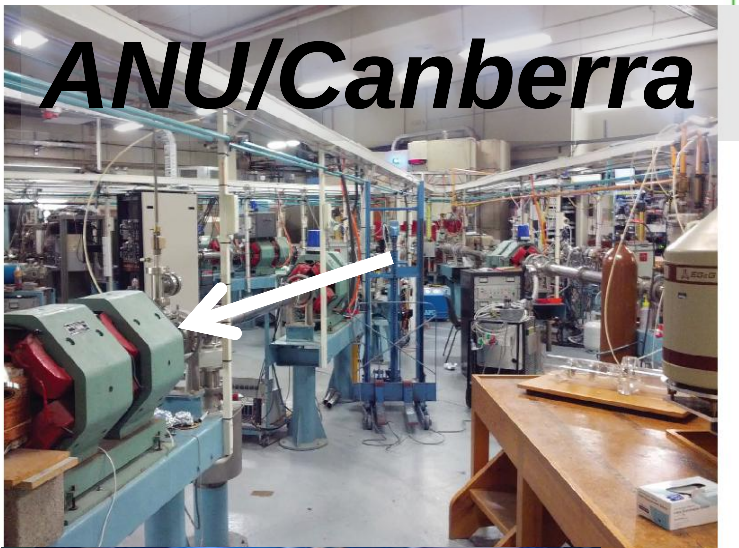


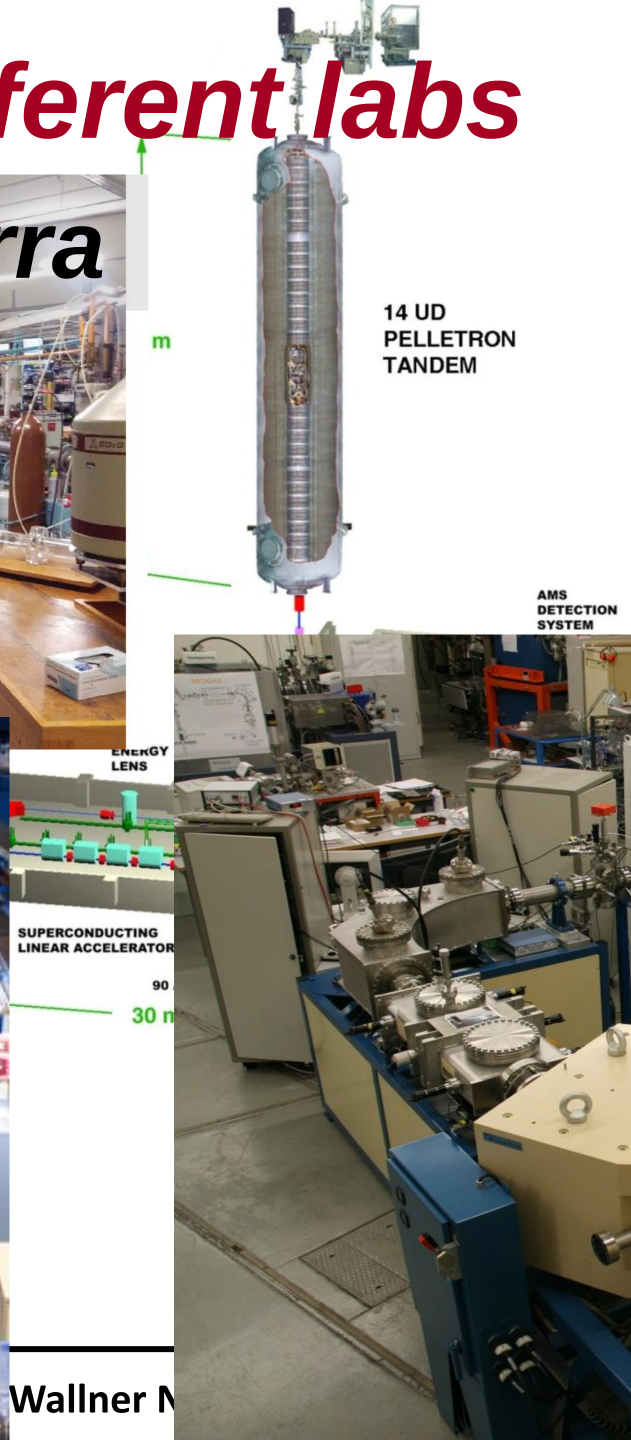


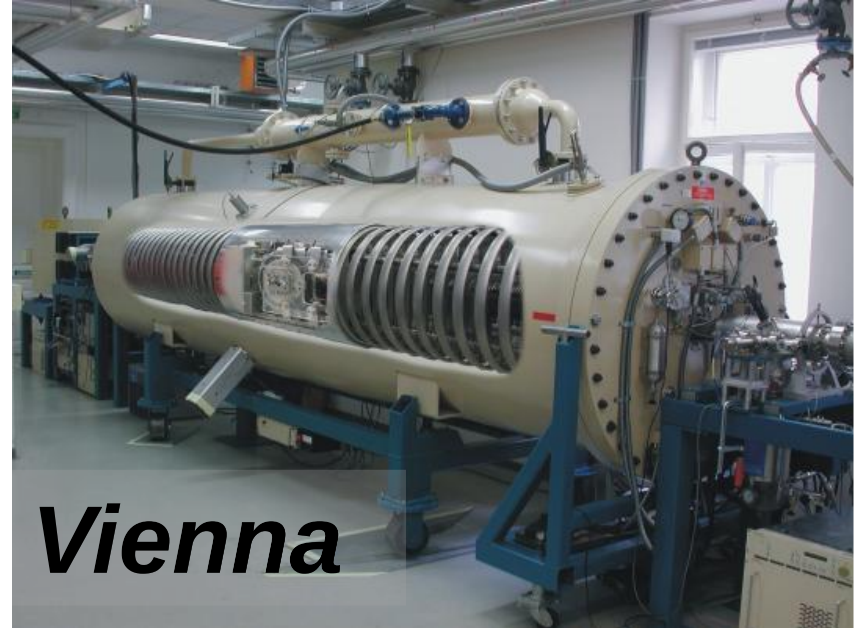
- products using the most sensitive
- technique: AMS

- **Atominstitut Vienna**: research reactor, thermal neutrons
- **IKI Budapest**: research reactor, thermal neutrons; cold neutrons
- **TSL Uppsala**: cyclotron, $^{7}Li(p,n)$: 45 MeV neutrons

AMS at different labs

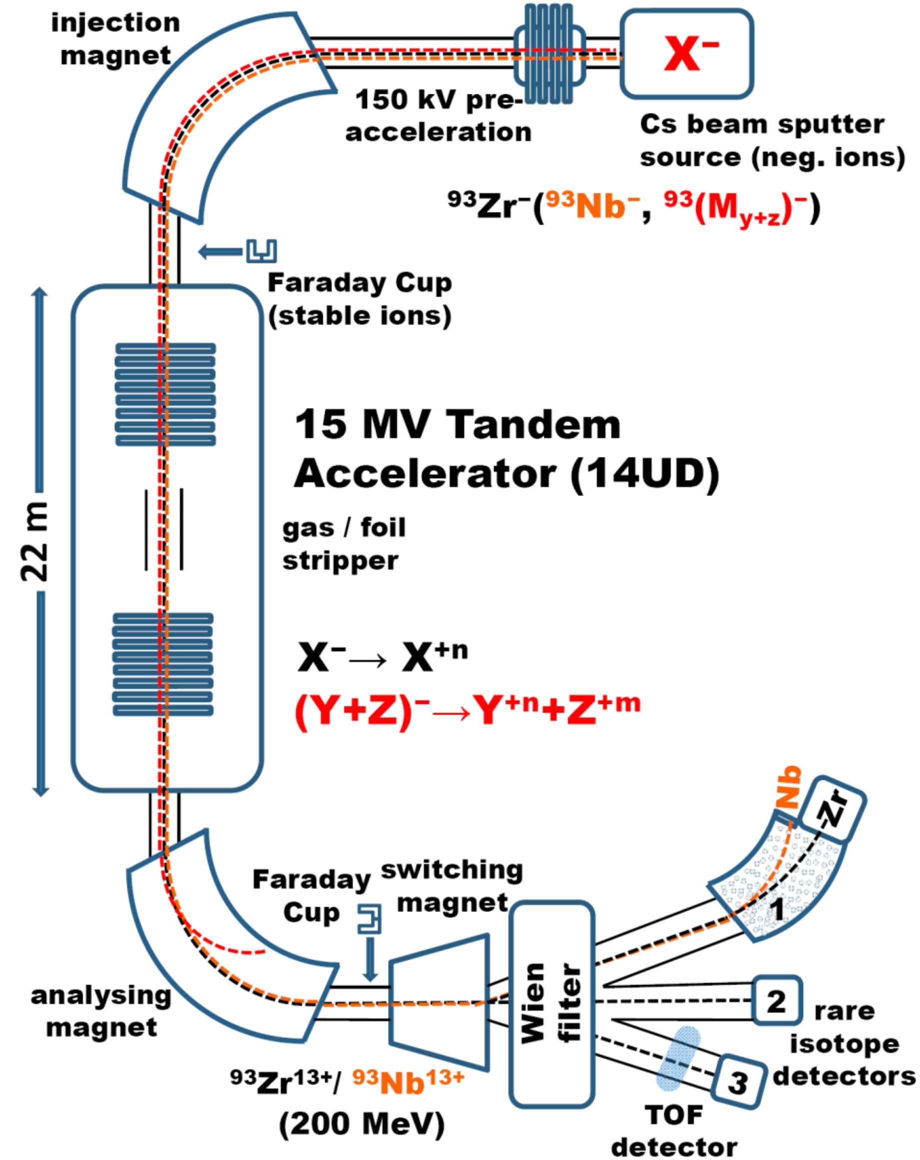






ANU's AMS facility

- AMS determines isotope ratios – atom counting technique
- $^{14}C/^{12}C$ – radiocarbon dating
- highest sensitivity: 10^{-12} – 10^{-16}
- no isobaric background (<-> ICPMS) (molecules are completely destroyed)
- isotopic background clearly identified



How to derive cross sections?

$$\sigma_{\exp} = \frac{N_{\text{product}}}{N_{\text{target}}} \times \frac{1}{\Phi}$$

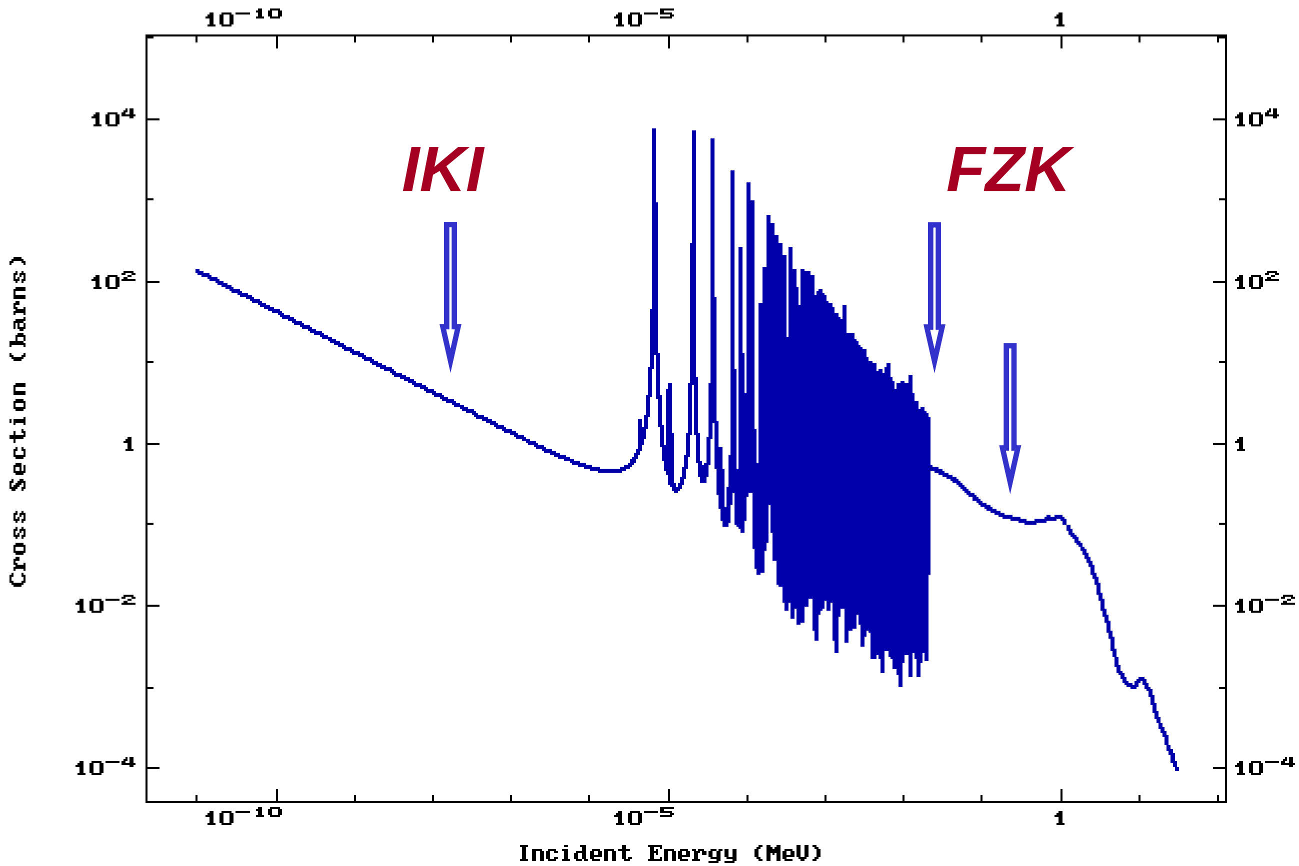
AMS-
isotope ratio

Monitor
(Au, Nb,…)

U

AMS monitor reaction (Au)

$^{238}U(n,\gamma)^{239}U$ -> ^{239}Np -> ^{239}Pu (24 kyr)



$^{235}U(n,\gamma)^{236}U$ (23 Myr) - simultaneously

^{236}U & ^{239}U: Production & Decay

^{236}U: $t_{1/2}$ = 23.4 Myr

^{239}U: $t_{1/2}$ = 23.5 min

^{239}Np: $t_{1/2}$ = 2.35 days

^{239}Pu: $t_{1/2}$ = 24.1 kyr

Analytical Strategy

Au for fluence → Sample ← ^{233}U and ^{242}Pu spike

↓

Chemical separation of U / Pu

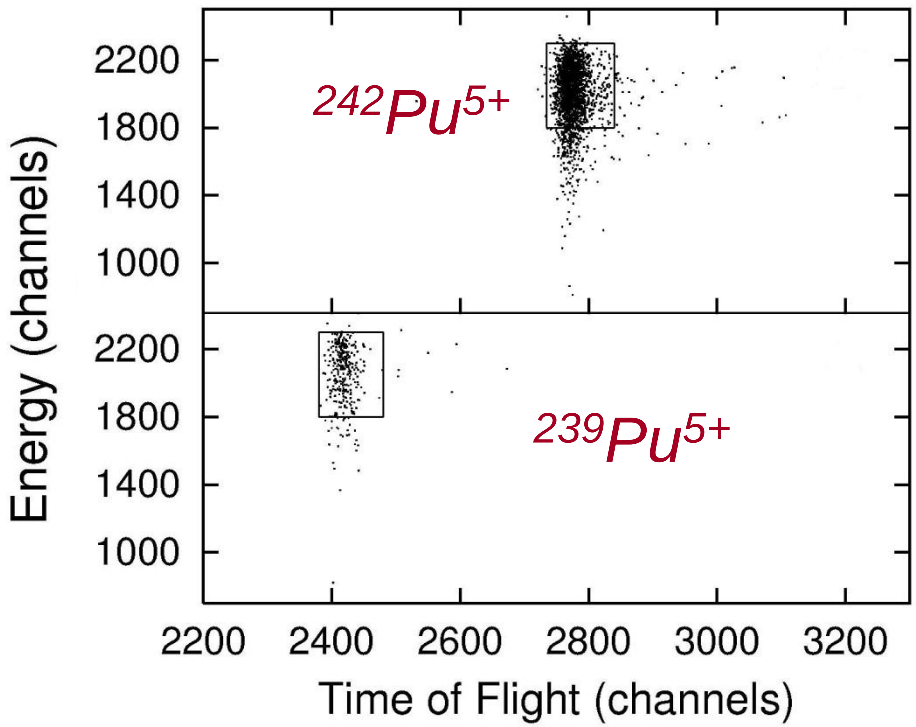
↓

AMS

↙ ↘

^{239}Pu / ^{242}Pu	^{233}U / ^{236}U / ^{238}U
↓	↓
$^{238}U(n,\gamma)^{239}U$	$^{235}U(n,\gamma)^{236}U$

typical AMS-spectrum for Pu measurements

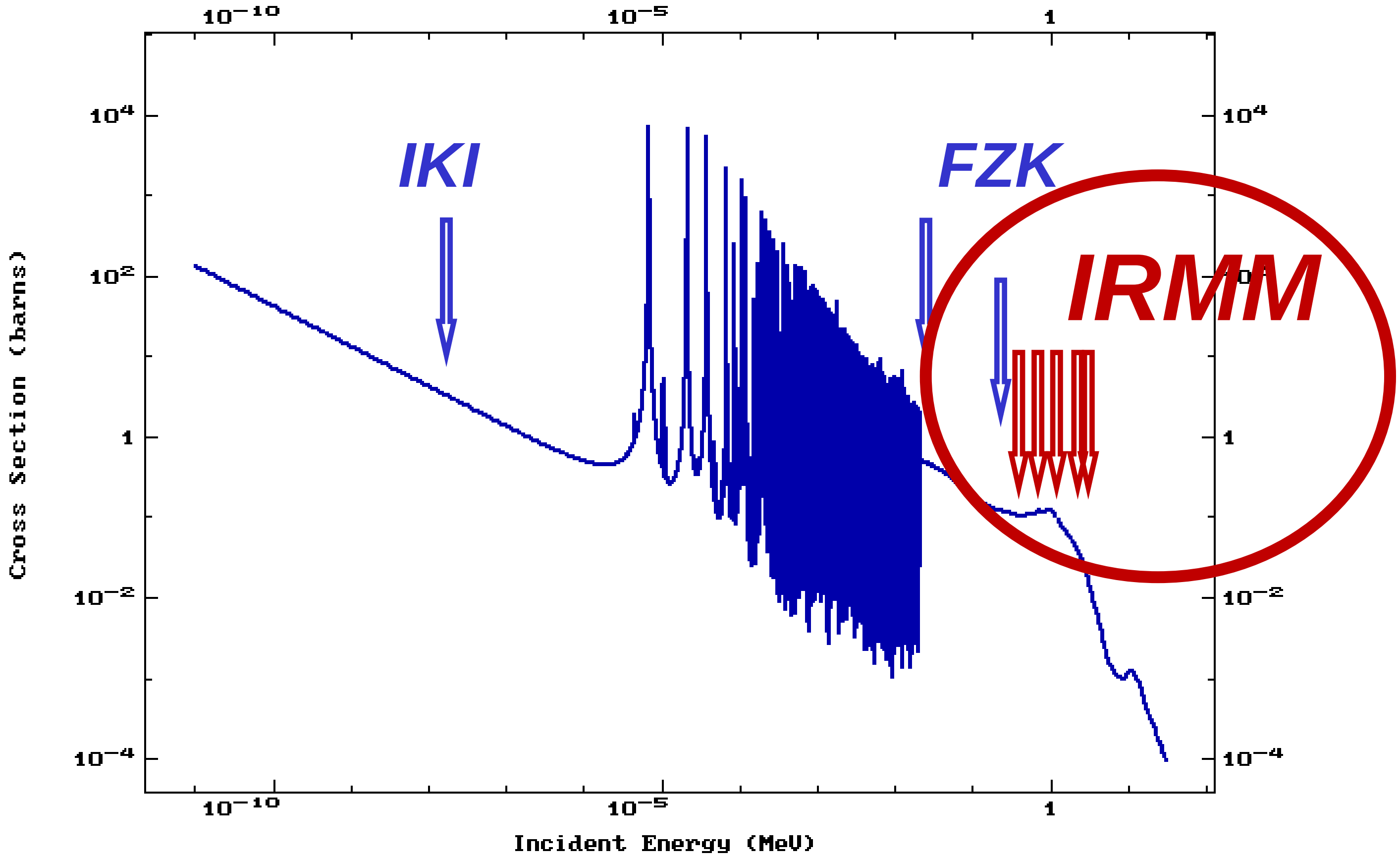


clear separation / no interference & ^{236}U-sensitivity: $^{236}U/^{238}U \sim 10^{-12}$

overall 10 AMS-beamtime sessions for $^{235}U(n,\gamma)$: +- 3 %

overall 6 AMS-beamtime sessions for $^{238}U(n,\gamma)$: +- 3 %

$^{238}U(n,\gamma)^{239}U \rightarrow {}^{239}Np \rightarrow {}^{239}Pu$ (24 kyr)



$^{232}Th(n,\gamma)^{233}Th \rightarrow {}^{233}Pa \rightarrow {}^{233}U$ (159 kyr)- simultaneously

^{233}Th & ^{239}U: Production & Decay: (n,γ)

low MeV

^{233}Th: $t_{1/2}$ = 22.3 min

^{233}Pa: $t_{1/2}$ = 27 days

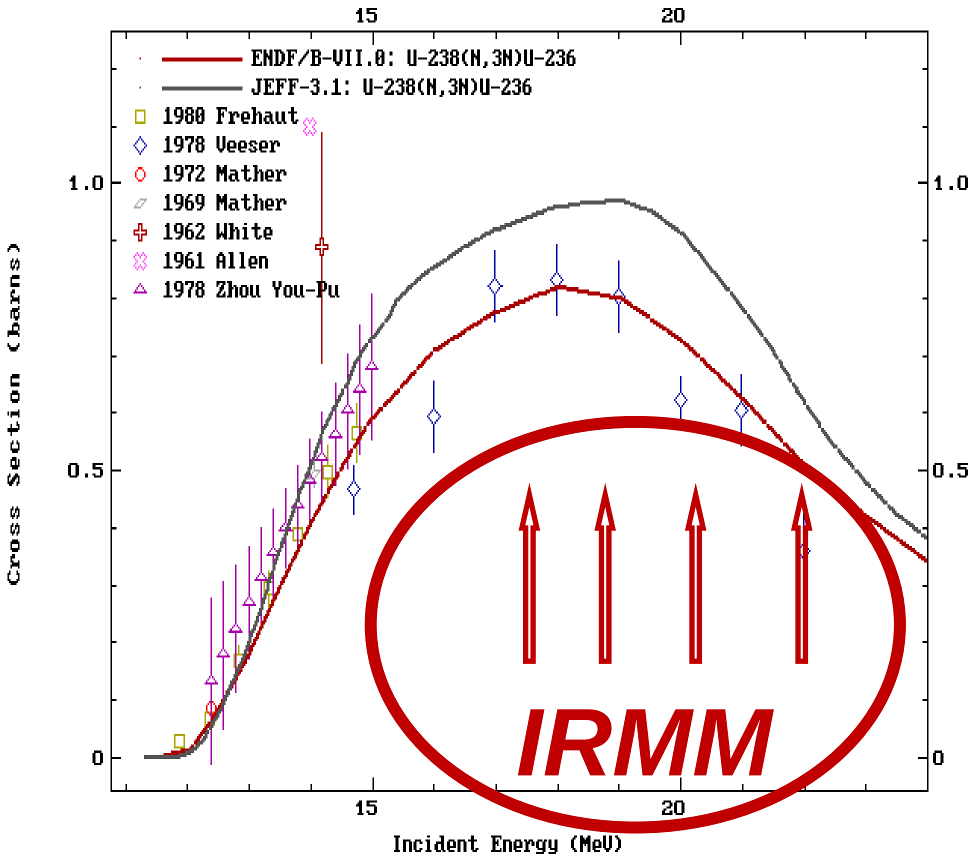
^{233}U: $t_{1/2}$ = 159 kyr

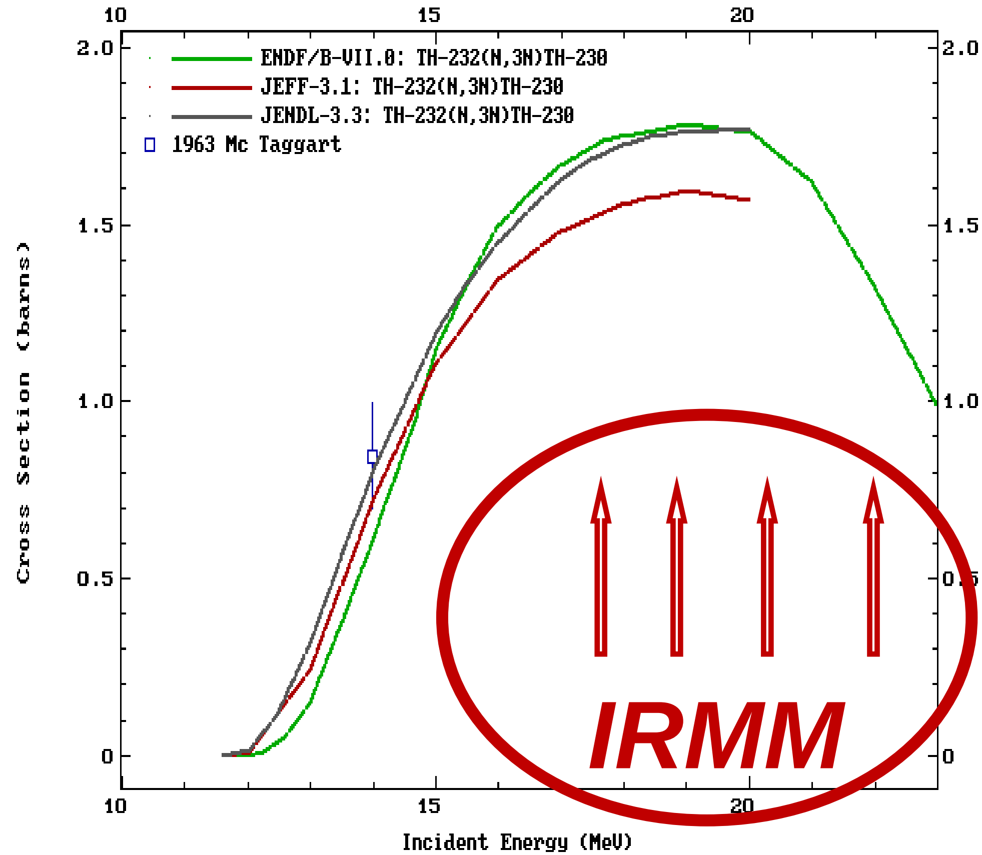
^{239}U: $t_{1/2}$ = 23.5 min

^{239}Np: $t_{1/2}$ = 2.35 days

^{239}Pu: $t_{1/2}$ = 24.1 kyr

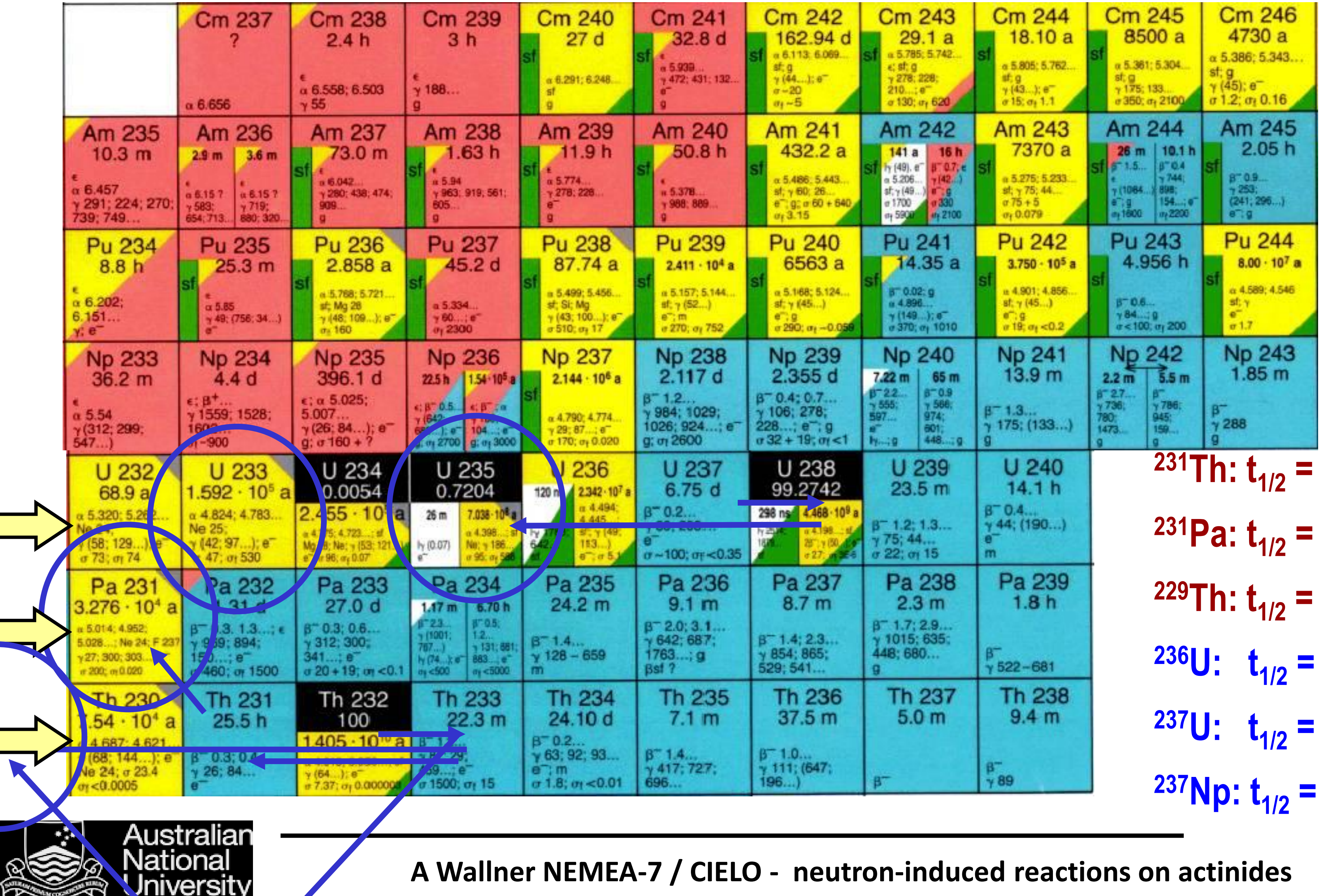
Fast neutron induced reactions: Th / U





U/Th simultaneously – the same geometry – the same fluence – (n,3n), (n,4n), (n,2n), (n,α) in the same irradiation

^{232}Th & ^{238}U: Production & Decay: (n,x)



fast n

^{231}Th: $t_{1/2}$ = 25.5 h

^{231}Pa: $t_{1/2}$ = 32.8 kyr

^{229}Th: $t_{1/2}$ = 7.88 kyr

^{236}U: $t_{1/2}$ = 23.4 Myr

^{237}U: $t_{1/2}$ = 6.75 days

^{237}Np: $t_{1/2}$ = 2.14 Myr

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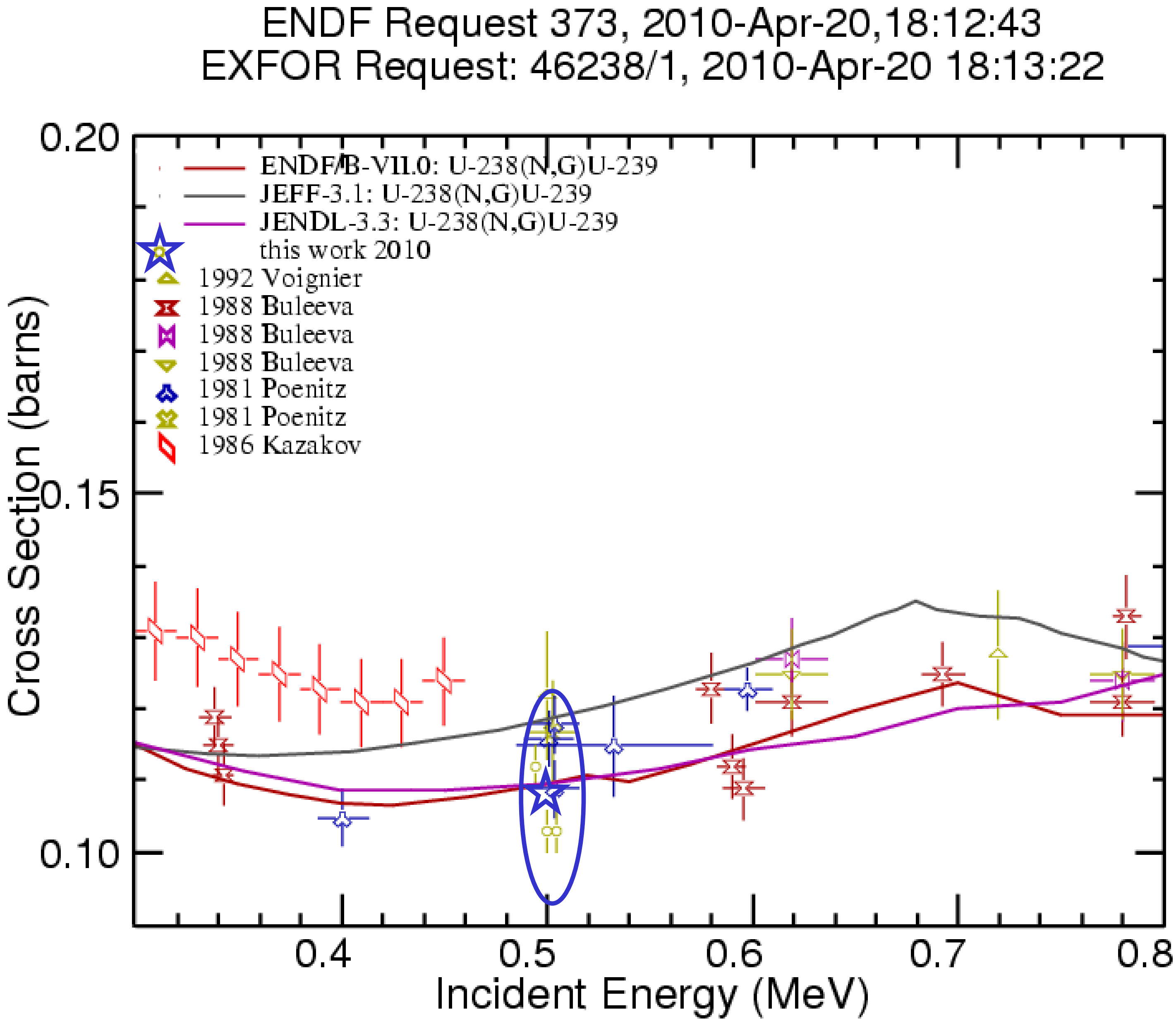


new data

$^{238}U(n,\gamma)^{239}U$

EXFOR database

300 – 800 keV

exp. data > 1980

Status – keV data

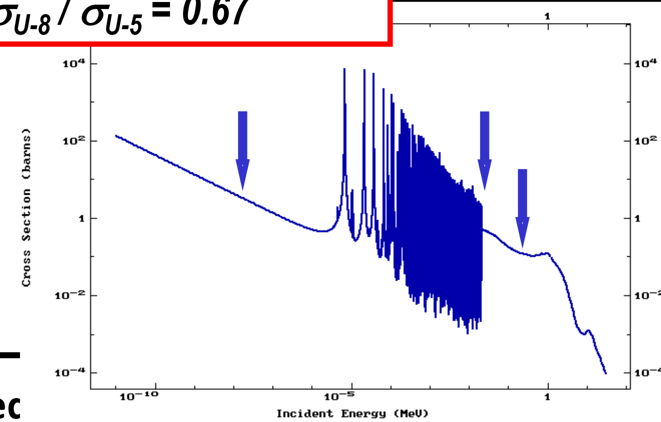
	$^{235}U(n,\gamma)^{236}U$ this work	$^{235}U(n,\gamma)^{236}U$ „recommended“	$^{238}U(n,\gamma)^{239}U$ this work	$^{238}U(n,\gamma)^{239}U$ „recommended“
$\sigma_{thermal}$	IKI-1: 99.5 ± 3 barn* IKI-2: 101.5 ± 3 barn*	98.96 barn 98.96 barn	IKI-1: reference for AMS IKI-2: reference for AMS	2.68 barn (2.72 barn JENDL)
$\sigma_{25\ keV}$	KIT-1: 646 ± 38 mbarn	ENDF: 679 mbarn (ENDF = JEFF)	KIT-1: 386 ± 25 mbarn	401 mbarn (ENDF,JEFF)
$\sigma_{426\ keV}$	KIT-2: 161 ± 7 mbarn	163 mbarn (ENDF,JEFF) 171 mbarn (JENDL)	KIT-2: 108 ± 4 mbarn	109 mbarn (ENDF, JENDL) 119 mbarn (JEFF)

this work (AMS ratio): $\sigma_{U\text{-}8} / \sigma_{U\text{-}5} = 0.60 \pm 0.05$
ENDF: $\sigma_{U\text{-}8} / \sigma_{U\text{-}5} = 0.59$

this work (AMS ratio): $\sigma_{U\text{-}8} / \sigma_{U\text{-}5} = 0.67 \pm 0.04$
ENDF: $\sigma_{U\text{-}8} / \sigma_{U\text{-}5} = 0.67$

- *relative to ^{233}U-spike*

AMS vs ENDF: $\sigma_{U\text{-}5}$ (25 keV) = 0.95 ± 0.06
AMS vs ENDF: $\sigma_{U\text{-}8}$ (25 keV) = 0.96 ± 0.06
AMS vs ENDF: $\sigma_{U\text{-}5}$ (426 keV) = 0.99 ± 0.04
AMS vs ENDF: $\sigma_{U\text{-}8}$ (426 keV) = 0.99 ± 0.04



PRL 109, 202506 (2012) PHYSICAL REVIEW LETTERS week ending 16 NOVEMBER 2012

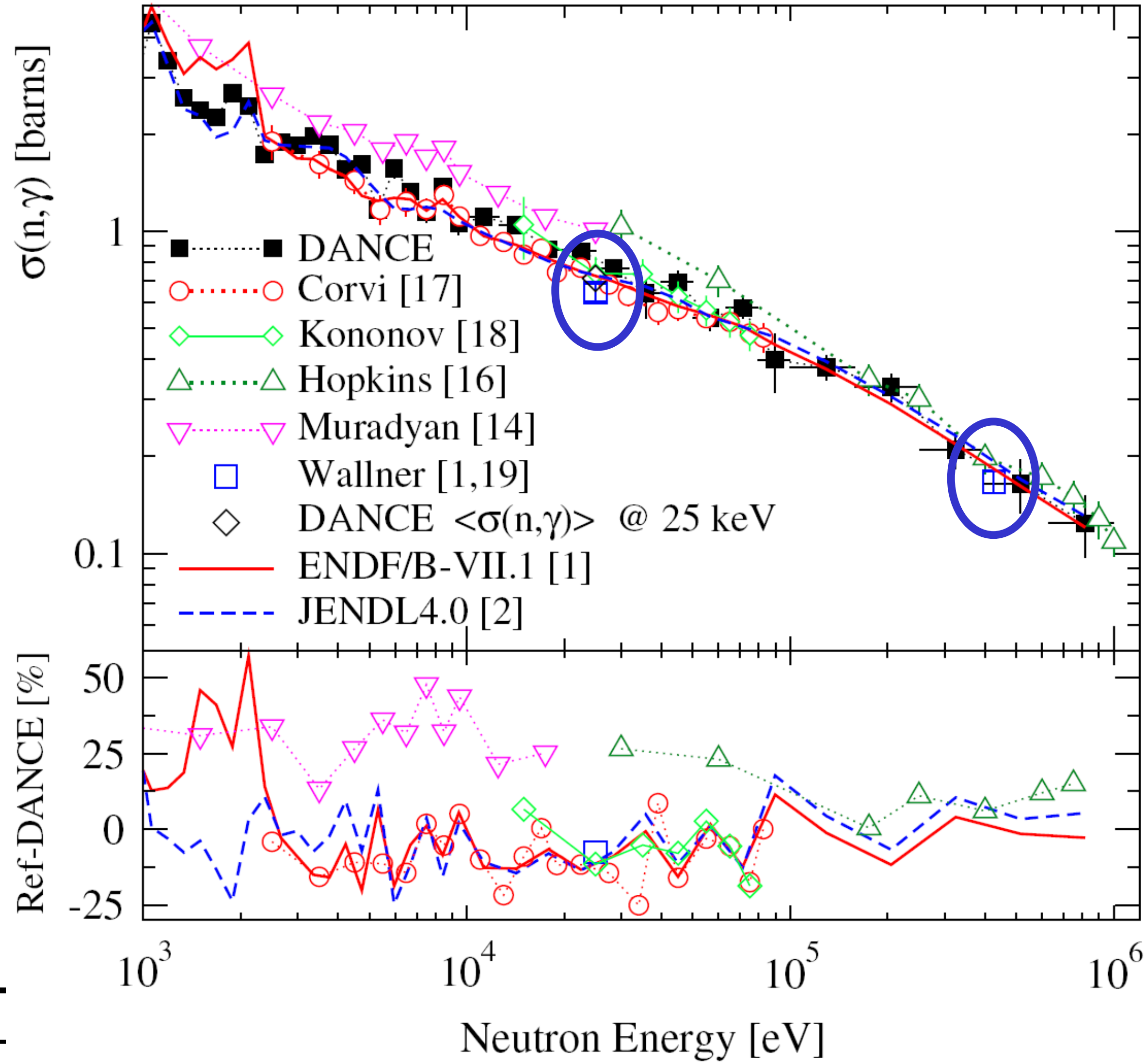
New Precision Measurements of the $^{235}U(n, \gamma)$ Cross Section

M. Jandel,[1,*] T. A. Bredeweg,[1] E. M. Bond,[1] M. B. Chadwick,[1] A. Couture,[1] J. M. O'Donnell,[1] M. Fowler,[1] R. C. Haight,[1] T. Kawano,[1] R. Reifarth,[1,†] R. S. Rundberg,[1] J. L. Ullmann,[1] D. J. Vieira,[1] J. M. Wouters,[1,‡] J. B. Wilhelmy,[1] C. Y. Wu,[2] and J. A. Becker[2]

25 keV – Maxwell-Boltzmann:

JANDEL - 2012:	σ_{U-5} = 0.70 ± 0.06
ENDF/B-VII.1	σ_{U-5} = 0.679
this work (AMS):	σ_{U-5} = 0.646 ± 0.038

AMS vs JANDEL: σ_{U-5} (25 keV) = 0.92 ± 0.10

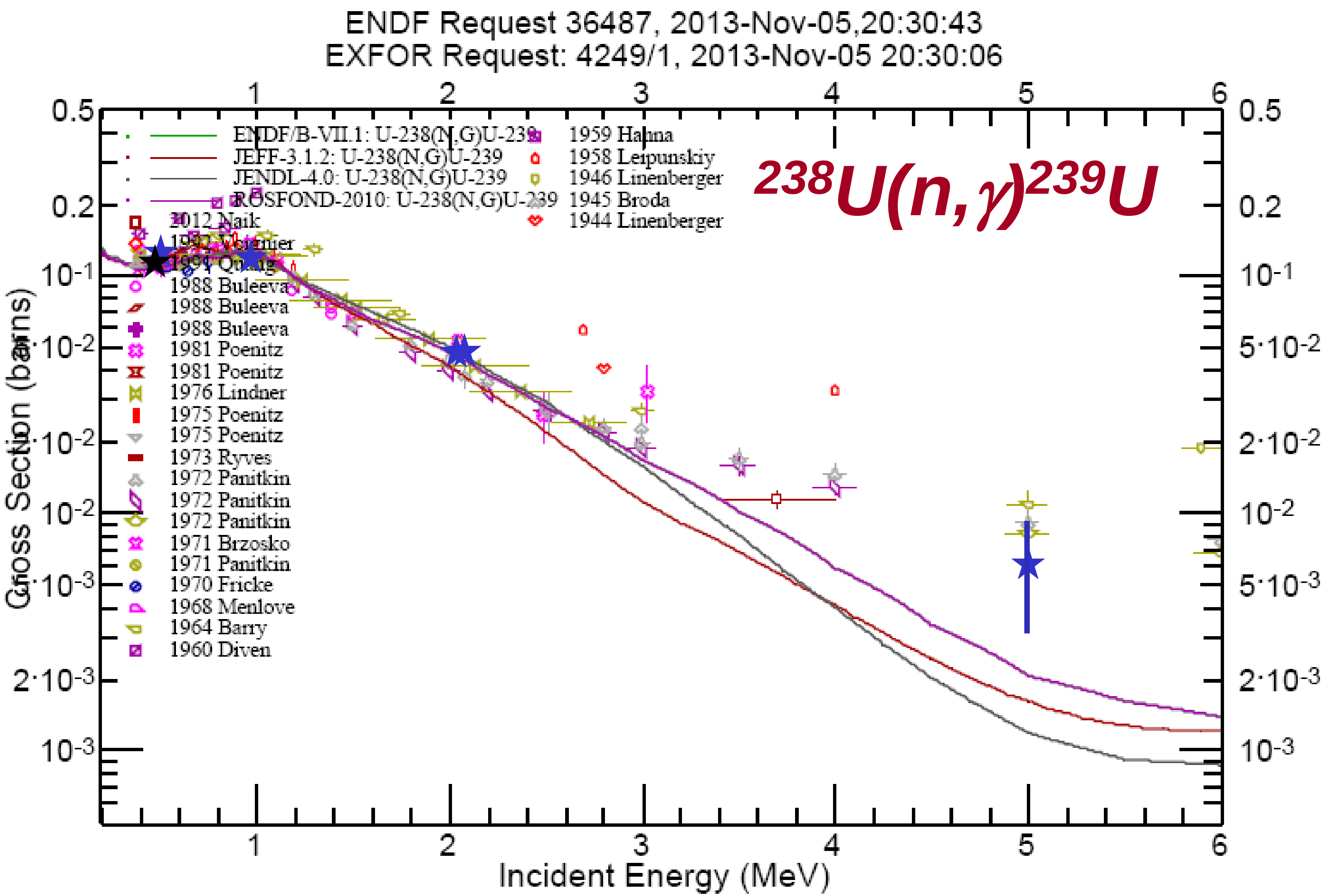


Status ^{232}Th(n,γ)^{233}Th & ^{238}U(n,γ)^{239}U for 0.5 / 1.0 / 2.1 / 5.0 MeV

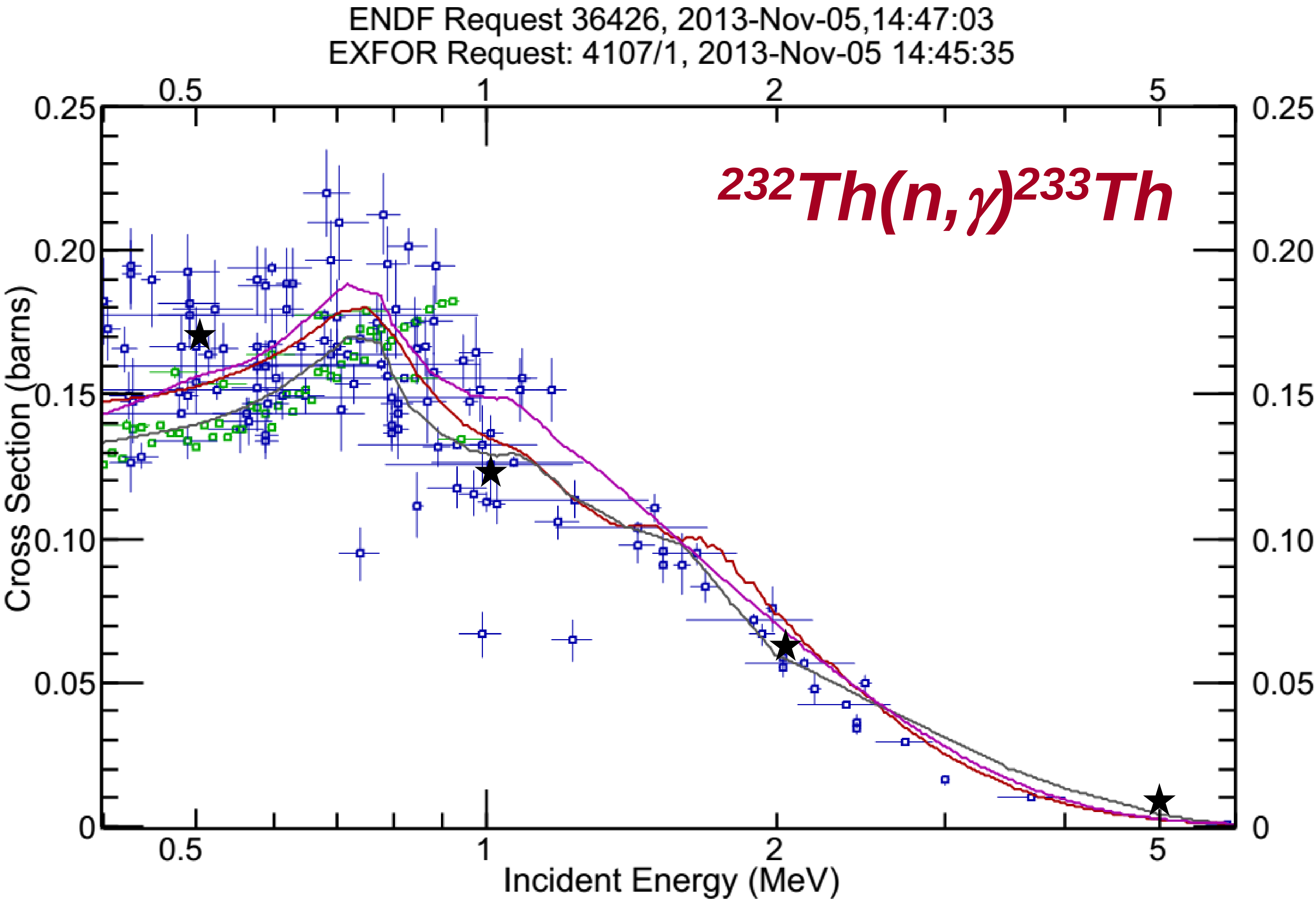
Preliminary data for ^{232}Th(n,γ) and ^{238}U(n,γ)

E_n (MeV)	Th-AMS (mbarn)	ENDF-Th (mbarn)	U-AMS (mbarn)	ENDF-U (mbarn)	AMS/ENDF - Th	AMS / ENDF - U
0.46	--		108 ± 4	109		1.00 ± 0.03
0.52	172 ± 15	153	123 ± 6	110	1.12 ± 0.10	1.12 ± 0.05
1.0	118 ± 10	136	108 ± 5	128	0.87 ± 0.07	0.85 ± 0.04
2.1	59 ± 3	68	42 ± 2	43	0.86 ± 0.03	0.97 ± 0.05
2.1		68	40 ± 7	43		0.92 ± 0.10
5.0	~ 10	2.8	6 ± 3	2.1	3.7	3 ± 2

AMS at the ANU



ENDF Request 36487, 2013-Nov-05,20:30:43
EXFOR Request: 4249/1, 2013-Nov-05 20:30:06
238U(n,γ)239U
ENDF/B-VII.1: U-238(N,G)U-239
JEFF-3.1.2: U-238(N,G)U-239
JENDL-4.0: U-238(N,G)U-239
ROSFOND-2010: U-238(N,G)U-239
1988 Buleeva
1988 Buleeva
1988 Buleeva
1981 Poenitz
1981 Poenitz
1976 Lindner
1975 Poenitz
1975 Poenitz
1973 Ryves
1972 Panitkin
1972 Panitkin
1972 Panitkin
1971 Brzosko
1971 Panitkin
1970 Fricke
1968 Menlove
1964 Barry
1960 Diven
1959 Hanna
1958 Leipunskiy
1946 Linenberger
1945 Broda
1944 Linenberger
Cross Section (barns)
Incident Energy (MeV)


ENDF Request 36426, 2013-Nov-05,14:47:03
EXFOR Request: 4107/1, 2013-Nov-05 14:45:35
232Th(n,γ)233Th
Cross Section (barns)
Incident Energy (MeV)
0.25
0.20
0.15
0.10
0.05
0
0.5
1
2
5

First results - $^{238}U(n,3n)^{236}U$

17.55 l

20.05 l

! Preliminary da

n“

n”

Preliminary data for $^{238}U(n,3n)^{236}U$ (VERA)

E_n (MeV)	AMS (barn)	evaluations (barn)
17.55	0.76 ± 0.04	0.7 – 1.0
18.80	0.89 ± 0.30	0.7 – 1.0
20.05	0.86 ± 0.04	0.6 – 0.9
22.00	0.42 ± 0.10	0.45 – 0.65

Fluence! ↔ number of neutrons/sample

First results - $^{232}Th(n,2n)^{231}Th$

VERY preliminary data for $^{232}Th(n,2n)^{231}Th$

(via ^{231}Pa: VERA & ETH & ANSTO)

E_n (MeV)	AMS-VERA (ratio)	AMS-ETH (ratio)	AMS-ANSTO (ratio)	AMS (barn)	evaluations (barn)
17.55	1.6 ± 0.3	1.5 ± 0.2	1.65 ± 0.3	0.20 ± 0.05	0.3 – 0.5
18.80	2.9 ± 0.3	3.0 ± 0.2	2.9 ± 0.3		0.25 – 0.45
20.05	29 ± 2	32 ± 1.2	24 ± 4	0.18 ± 0.01	0.2 – 0.4
22.00	2.2 ± 0.2	2.0 ± 0.2	2.2 ± 0.2		~ 0.2

First results - $^{232}Th(n,4n+\alpha)^{229}Th$

Preliminary data for $^{232}Th(n,4n+\alpha)$ (VERA & ETH)

E_n (MeV)	AMS (mbarn)	evaluations (mbarn)
17.55	< 5	0.2 - 2
20.05	6 ± 2	10 - 50
22.0	345 ± 35	~200 (±×3)

AMS & neutron fluence

^{236}U

$$\frac{\sigma_{U-5(n,\gamma)}}{\sigma_{U-8(n,\gamma)}} = \frac{\dfrac{^{236}U}{^{235}U}}{\dfrac{^{239}Pu}{^{238}U}}$$

AMS-ratio only !!!

this work (AMS ratio):	$\sigma_{U\text{-}8} / \sigma_{U\text{-}5}$ = *0.60* ± 0.05
ENDF:	$\sigma_{U\text{-}8} / \sigma_{U\text{-}5}$ = *0.59*
this work (AMS ratio):	$\sigma_{U\text{-}8} / \sigma_{U\text{-}5}$ = *0.67* ± 0.04
ENDF:	$\sigma_{U\text{-}8} / \sigma_{U\text{-}5}$ = *0.67*

systematic study of Maxwellian-averaged xs from precise AMS measurements

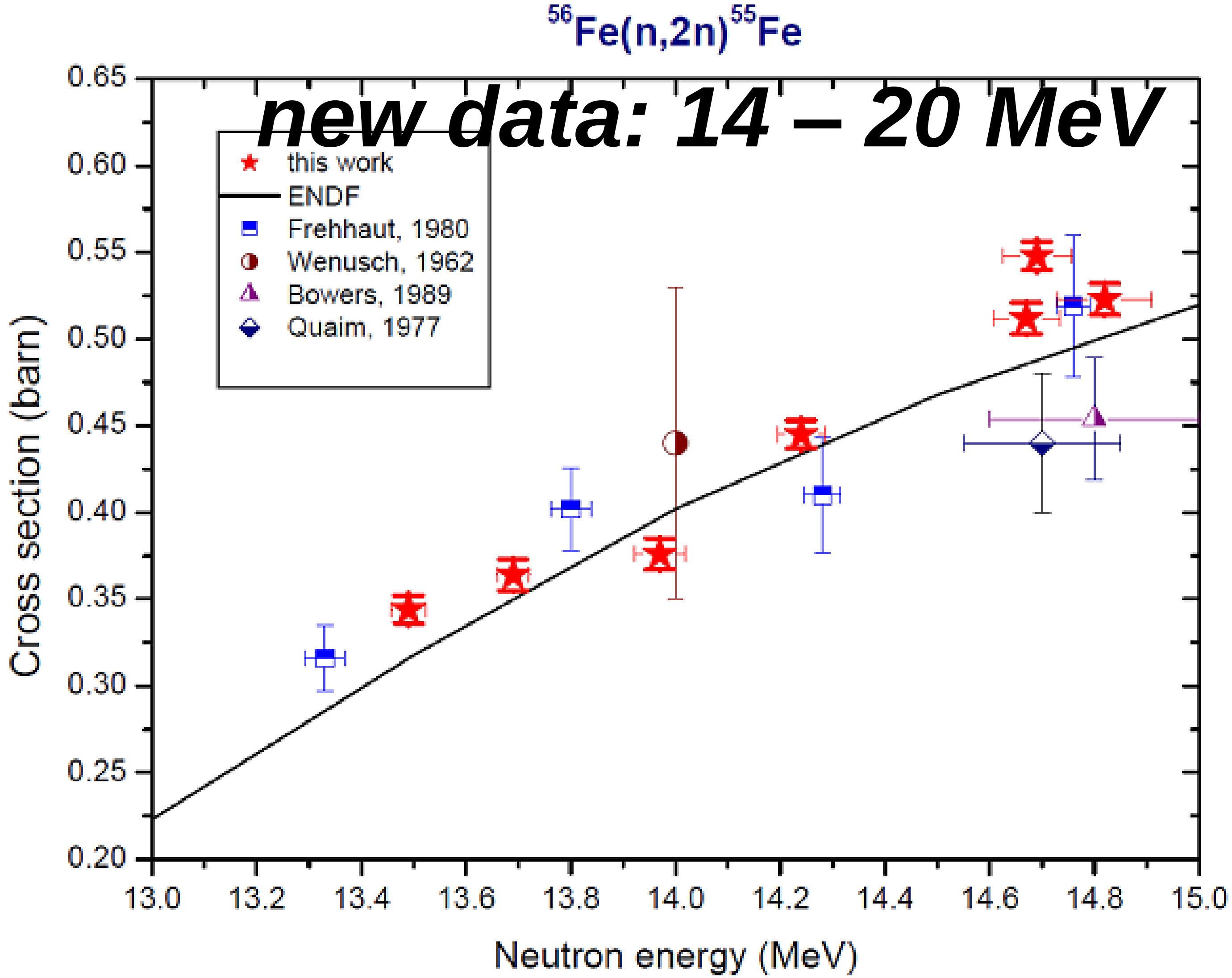
addendum:

Fe – AMS da

new data: 14 – 20 MeV

long-lived reaction products:

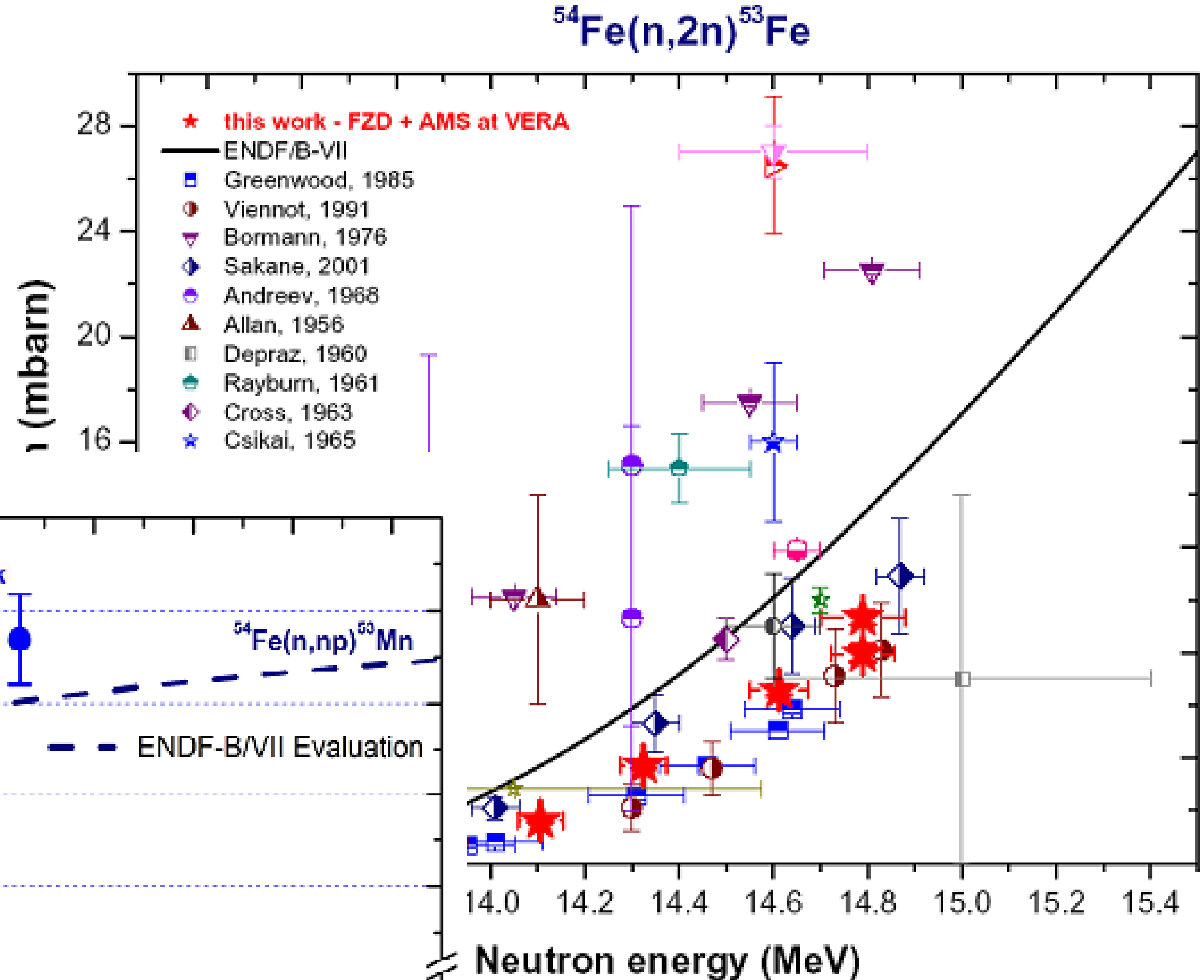
- $^{56}Fe(n,2n)^{55}Fe$
- $^{54}Fe(n,2n)^{53}Fe$ [$\rightarrow$ ^{53}M
- $^{54}Fe(n,\gamma)^{55}Fe$
- $^{54}Fe(n,np+d)^{53}Fe \rightarrow$ ^{53}M
- $^{58}Ni(n,\alpha)^{55}Fe$

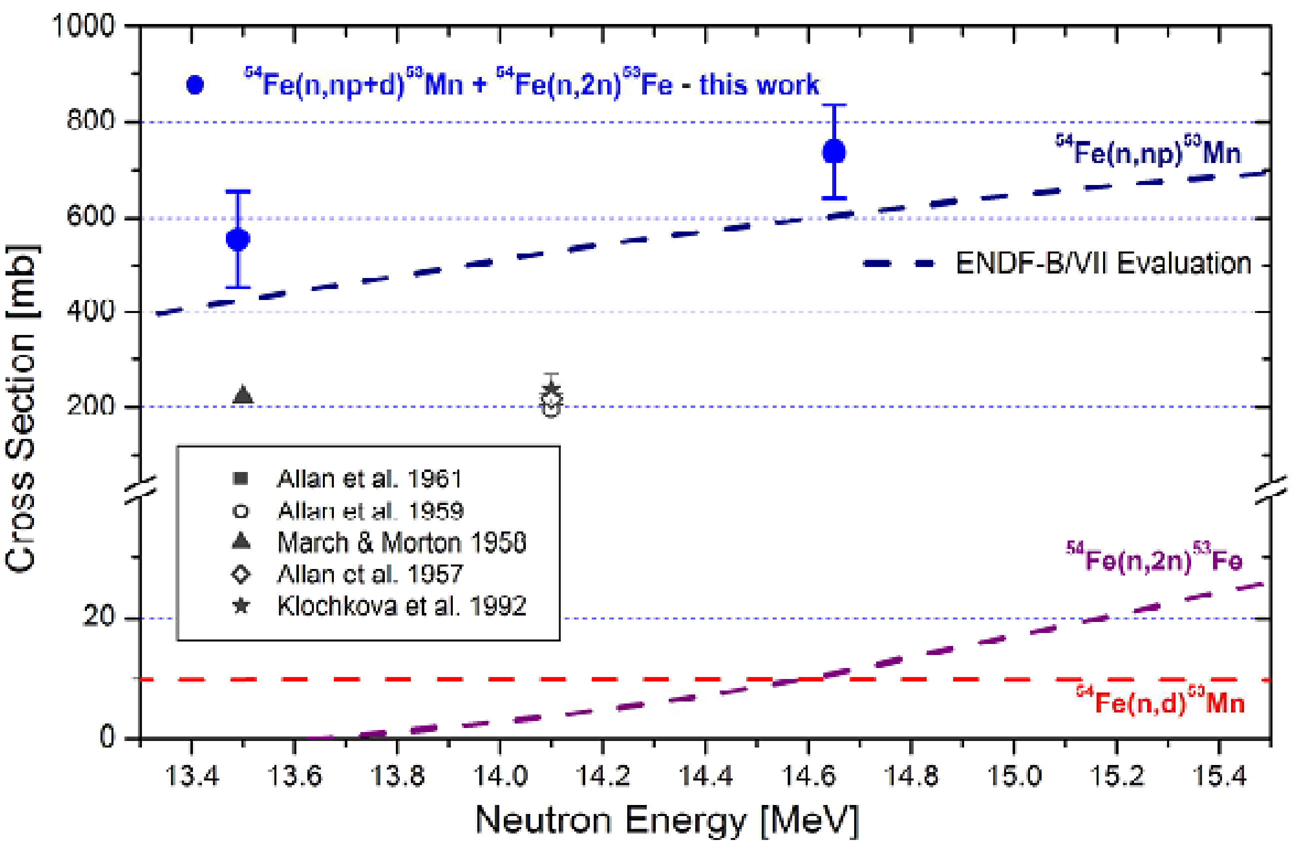


^{55}Fe - AMS: ± 2%

^{53}Fe / ^{53}Mn

^{53}Mn: $t_{1/2}$ = 3.7 My





Conclusion - actinides

- AMS represents an independent method for studying nuclear reactions leading to long-lived radionuclides
- direct method: atom counting as isotope ratio; i.e. no sophisticated data reduction or corrections required
- not a universal method – AMS is used for some 25 radionuclides
- if it can be applied – usually very (the most) sensitive method
- can serve as an important tool to provide some anchor points
- can it help to solve the Au-standard issue?
- new neutron-producing facilities (FRANZ, Frankfurt, Germany), SARAF (Israel) offer a much higher keV-neutron flux

Comparison AMS – other meas. / ENDF

Reaction		AMS (25keV)	other detection technique (KADONIS)	ENDF/B-VII.1/KADONIS
$^{9}Be(n,\gamma)^{10}Be$	EXP	9.5 μbarn	13 μbarn	0.88 ± 0.17
$^{13}C(n,\gamma)^{14}C$	EXP	13.3 μbarn	20 μbarn	-- (JENDL 13, JEFF 7μb)
$^{14}N(n,p)^{14}C$	EXP	1.74 mbarn	2 mbarn	-- (ENDF: 1.70 mbarn)
$^{35}Cl(n,\gamma)^{36}Cl$	EXP	8.84 mbarn	11.3 mbarn	0.78 ± 0.03
$^{40}Ca(n,\gamma)^{41}Ca$	MACS	5.74 mbarn	6.7 mbarn	0.89 ± 0.07
$^{54}Fe(n,\gamma)^{55}Fe$	EXP	mbarn	29.6 mbarn	0.73 ± 0.06
$^{235}U(n,\gamma)^{236}U$	EXP	646 ± 38 mbarn	--	-- (ENDF: 679 mbarn)
$^{238}U(n,\gamma)^{239}U$	EXP	386 ± 25 mbarn	--	-- (ENDF: 401 mbarn)

AMS is generally lower than ‚other' techniques, but

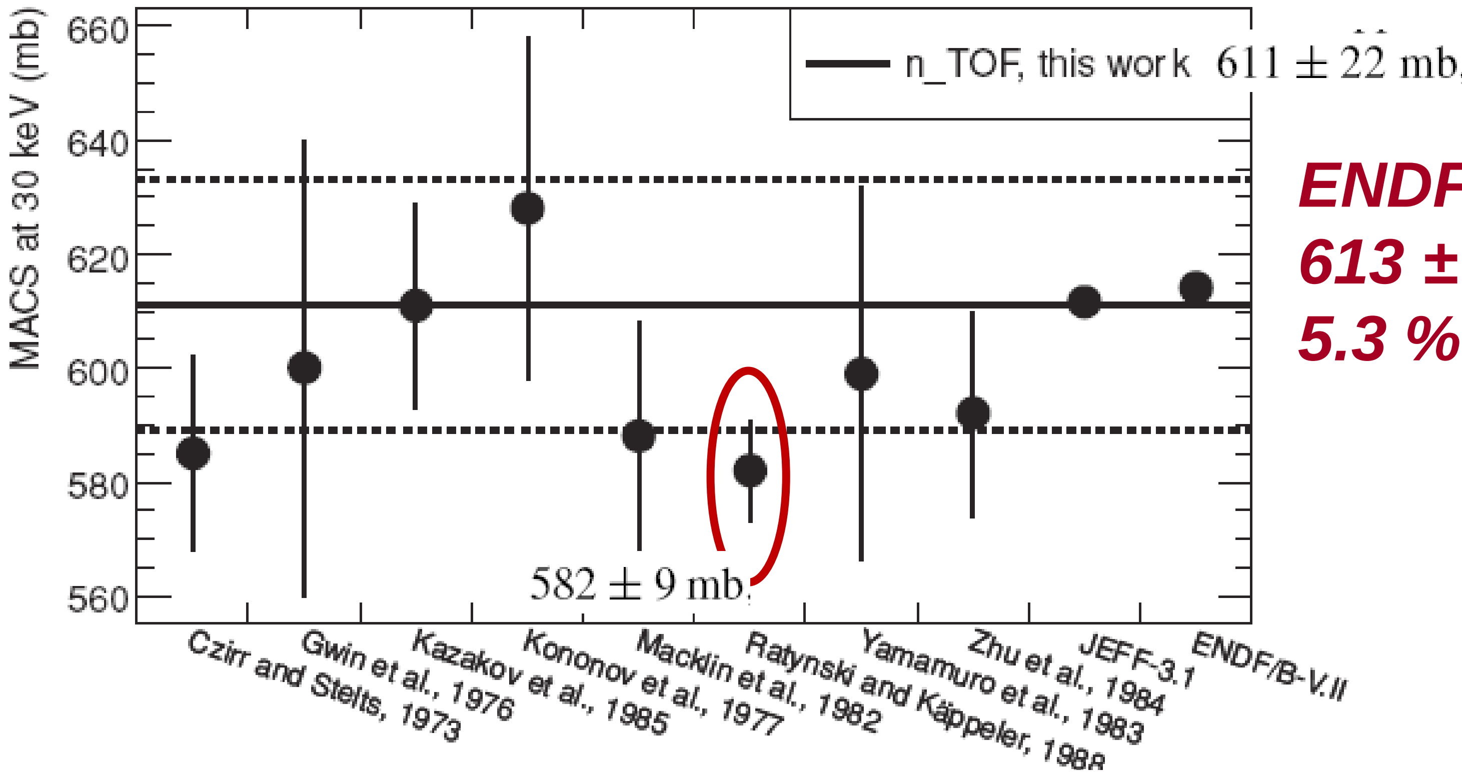
- fluence value via $^{197}Au(n,\gamma) \rightarrow$ Au cross section enters directly! (see above)
- background in other methods ?
- AMS: individual reactions via independent reference materials $\rightarrow$ no correlation in AMS

AMS data (not U) relative to Au- Ratynski-Käppeler for 25 keV

all activations with the same geometry at KIT – as used in "Ratynski & Käppeler

30 keV MACS for ^{197}Au(n,γ)

(Maxwellian-averaged cross sections)



ENDF/B-VII.1
613 ± 8 mb
5.3 %

PHYSICAL REVIEW C **83**, 034608 (2011) C. LEDERER *et al.*

^{197}Au(n,γ) cross section in the unresolved resonance region