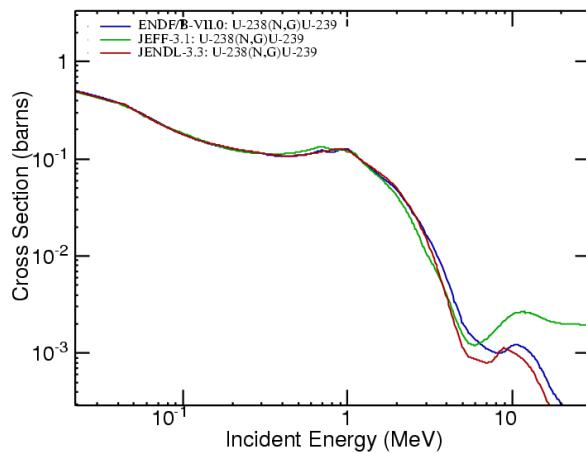
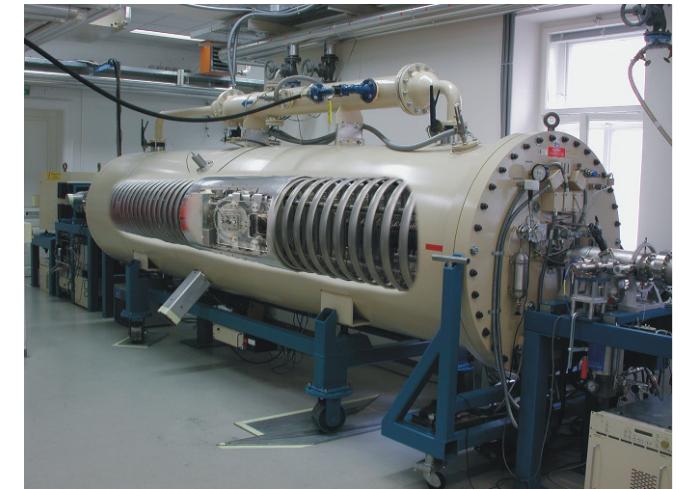
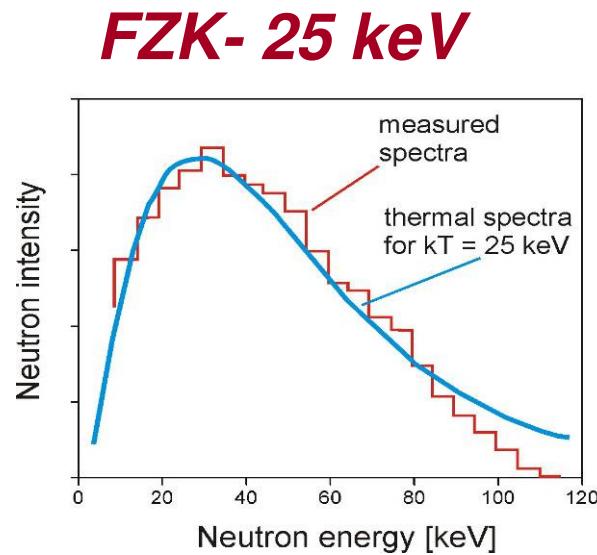


Neutron Capture Studies of ^{235}U and ^{238}U via AMS



$^{238}\text{U}(n,\gamma)^{239}\text{U} - \text{eval.}$



VERA

A. Wallner

VERA, Faculty of Physics, University of Vienna, Austria

Content

- *Method: combination of activation and AMS*
- *neutron capture cross section of $^{235,238}\text{U}$ in the keV region – a prime example for applying AMS*
- *AMS at VERA – overview*
- *first results*
- *other nuclides for neutron reaction data via AMS - outlook*

Activation (I) → AMS-Measurement (II) fusion / fission / ADS products

(1) Recent neutron irradiations:

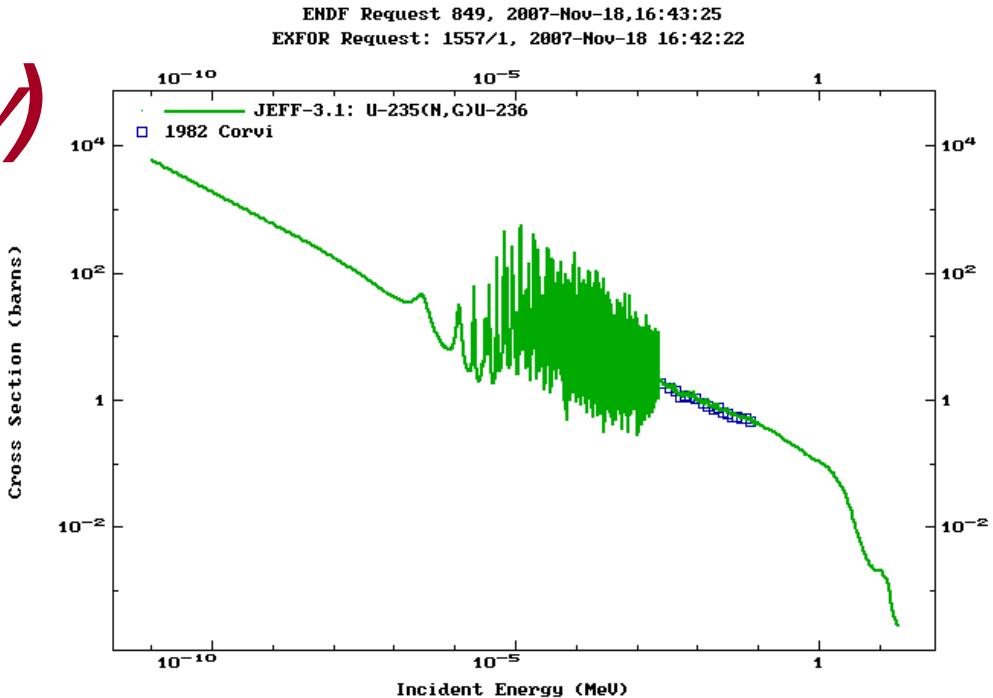
- **FZ Karlsruhe:** 3.7 MV Van de Graaff: $^7\text{Li}(\text{p},\text{n})^7\text{Be}$: 25 keV Maxwell-Boltzmann, 120 keV, 180 keV, 500 keV, ...)
- **IRMM Geel:** 7 MV Van de Graaff: ($\text{T}(\text{d},\text{n})^4\text{He}$: $E_n = 14 - 20 \text{ MeV}$)
- **FZ Dresden/Rossendorf:** 300 kV ($\text{T}(\text{d},\text{n})^4\text{He}$: $E_n = 13.3 - 14.9 \text{ MeV}$)
- **Atominstitut Vienna:** research reactor, thermal neutrons
- **IKI Budapest:** research reactor, thermal neutrons; cold neutrons

(2) AMS measurements e.g. at VERA: from Be to ^{238}U , Pu

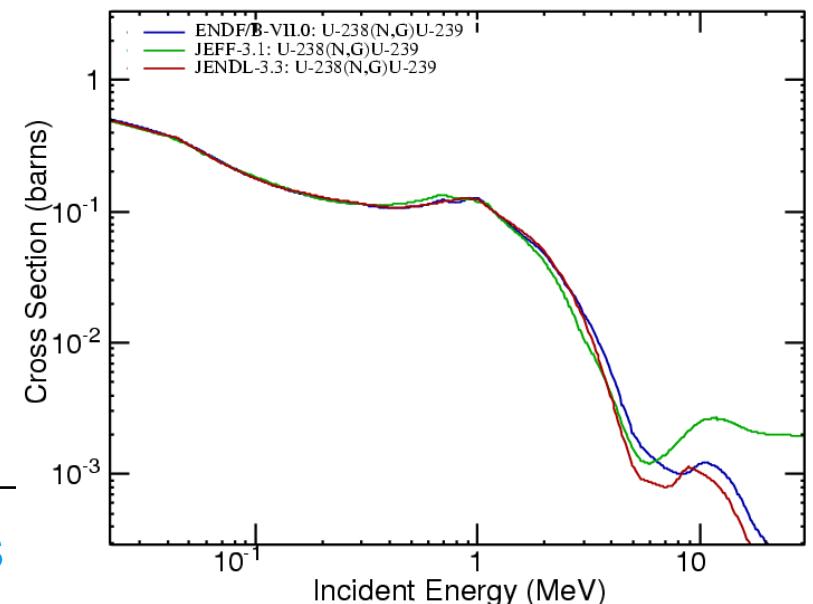
Motivation: $^{235}/^8U(n,\gamma)$

- ratio of capture to fission of the fissile isotopes: capture on ^{235}U
- ^{235}U : existing data via TOF and detections of prompt γ -rays;
- ^{238}U also decay of ^{239}Np
- since 1980 only 1 measurement in the keV region for ^{235}U (1982 Corvi et al.)
- Activation + AMS is an independent method; no influence from fission channel
- may serve as proof-of-principle measurement - to be extended to other reactions

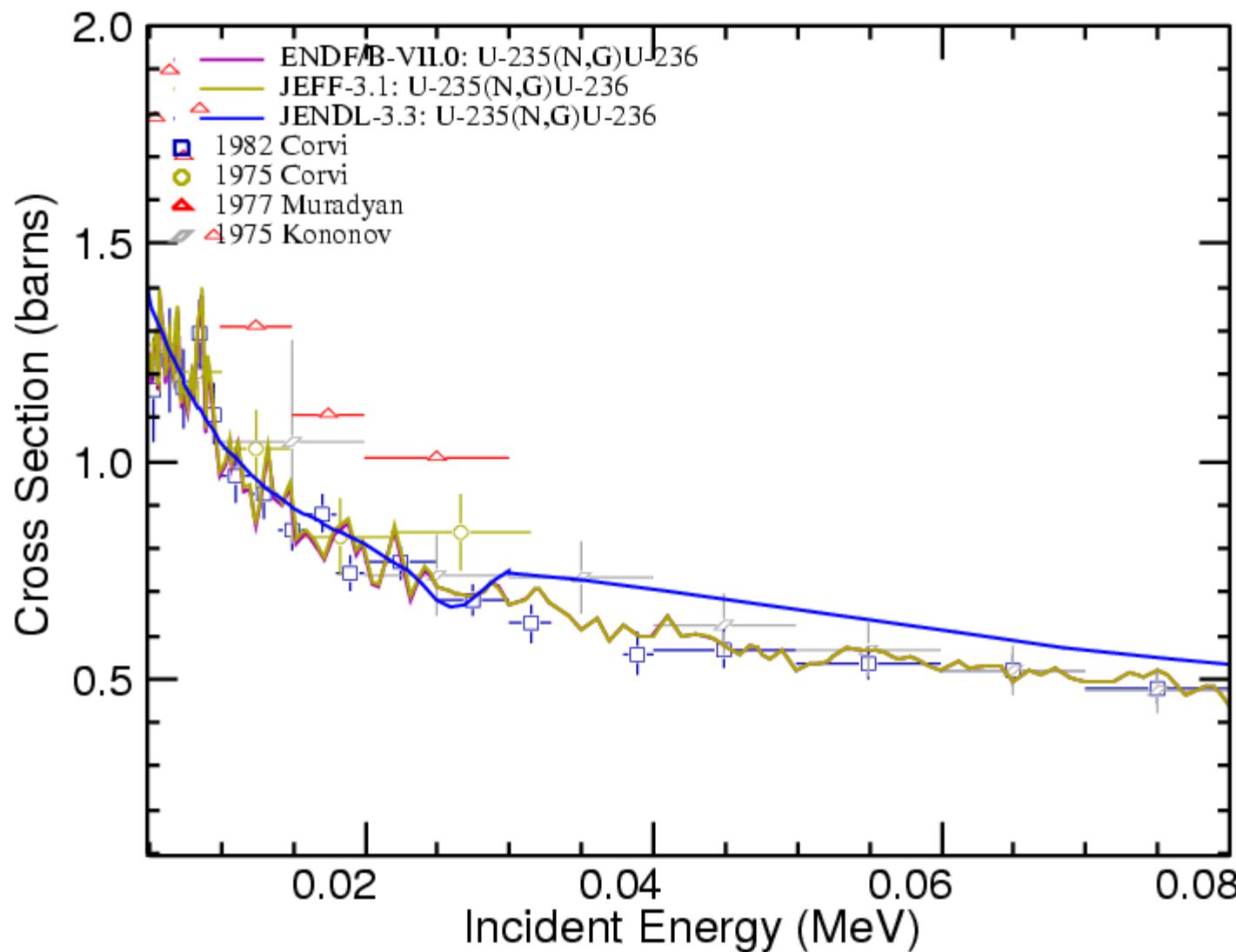
! discrepancy of evaluations !



ENDF Request 352, 2010-Apr-20, 16:00:00



ENDF Request 360, 2010-Apr-20, 17:26:55
EXFOR Request: 46218/1, 2010-Apr-20 16:38:59



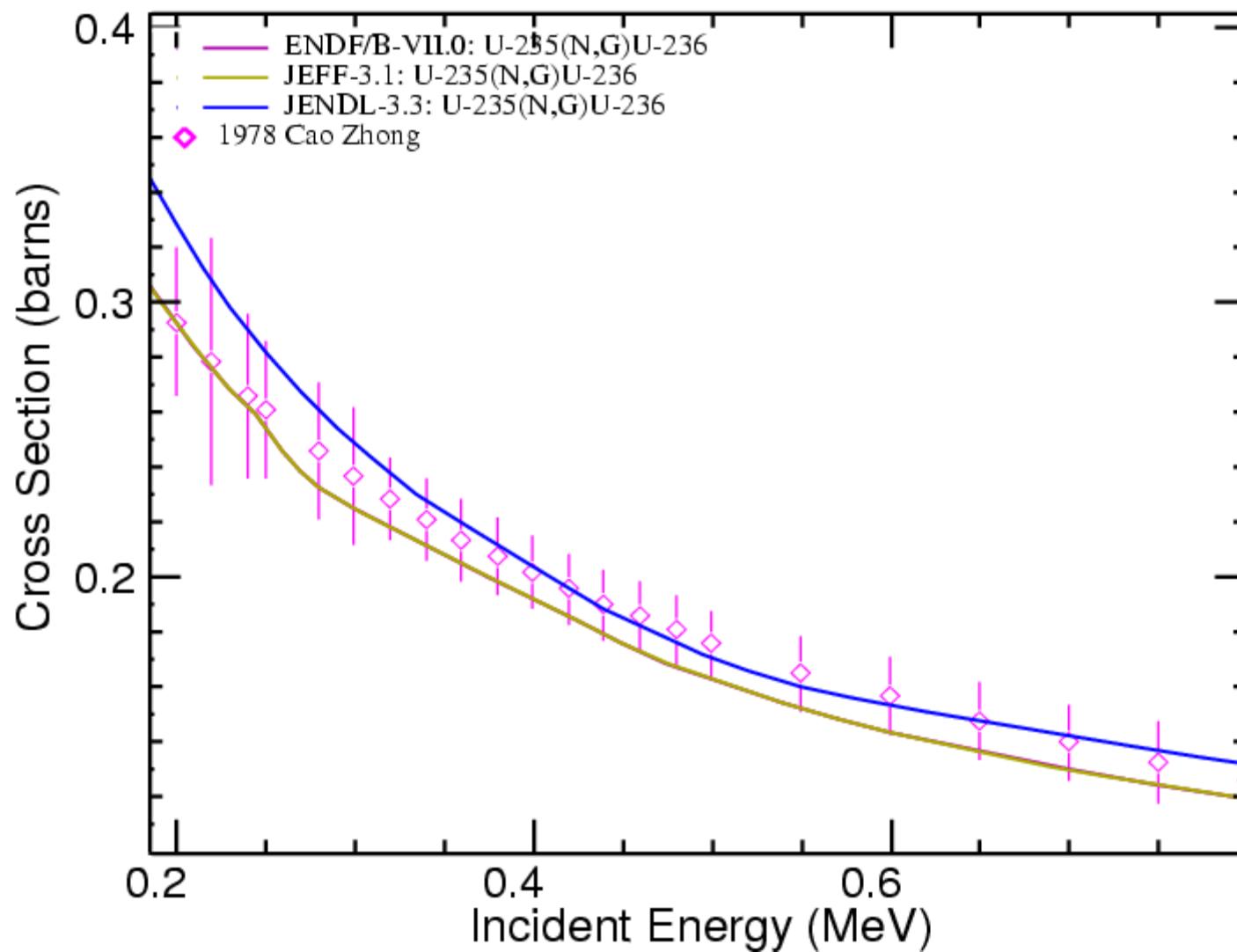
$^{235}\text{U}(\text{n},\gamma)^{236}\text{U}$

EXFOR database

ENDF = JEFF

5 – 80 keV

ENDF Request 360, 2010-Apr-20, 17:26:55
EXFOR Request: 46218/1, 2010-Apr-20 16:38:59



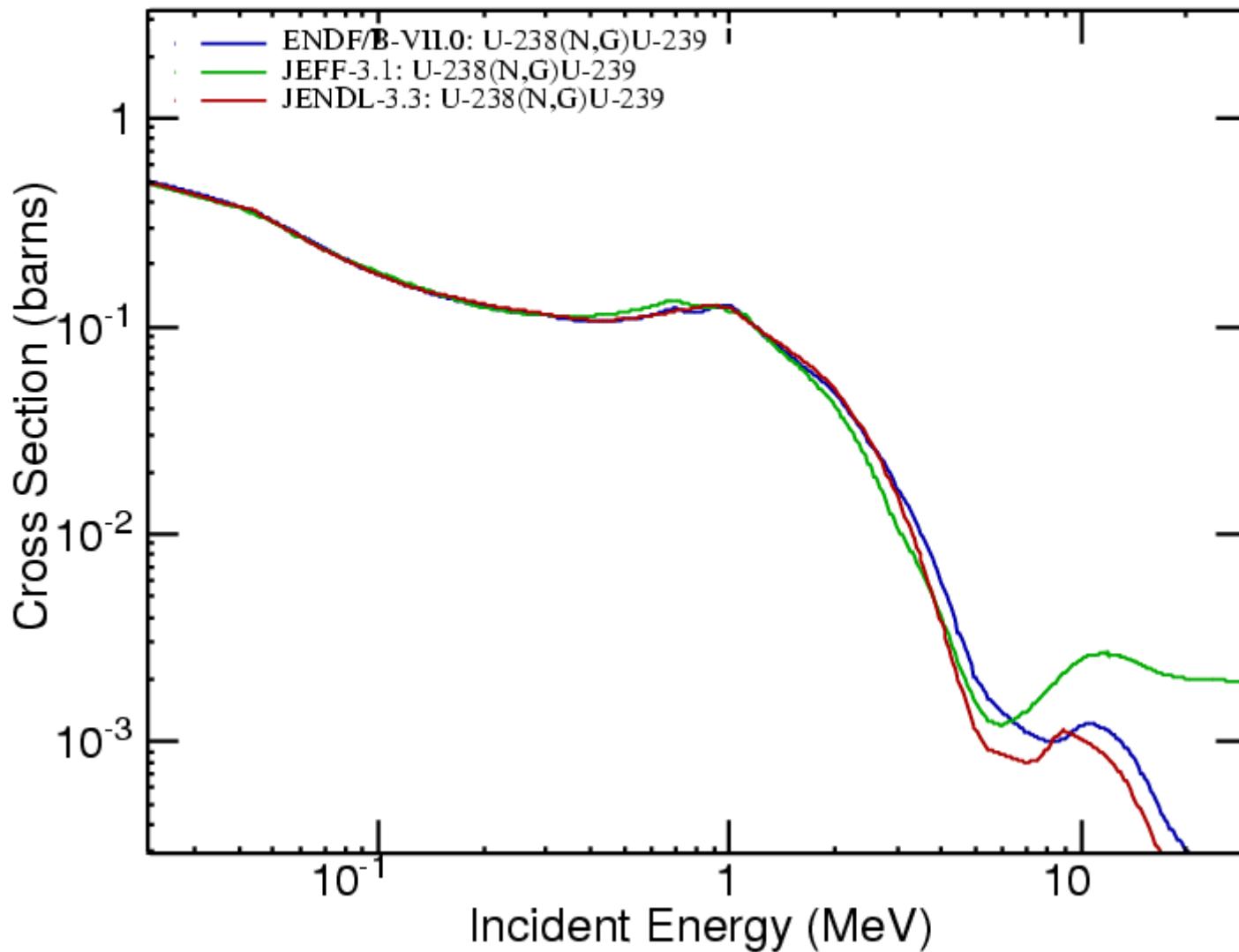
$^{235}\text{U}(\text{n},\gamma)^{236}\text{U}$

EXFOR database

ENDF = JEFF

180 – 800 keV

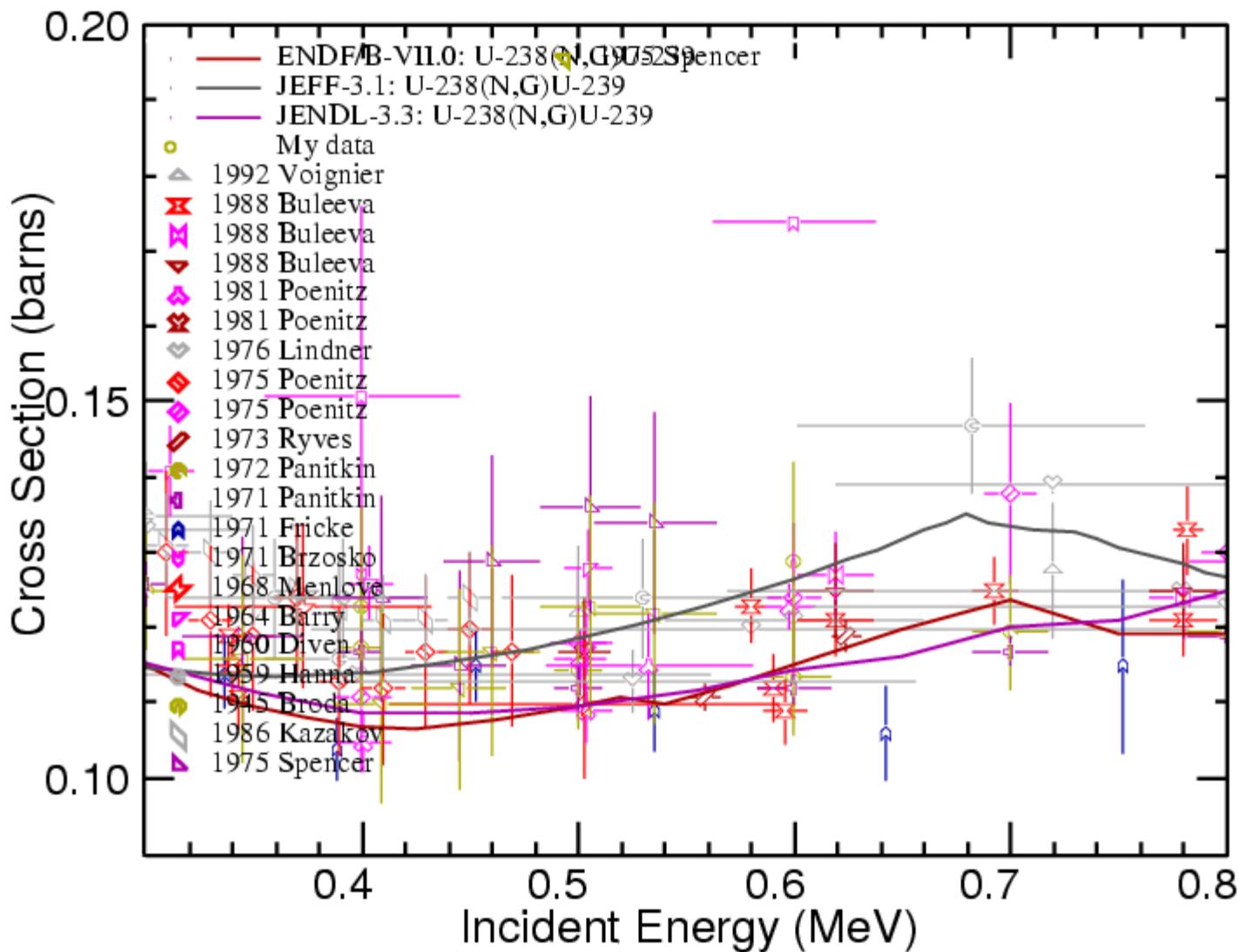
ENDF Request 352, 2010-Apr-20, 16:00:00



IAEA database

20 keV– 20 MeV

ENDF Request 352, 2010-Apr-20, 16:00:00
EXFOR Request: 46235/1, 2010-Apr-20 18:03:30



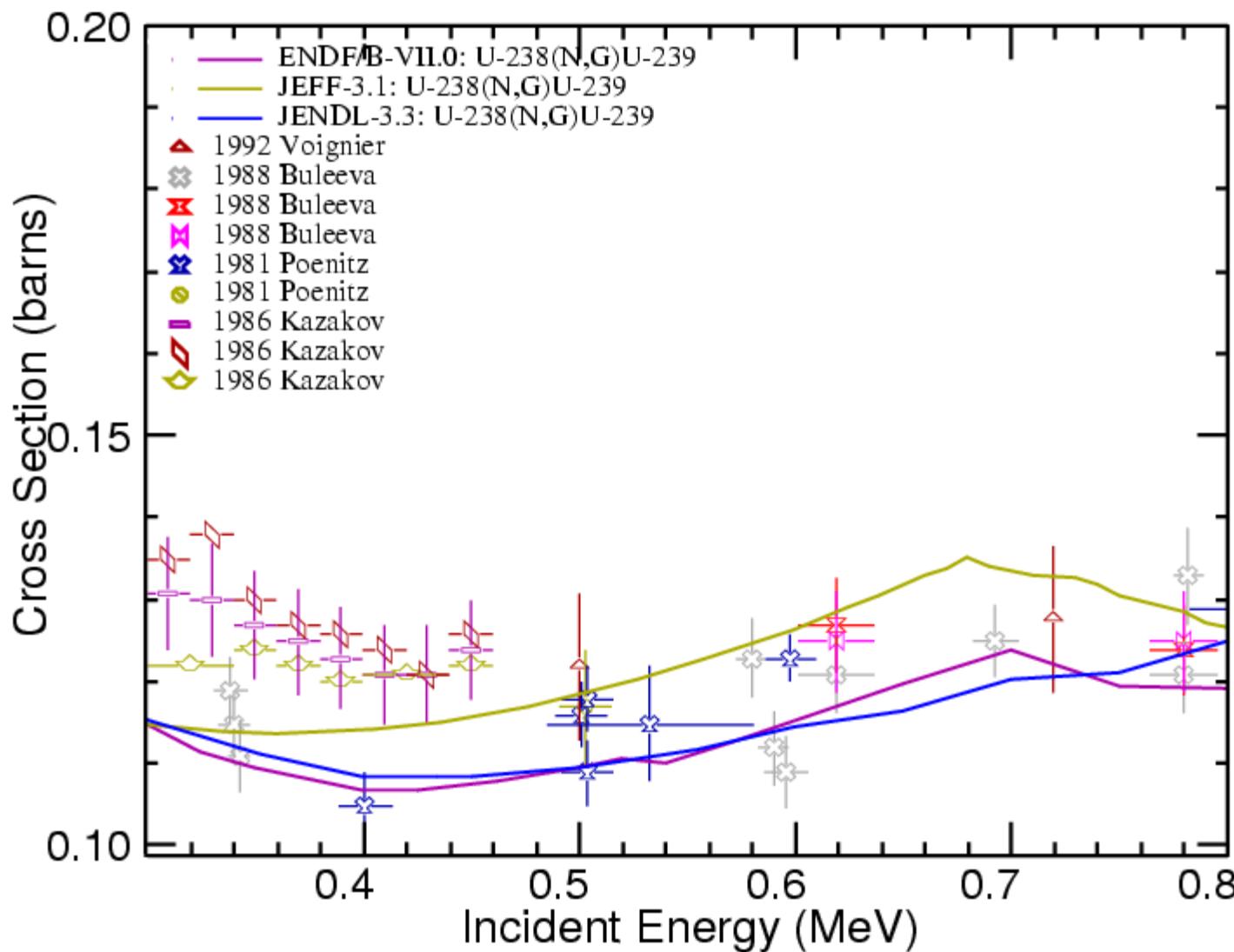
$^{238}\text{U}(\text{n},\gamma)^{239}\text{U}$

EXFOR database

300 – 800 keV

exp. data

ENDF Request 356, 2010-Apr-20, 16:27:00
EXFOR Request: 46190/1, 2010-Apr-20 16:04:42



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

EXFOR database

300 – 800 keV

exp. data > 1980

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

	Cm 237 ?	Cm 238 2.4 h	Cm 239 3 h	Cm 240 27 d	Cm 241 32.8 d	Cm 242 162.94 d	Cm 243 29.1 a	Cm 244 18.10 a	Cm 245 8500 a	Cm 246 4730 a	
	α 6.656	ϵ α 6.558; 6.503 γ 55	ϵ α 6.558; 6.503 γ 188... g	ϵ α 6.291; 6.248... sf g	ϵ α 5.939... sf g	ϵ α 6.113; 6.069... sf; g γ (44...); e^- σ = 20 σ_f = 5	ϵ α 5.785; 5.742... sf; g γ (278; 228; 210...); e^- σ = 15; σ_f = 1.1	ϵ α 5.805; 5.762... sf; g γ (43...); e^- σ = 15; σ_f = 1.1	ϵ α 5.381; 5.304... sf; g γ (175; 133...); σ = 350; σ_f = 2100	ϵ α 5.386; 5.343... sf; g γ (45); e^- σ = 1.2; σ_f = 0.16	
Am 235 10.3 m	Am 236 2.9 m ϵ α 6.457 γ 291; 224; 270; 739; 749...	Am 237 3.6 m ϵ α 6.15 ? γ 583; 654; 713... σ = 880; 320...	Am 238 73.0 m ϵ α 6.042... γ 280; 438; 474; 908... g	Am 239 1.63 h ϵ α 5.94 γ 963; 919; 561; 805... g	Am 240 11.9 h ϵ α 5.774... γ 278; 228... g	Am 240 50.8 h ϵ α 5.378... γ 300; 809... g	Am 241 432.2 a ϵ α 5.486; 5.443... sf; γ 80; 26... e^- ; g; σ = 60 + 840 σ_f = 3.15	Am 242 141 a ϵ α 5.486; 5.443... sf; γ (49); e^- σ = 0.7; σ_f = 142... σ = 206... σ_f = 142... σ = 1700; σ_f = 330 σ_f = 590; σ_f = 2100	Am 243 16 h ϵ α 5.275; 5.233... sf; γ 75; 44... σ = 75 + 5 σ_f = 0.079	Am 244 7370 a ϵ α 5.275; 5.233... γ 744; 898; 154...; e^- σ = 1600; σ_f = 2200	Am 245 2.05 h ϵ α 5.275; 5.233... γ 253; (241; 296...); e^- ; g
Pu 234 8.8 h ϵ α 6.15... γ ; e^-	Pu 235 25.3 m ϵ α 5.85 γ 48; (756; 34...); e^- σ_f = 160	Pu 236 2.858 a ϵ α 5.788; 5.721... sf; Mg 26... γ (48; 109...); e^- σ_f = 2300	Pu 237 45.2 d ϵ α 5.334... γ 60...; e^- σ_f = 2300	Pu 238 87.74 a ϵ α 5.499; 5.452... sf; Si; Mg 26... γ (43; 100...); σ = 510; σ_f = 17	Pu 239 2.411 $\cdot 10^4$ a ϵ α 5.157; 5.124... sf; γ (52...); e^- ; m σ = 270; σ_f = 752	Pu 240 6563 a ϵ α 5.168; 5.124... sf; γ (45...); e^- ; m σ = 290; σ_f = 0.058	Pu 241 14.35 a ϵ α 5.02; g γ 4.896... e^- ; g σ = 370; σ_f = 1010	Pu 242 3.750 $\cdot 10^5$ a ϵ α 4.901; 4.856... sf; γ (45...); e^- ; g σ = 19; σ_f < 0.2	Pu 243 4.956 h ϵ α 4.901; 4.856... β^- 0.6... γ 84...; g σ < 100; σ_f = 200	Pu 244 8.00 $\cdot 10^7$ a ϵ α 4.589; 4.546... sf; γ e^- σ = 1.7	
Np 233 36.2 m ϵ α 5.54 γ (312; 299; 547...); σ_f = 900	Np 234 4.4 d ϵ ; β^+ ... γ 1559; 1528; 1602... σ_f = 900	Np 235 396.1 d ϵ ; α 5.025; 5.007... γ (26; 84...); e^- g; σ_f = 2700	Np 236 22.5 h ϵ ; β^- 0.5... γ (642...); e^- σ_f = 104...; e^- g; σ_f = 3000	Np 237 2.144 $\cdot 10^6$ a ϵ α 4.775; 4.723...; sf Mg 26; Ne 25; γ (53; 121...); σ_f = 98; σ_f = 0.07	Np 238 2.117 d ϵ α 4.494; 4.445... γ 984; 1029; 1026; 924...; e^- σ_f = 2600	Np 239 2.355 d ϵ α 4.04; 0.7... γ 106; 278... 228...; e^- ; g σ = 32 + 19; σ_f < 1	Np 240 7.22 m ϵ α 3.2... γ 555; 597... e^- ; g σ_f = 601; σ_f = 448; g	Np 241 13.9 m ϵ α 2.2... γ 566; 974; 601; 448...; g	Np 242 2.2 m ϵ α 2.7... γ 786; 780; 945... 1473...; g	Np 243 1.85 m ϵ α 2.7... γ 288; g	
U 232 68.9 a ϵ α 5.262... γ (58; 129...); e^- σ_f = 74	U 233 1.592 $\cdot 10^5$ a ϵ α 4.824; 4.783... Ne 25; γ (42; 97...); e^- σ_f = 47; σ_f = 530	U 234 0.0054 ϵ α 4.775; 4.723...; sf Mg 26; Ne 25; γ (53; 121...); σ_f = 98; σ_f = 0.07	U 235 0.7204 ϵ α 4.494; 4.445... γ 783; 642...; g σ_f = 51	U 236 120 ns ϵ α 4.398... γ 783; 642...; g σ_f = 51	U 237 0.75 d ϵ α 4.494; 4.445... γ 80; 208... e^- ; m σ_f = 100; σ_f < 0.35	U 238 99.2742 ϵ α 4.468 $\cdot 10^9$ a γ 28; γ (80...); σ_f = 27; σ_f < 6	U 239 23.5 m ϵ α 1.2; 1.3... γ 75; 44... σ = 22; σ_f = 15	U 240 14.1 h ϵ α 0.4... γ 144; (190...); e^- m			
Pa 231 3.276 $\cdot 10^4$ a α 5.014; 4.952; 5.028...; Ne 24; F 237 γ 27; 300; 303...; e^- σ_f = 200; σ_f = 0.020	Pa 232 1.31 d β^- 0.3; 1.3...; e^- γ 969; 894; 150...; e^- σ_f = 460; σ_f = 1500	Pa 233 27.0 d β^- 0.3; 0.6... γ 312; 300... 341...; e^- σ_f = 20 + 19; σ_f < 0.1	Pa 234 1.17 m β^- 2.3... γ (100); 787...; g σ_f < 500	Pa 235 24.2 m β^- 0.5... γ 131; 861; 883...; e^- σ_f = 5000	Pa 236 9.1 m β^- 2.0; 3.1... γ 642; 687; 1763...; g Bsf ?	Pa 237 8.7 m β^- 1.4; 2.3... γ 854; 865; 529; 541... g	Pa 238 2.3 m β^- 1.7; 2.9... γ 1015; 635; 448; 680... g	Pa 239 1.8 h β^- γ 522...681			
Th 230 7.54 $\cdot 10^4$ a α 4.687; 4.621... γ (68; 144...); e^- Ne 24; σ = 23.4 σ_f < 0.0005	Th 231 25.5 h β^- 0.3; 0.4... γ 26; 84... g	Th 232 100 β^- 1.2... γ 87; 29; 459...; e^- σ_f = 1500; σ_f = 15	Th 233 22.3 m β^- 0.2... γ 63; 92; 93... σ_f = 1.8; σ_f < 0.01	Th 234 24.10 d β^- 1.4... γ 417; 727; 696... g	Th 235 7.1 m β^- 1.4... γ 417; 727; 696... g	Th 236 37.5 m β^- 1.0... γ 111; (647; 196...)	Th 237 5.0 m β^-	Th 238 9.4 m β^- γ 89			

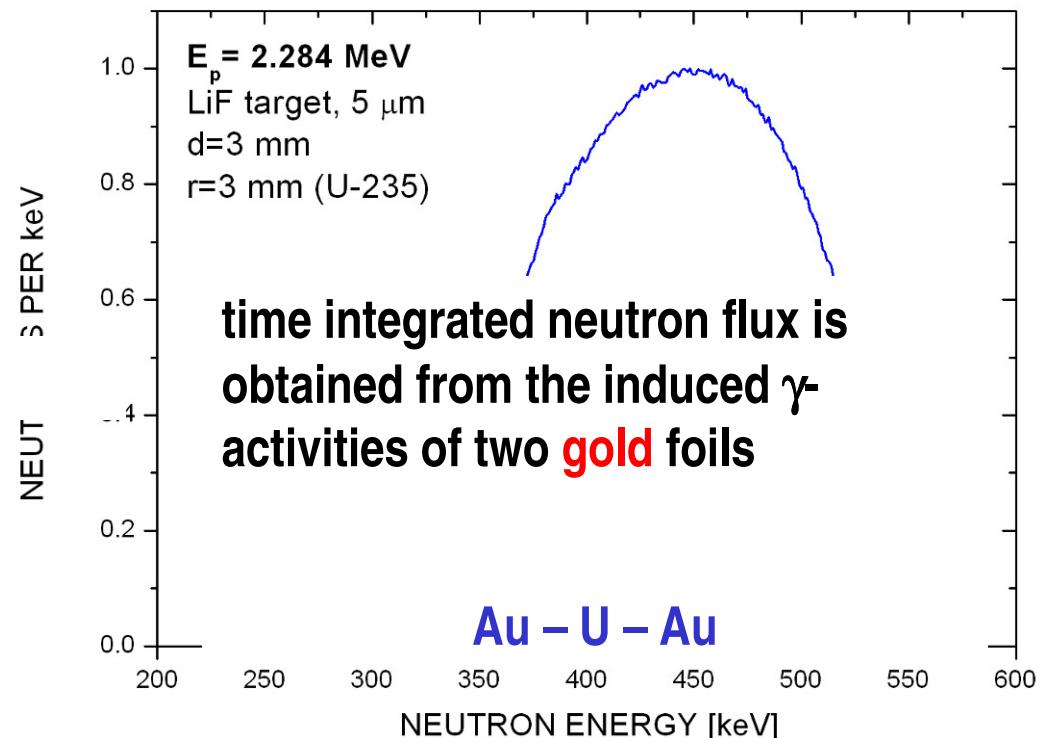
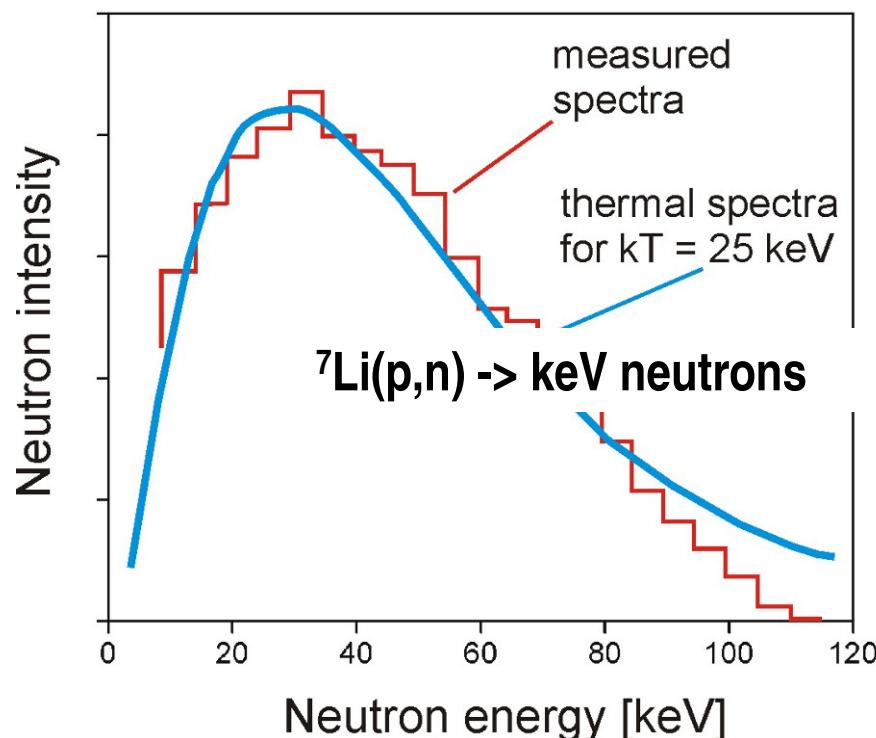
$$^{236}\text{U}: t_{1/2} = 23.4 \text{ Myr}$$

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

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

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

1st-irradiation FZK: 25 / 500 keV neutrons (EFNUDAT)



neutron energy: ~ 25 keV

fluence: ~ $1.7 \times 10^{15} \text{ ncm}^{-2}$

cross section: ~ 690 / 450 mb (U-5 / U-8)

~ 450 keV

~ $4 \times 10^{15} \text{ ncm}^{-2}$

~ 163 / 109 mb

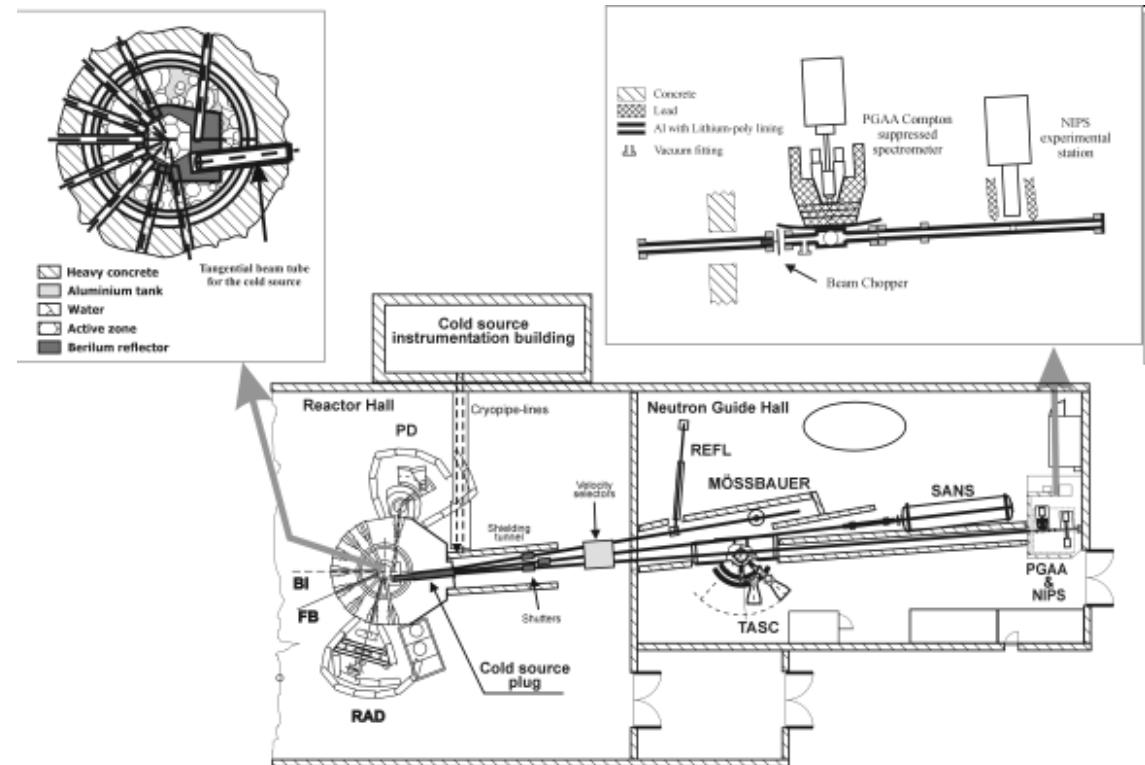
© Rene Reifarth et al.: PINO

irradiation at Budapest (IKI): cold neutrons

$^{235/8}\text{U}(n,\gamma)^{236/9}\text{U}$ – ENUDAT project

- Neutron irradiations at Budapest
- activations with cold neutrons
- time integrated neutron flux from
- γ -activities of gold foils and of
- Au powder mixed into the U sample
- ^{nat}U : ^{239}Np activity as additional internal monitor

Au – (U+Au) – Au



$^{235/8}U(n,\gamma)^{236/9}U$ – sample summary

- U samples with natural isotopic composition (0.72 % ^{235}U & 99.28 % ^{238}U)
- IRMM-BC0206: certified for stoichiometry - U_3O_8 & certified $^{235}U/^{238}U$ isotope ratio
- no enrichment; no reactor history
- selected sample: prior to neutron irradiation was found to be low ($^{236}U/^{238}U$ few $\times 10^{-12}$)

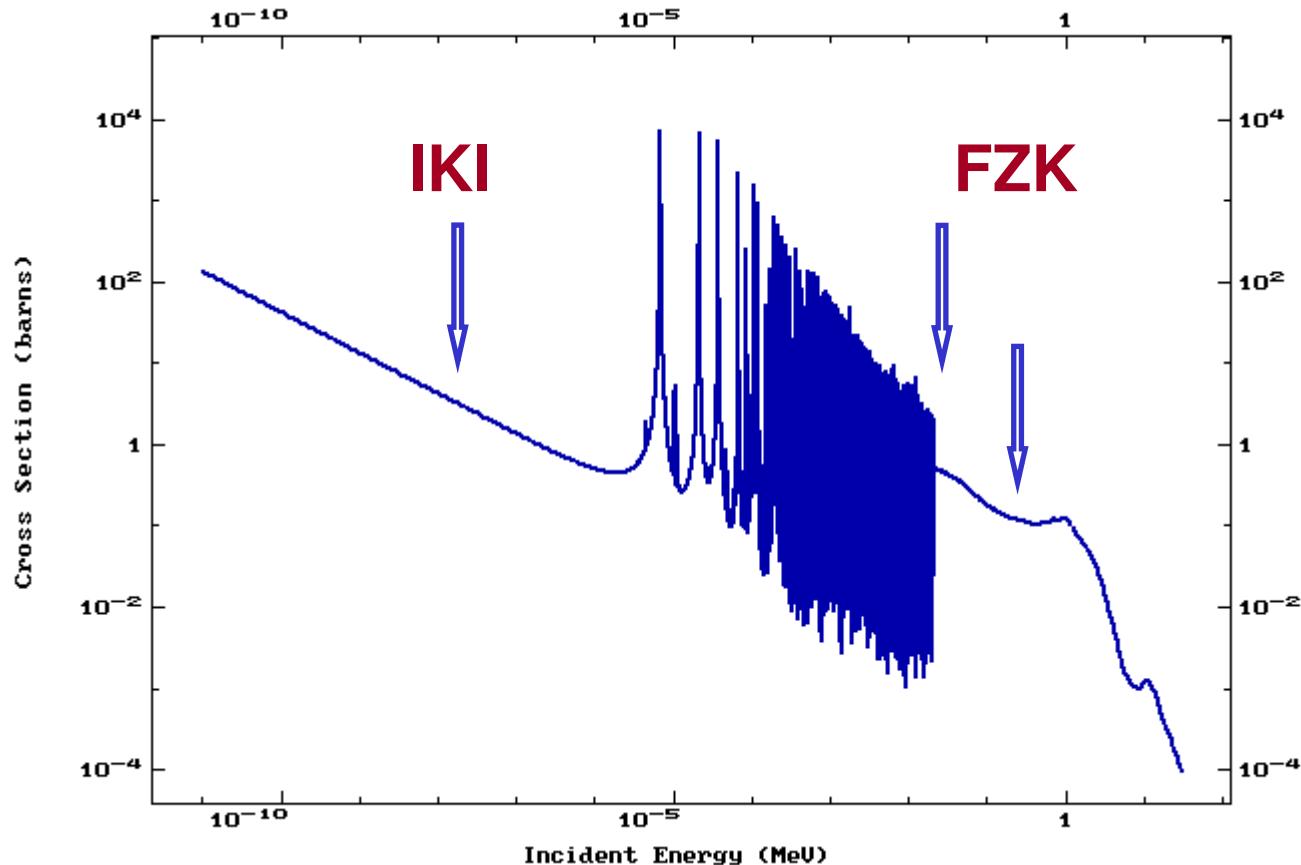
- FZK: 25 keV irradiation finished ($1.8 \times 10^{15} n \text{ cm}^{-2}$) 1 pellet 55 mg Au- U_3O_8 - Au
500 keV irradiation finished ($4.3 \times 10^{15} n \text{ cm}^{-2}$) 1 pellet 55 mg Au- U_3O_8 - Au
- IKI: cold neutrons finished (0.7 & $3 \times 10^{14} n \text{ cm}^{-2}$) 2 pellets 70 mg (Au- U_3O_8 +Au)- Au

- VERA: AMS measurements 2009-2010

expected isotope ratios: $^{236}U/^{238}U$ ca. $5-8 \times 10^{-12}$ and 2×10^{-11}

$^{239}Pu/^{242}Pu$: no background interference

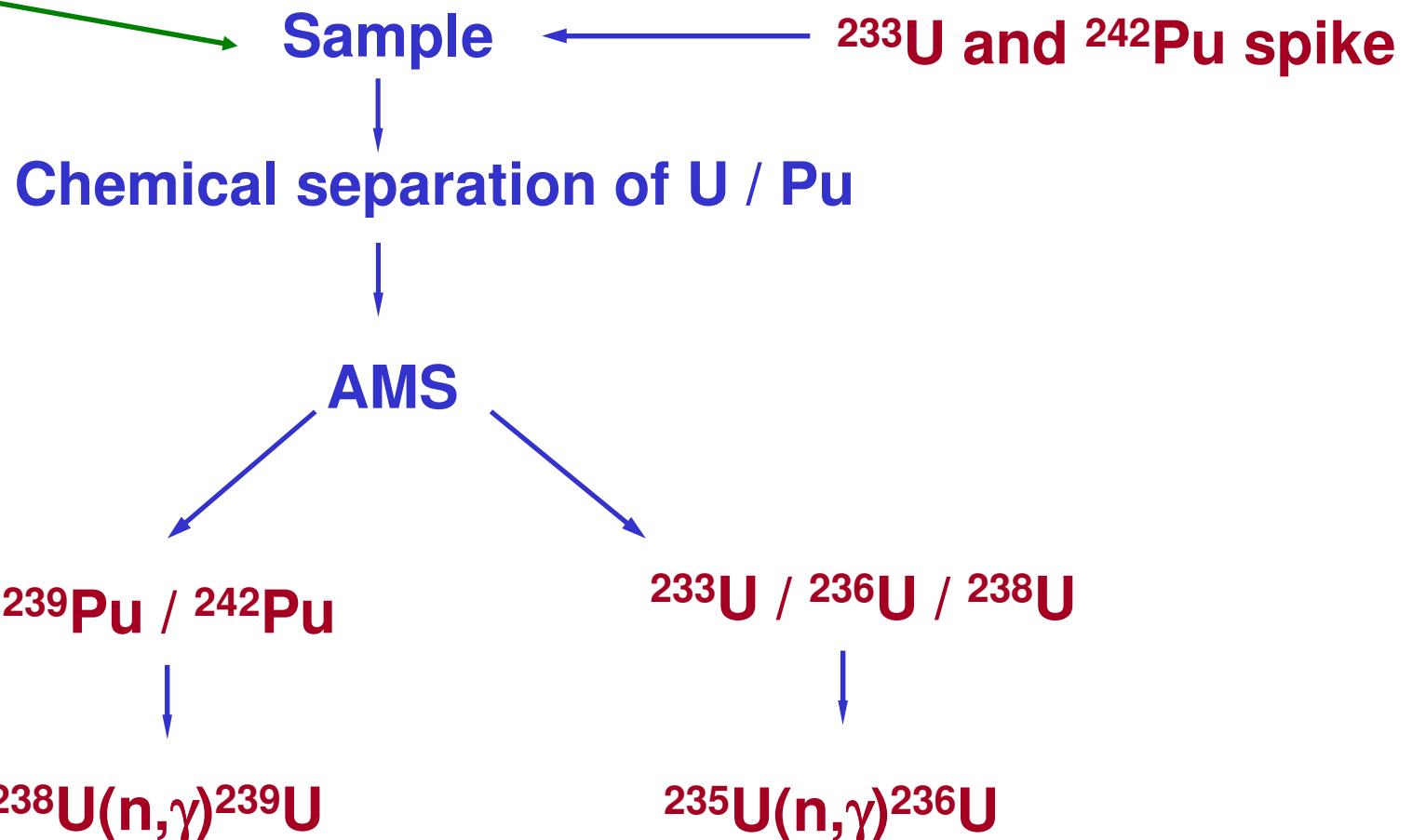
$^{238}U(n,\gamma)^{239}U \rightarrow ^{239}Np \rightarrow ^{239}Pu$ (24 kyr)



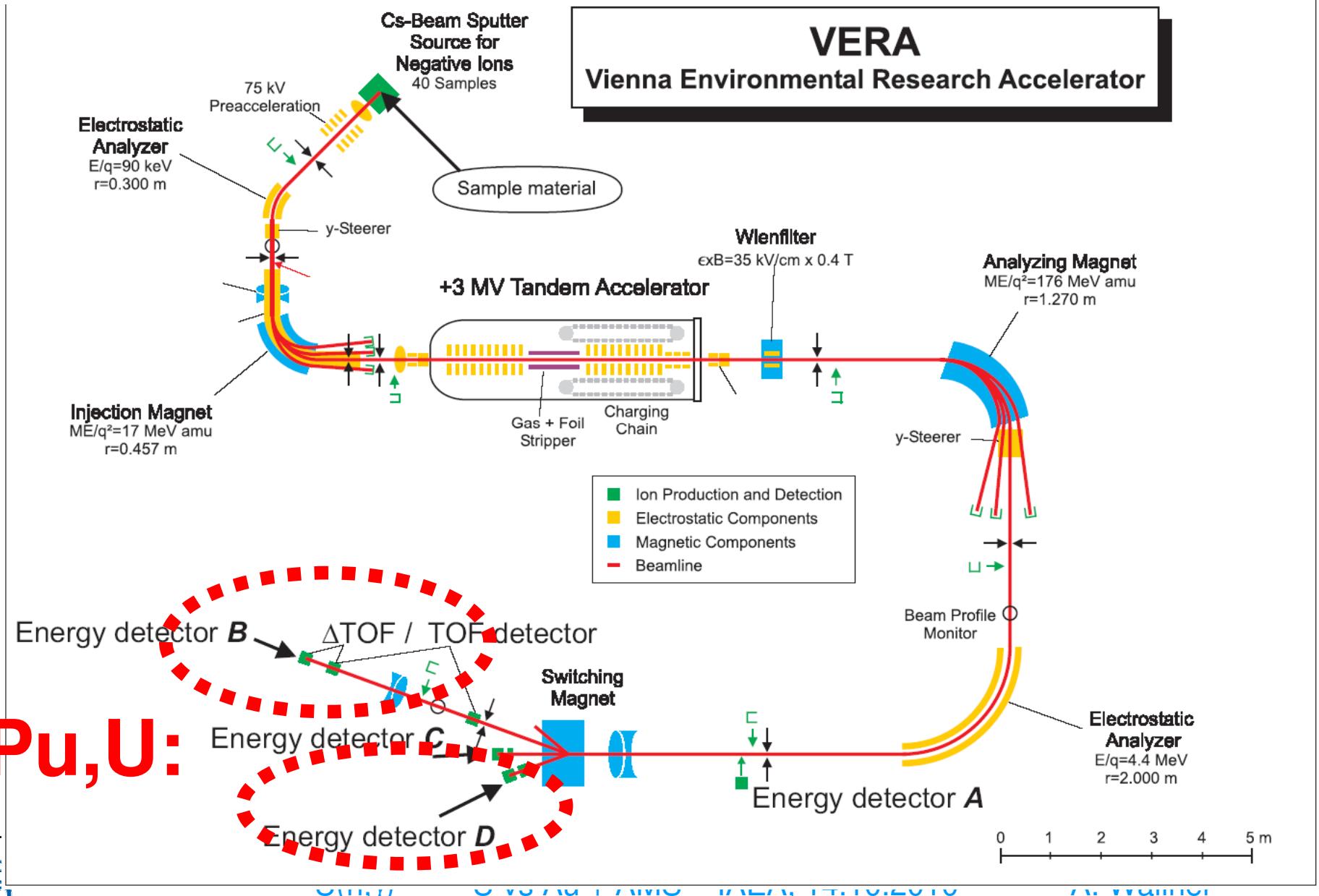
$^{235}U(n,\gamma)^{236}U$ (23 Myr) - simultaneously

Analytical Strategy

Au for fluence

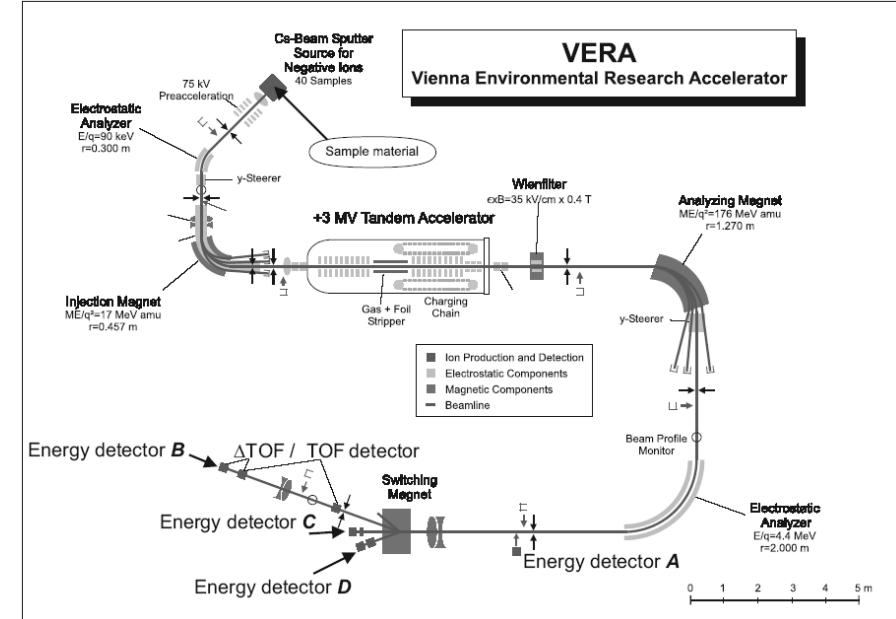


2nd: AMS measurement



2nd: $^{235/8}U(n,\gamma)^{236/9}U$ via AMS

- AMS determines isotope ratios
- no interfering isobaric background (<-> ICPMS)
(molecules are completely destroyed)
- isotopic background strongly reduced



$^{236}\text{U}/^{235}\text{U}$ isotope ratio at VERA

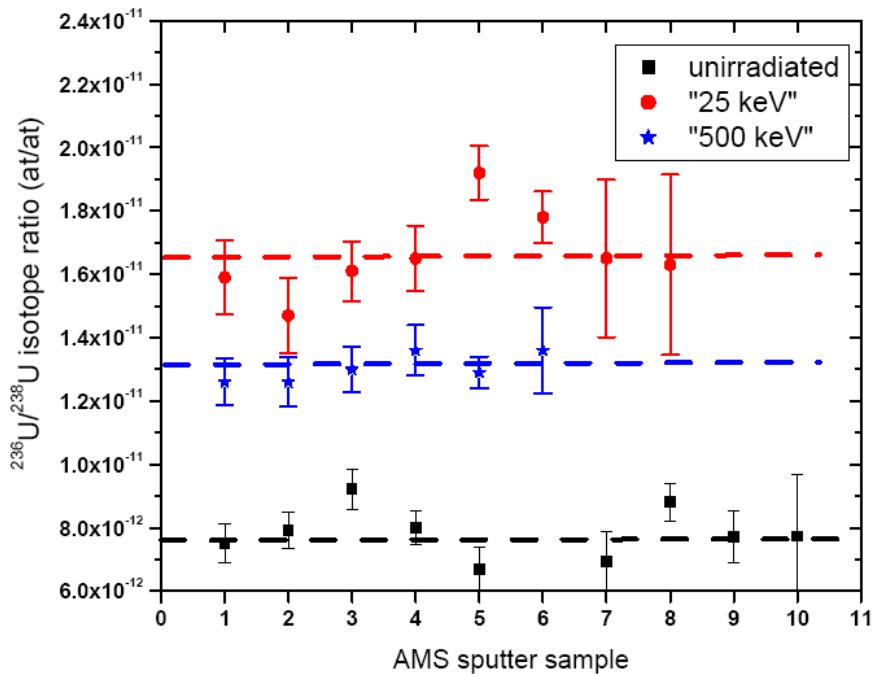
- isotope ratio via fast switching between "stable" and radioisotope (5 s^{-1})
- *precision limited* due to beam losses along detection beamline + background
- ^{236}U blank background of unirradiated sample

$^{239}\text{Pu}/^{238}\text{U}$ isotope ratio at VERA

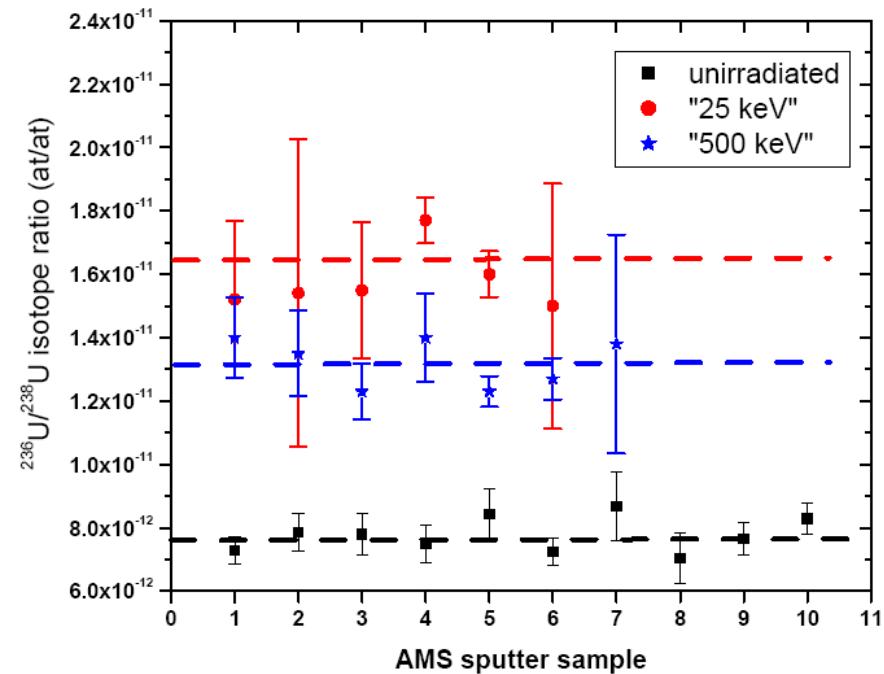
- isotope ratio via switching between reference isotope ^{242}Pu and ^{239}Pu
- *precision limited* due to beam losses along detection beamline + background
- no significant background for ^{239}Pu

$^{235}\text{U}(n,\gamma)^{236}\text{U}$: AMS results - 25 and 500keV

beamtime I

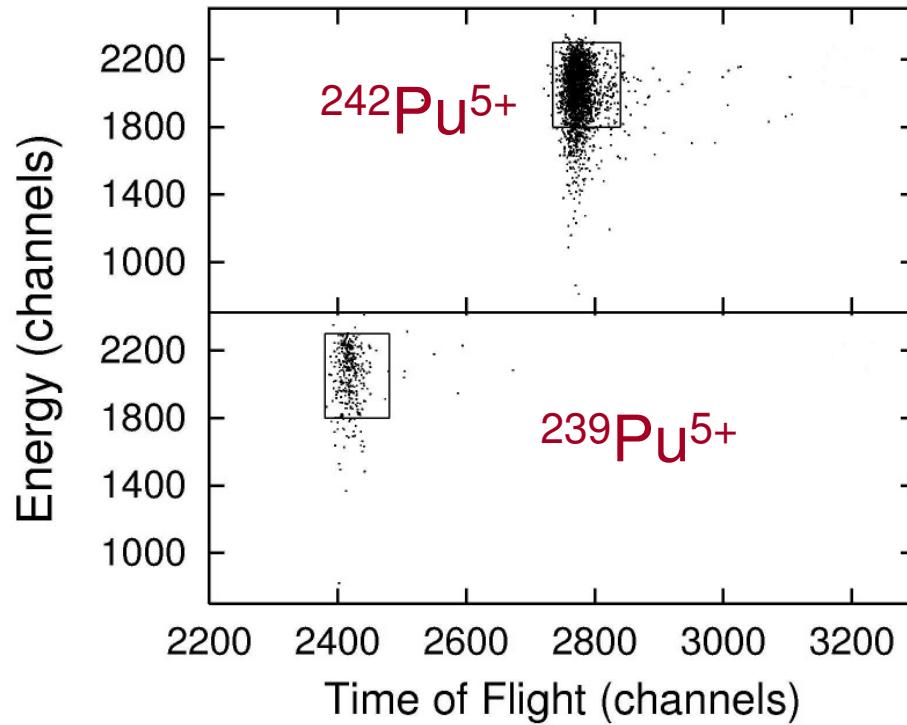


beamtime II



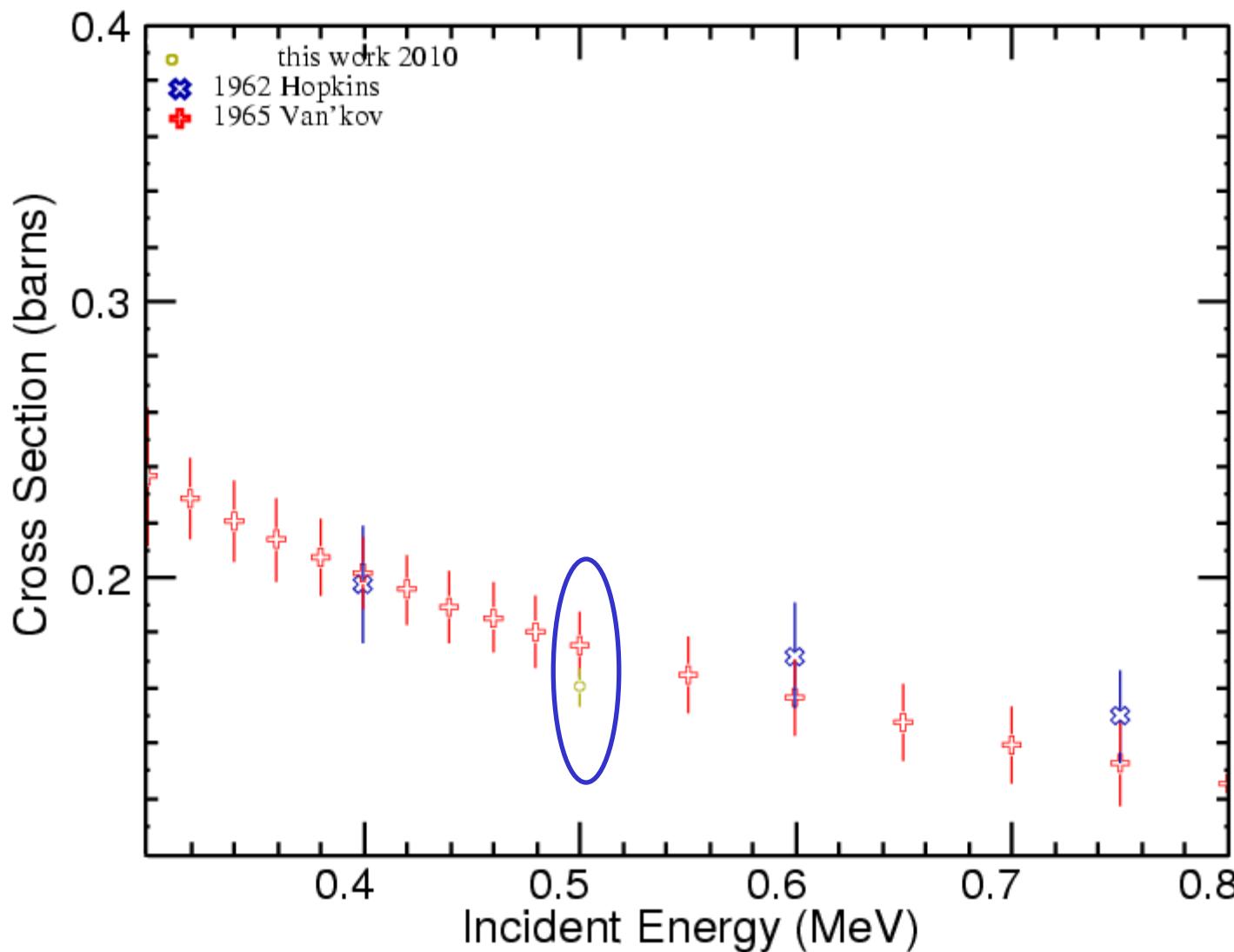
all data normalized to our in-house standard: $\pm 4\%$

typical AMS-spectrum for Pu measurements



92-U-235(N,G)92-U-236
EXFOR Request: 46230/1, 2010-Apr-20 17:35:45

Results



$^{235}\text{U}(\text{n},\gamma)^{236}\text{U}$

EXFOR database

300 – 800 keV

ENDF Request 369, 2010-Apr-20, 17:55:59
EXFOR Request: 46230/1, 2010-Apr-20 17:35:45

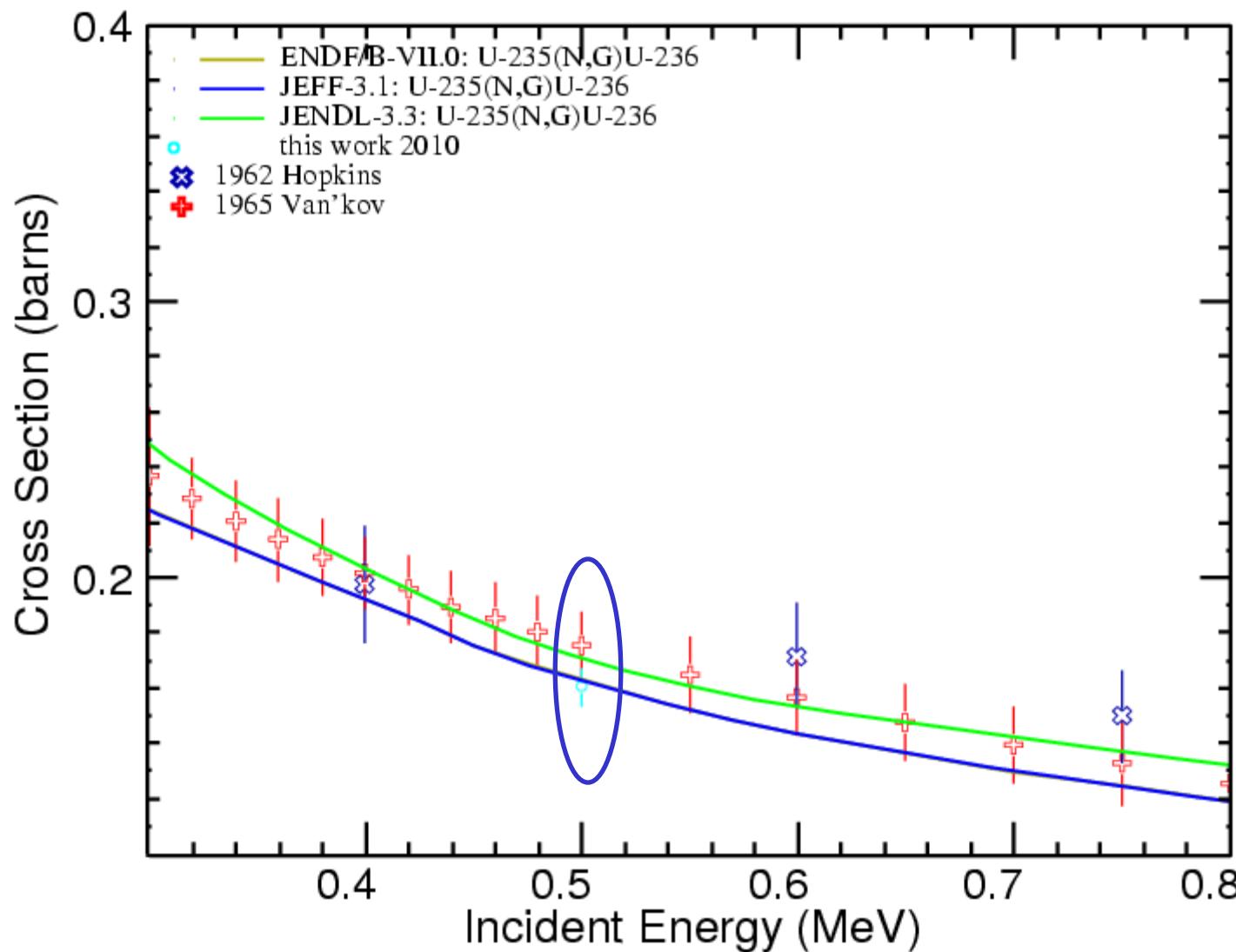
Results



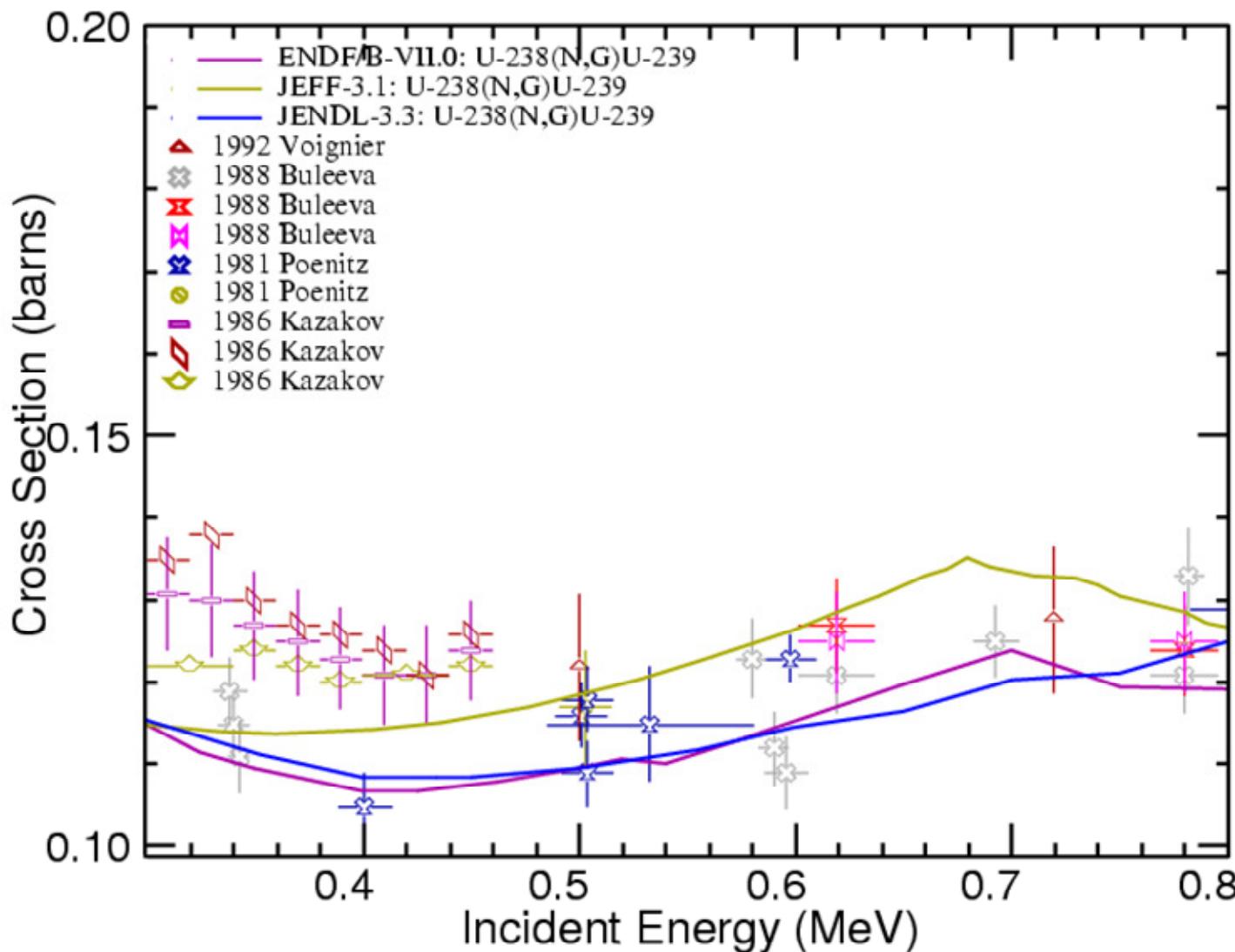
EXFOR database

ENDF = JEFF

300 – 800 keV



ENDF Request 356, 2010-Apr-20, 16:27:00
EXFOR Request: 46190/1, 2010-Apr-20 16:04:42



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

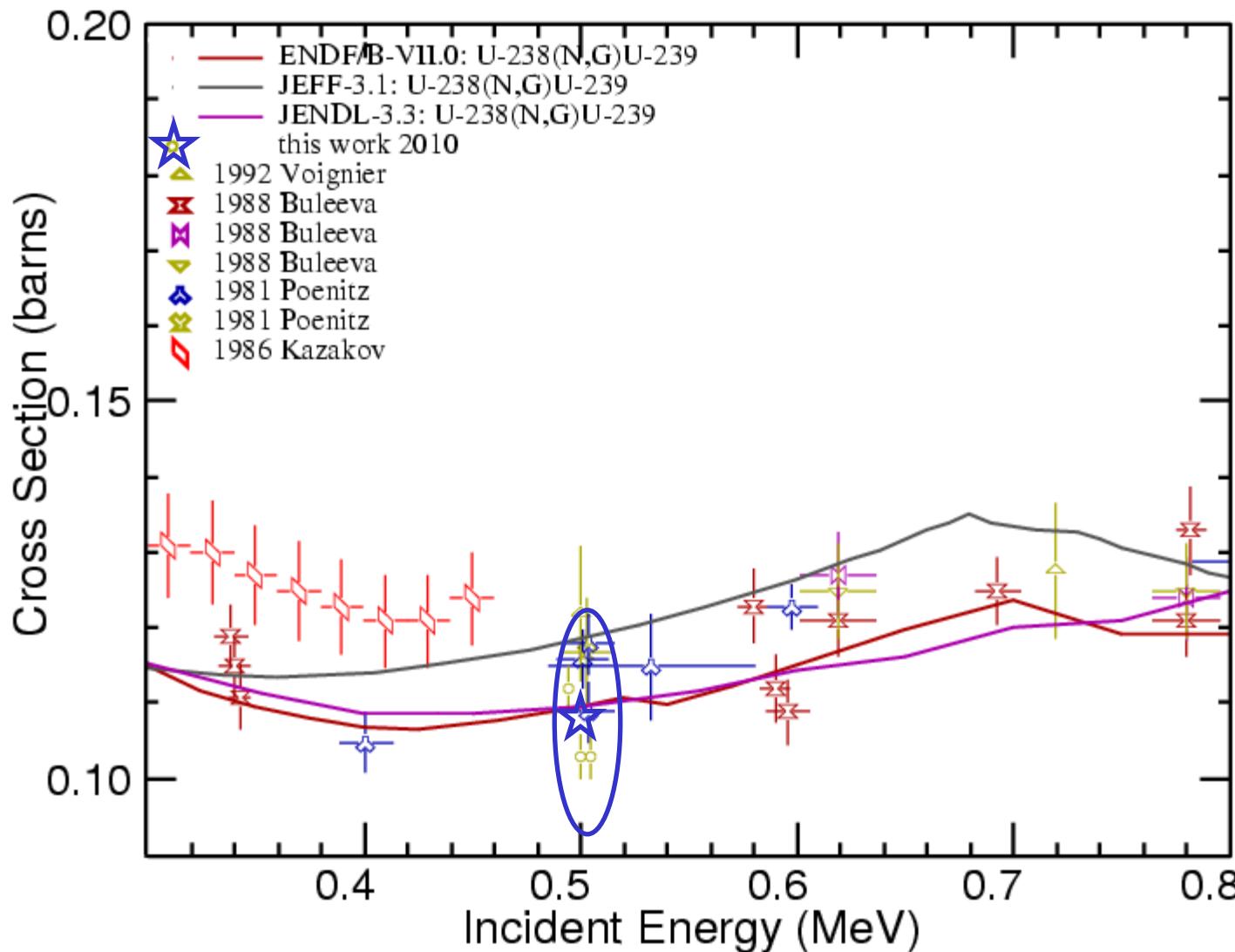
EXFOR database

300 – 800 keV

exp. data > 1980

ENDF Request 373, 2010-Apr-20,18:12:43
EXFOR Request: 46238/1, 2010-Apr-20 18:13:22

new data



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

EXFOR database

300 – 800 keV

exp. data > 1980

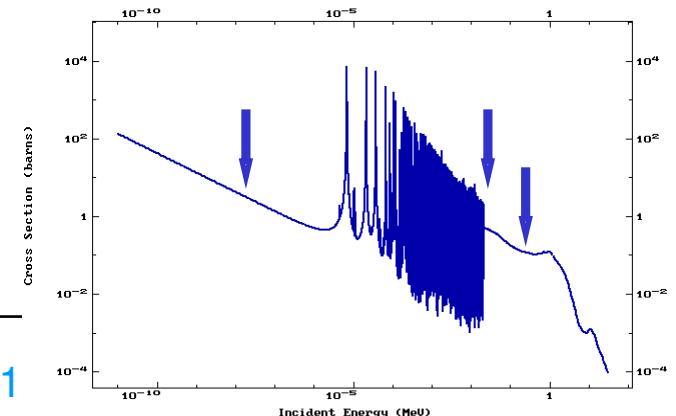
Status – preliminary data

	$^{235}\text{U}(\text{n},\gamma)^{236}\text{U}$ this work	$^{235}\text{U}(\text{n},\gamma)^{236}\text{U}$ „recommended“	$^{238}\text{U}(\text{n},\gamma)^{239}\text{U}$ this work	$^{238}\text{U}(\text{n},\gamma)^{239}\text{U}$ „recommended“
σ_{thermal}	IKI-1: 102 ± 1 barn* IKI-2: 103 ± 2 barn*	98.96 barn 98.96 barn	IKI-1: -- IKI-2: --	2.68 barn (2.72 barn JENDL)
$\sigma_{25 \text{ keV}}$	FZK-1: 591 ± 25 mbarn	ENDF: $\sigma_{\text{U-8}} / \sigma_{\text{U-5}} = 0.61$ this work (AMS ratio): $\sigma_{\text{U-8}} / \sigma_{\text{U-5}} = 0.63$ (ENDF = JEFF)	mean	360 mbarn calculated (ENDF = JEFF)
$\sigma_{500 \text{ keV}}$	FZK-2: 158 ± 7 mbarn	160-185 mbarn (ENDF, JEFF) ?? Spectrum??	FZK-2_a: 112 ± 2 mbarn** FZK-2_b: mean: 109 mbarn ** FZK-2_c: 104 ± 2 mbarn**	109 mbarn (ENDF, JENDL) 119 mbarn (JEFF)

* relative to $^{236}\text{U}/^{238}\text{U}$ in-house standard

= data relative to ^{233}U -spike - 3.5% lower - ok

** slightly different values expected due to inhomogenous activation



AMS: limitations

- isotope ratios (e.g. $^{236}\text{U}/^{238}\text{U}$) measured:
 - interference with background: VERA ca. $< 10^{-12}$ for ^{236}U
 - isobaric interference (e.g. ^{235}UH) – not an issue for AMS
 - isotopic interference (e.g. ^{235}U , ^{238}U): e.g. $^{236}\text{U}/^{238}\text{U} < 10^{-12}$
 - ev. m/q interference (count-rate interference - detector busy)
- number of nuclei in sample
 - determines statistical uncertainty
 - typical overall efficiency: $10^{-3} - 10^{-4}$

Examples - limitations

U: isotope ratios

20 mg irradiated with 10^{15} n; 700 mb

$$^{236}\text{U}/^{235}\text{U} = 7 \cdot 10^{-10}$$

$$^{236}\text{U}/^{238}\text{U} = 5 \cdot 10^{-12}$$

$$N_{^{236}} = 3 \cdot 10^8 \rightarrow \text{AMS: } > 10^5 \text{ counts in detector}$$

U: number of nuclei; example: $^{238}\text{U}(n,\gamma)^{239}\text{U} \rightarrow (0.5 \text{ h}) \rightarrow ^{239}\text{Np} \rightarrow (2.4 \text{ d}) \rightarrow ^{239}\text{Pu}$ ($t_{1/2} = 24 \text{ 000 a}$)

$$N_{^{239}} = 10^7 ! \text{ (required for AMS counting} \rightarrow \approx 10^3 \text{ detected)}$$

$$\sigma\phi = 1 \text{ barn} \cdot 10^{15} \text{ n cm}^{-2}$$

$$= 10^{-9}$$

$$N_{^{238}} = N_{^{239}} / \sigma\phi \text{ (required U-atoms)}$$

$$= 10^{16} \text{ ats (4 ug material)} + \text{extraction of } ^{239}\text{Pu from U-bulk} \\ (\text{with well-known spike})$$

Collaboration:

- VERA (Univ. Vienna): A. Wallner, O. Forstner, R. Golser, W. Kutschera, A. Priller, P. Steier, F. Quinto, K. Buczak, C. Lederer, G. Wallner
- FZK (Karlsruhe) / GSI: F. Käppeler, I. Dillmann
- IKI Budapest T. Belgya, L. Szentmiklosi
- IAEA (Vienna): A. Mengoni
- Atominstitut (Vienna): M. Bichler
- ...

$^{238}U(n,\gamma)^{239}U \rightarrow ^{239}Pu/^{242}Pu$ with AMS

- pellet was divided into 3 to 4 individual pieces
- dissolved
- well-known $^{233}U + ^{242}Pu$ spike added
- U / Pu column separation
- U- and Pu-Oxide powder
- pressed into AMS sample holder
- AMS: U: ^{236}U vs ^{233}U and vs $^{238}(^{235})U$
- AMS: Pu: ^{239}Pu vs ^{242}Pu



$$Z = N \cdot \sigma_{E_n} \cdot \Phi_{tot} \cdot f_b$$

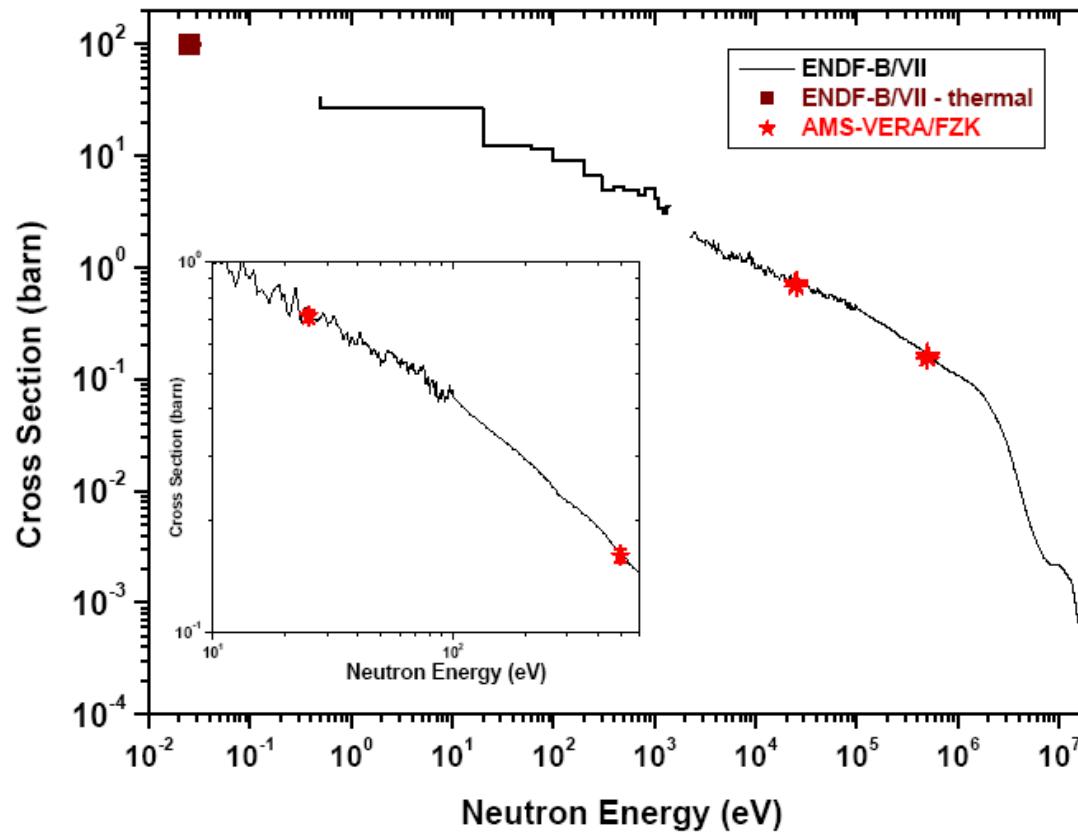
$$\sigma_{E_n} = \frac{Z}{N} \cdot \frac{1}{\Phi_{tot} \cdot f_b}$$

e.g. $\sigma_{E_n} = \frac{^{239}U}{^{238}U} \cdot \frac{1}{\Phi_{tot}} = \frac{^{239}Pu}{^{238}U} \cdot \frac{1}{\Phi_{tot}}$ for $T_{wait} \gg T_{1/2}(^{239}U)$

$$= \frac{^{239}Pu}{^{242}Pu} \cdot \frac{^{242}Pu}{^{238}U} \cdot \frac{1}{\Phi_{tot}}$$
 for $^{238}U(n,\gamma)^{239}U$

AMS mass Au

$^{235}\text{U}(n,\gamma)^{236}\text{U}$



preliminary data

+ adjust for broad neutron energy distribution

Summary U-activations

- new AMS measurements for thermal, 25 keV (MB) and 500 keV neutron energy
- data evaluation in progress: 500 keV \rightarrow < 5% (3%) uncertainty seems reasonable

$^{235}\text{U}(\text{n},\gamma)^{236}\text{U}$:

- thermal value as reference for 25/500 keV
- in-house AMS standard
- ^{233}U spike as additional standard

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

- thermal value as reference for 25/500 keV
- ^{242}Pu spike as additional standard

Extension to other minor actinides

U: isotope ratios

20 mg irradiated with 10^{15} n cm $^{-2}$; 700 mb

$$^{236}\text{U}/^{235}\text{U} = 7 \cdot 10^{-10}$$

$$^{236}\text{U}/^{238}\text{U} = 5 \cdot 10^{-12} \text{ (natural sample)}$$

$$N_{^{236}} = 3 \cdot 10^8 \text{ atoms} \rightarrow \text{AMS: } \sim 10^6 \text{ counts in detector}$$

Am: example: $^{241}\text{Am}(n,\gamma)^{242g}\text{Am} \rightarrow (16\text{h}) \rightarrow ^{242}\text{Cm} (163\text{ d})$

$$N_{^{242}} = 10^7 ! \text{ (required for AMS counting} \rightarrow \approx 10^3 \text{ detected)}$$

$$\sigma\phi = 700 \text{ mb} \cdot 10^{15} \text{ n cm}^{-2}$$

$$= 7 \cdot 10^{-10}$$

$$N_{^{241}} = N_{^{242}} / \sigma\phi$$

$$= 10^{16} \text{ ats} \quad (< 0.01 \text{ mg material}) \quad + !! \text{ Extract Cm from Am-bulk!!}$$

(400 cts ... 5 % statistics: 10^{15} ats ($< \mu\text{g!}$)

\leftrightarrow Ion source contamination! (^{241}Pu)