European Commission

Cross Section Measurements and Uncertainties of Cross Section Data

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ec.europa.eu/jrc/en/institutes/irmm

What will I cover?

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- Neutron-induced fission
- Fission quantities
  - The fission cross-section, σ_f
  - The mean number of neutrons emitted per fission: nubar or ν
  - The mean energy distribution of a fission neutron: neutron spectrum or χ_ν
  - The delayed neutron fraction, ν_d; fragments with β-delayed neutron emission
  - Fission fragments (FF): yields (FY), kinetic energy (TKE), angular distribution (ad), gammas

Overview High Priority Request List

Requests concerning fission

Backed by sensitivity

- ^{238}Pu(n,f) sig, nubar
- ^{239}Pu, ^{235}U prompt gammas, 7.5%, 0-10MeV
- ^{240}Pu(n,f) sig, nubar
- ^{241}Pu(n,f)
- ^{242}Pu(n,f)
- ^{241}Am(n,f)
- ^{242m}Am(n,f)
- ^{242}Cm(n,f)
- ^{244}Cm(n,f)
- ^{245}Cm(n,f)

General

- ^{233}U(n,f) sig,nubar th-10keV, 0.5%
- ^{239}Pu(n,f)/(n,g) eta/alpha 1%
- ^{243}Am(n,f) n-spectrum th-10MeV, 10%
- ^{244}Cm(n,f) n-spectrum th-10MeV, 10%

Most fission cross sections about 2% uncertainty

Nubar 1%-2%.

Uncertainty reduction is the important theme for most isotopes on the list.

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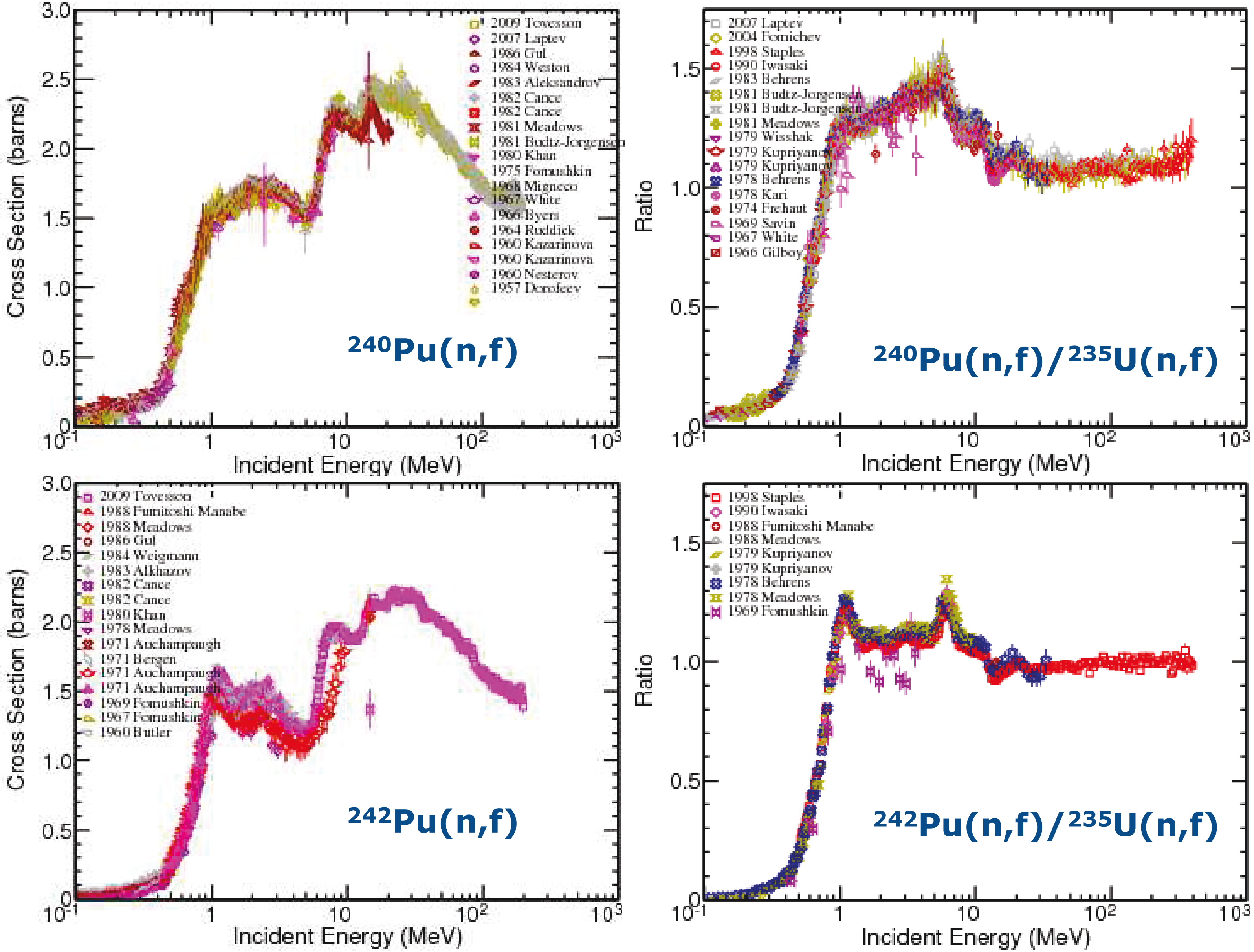
Example: ^{240}Pu(n,f) and ^{242}Pu(n,f) status



Despite considerable effort 10% spread

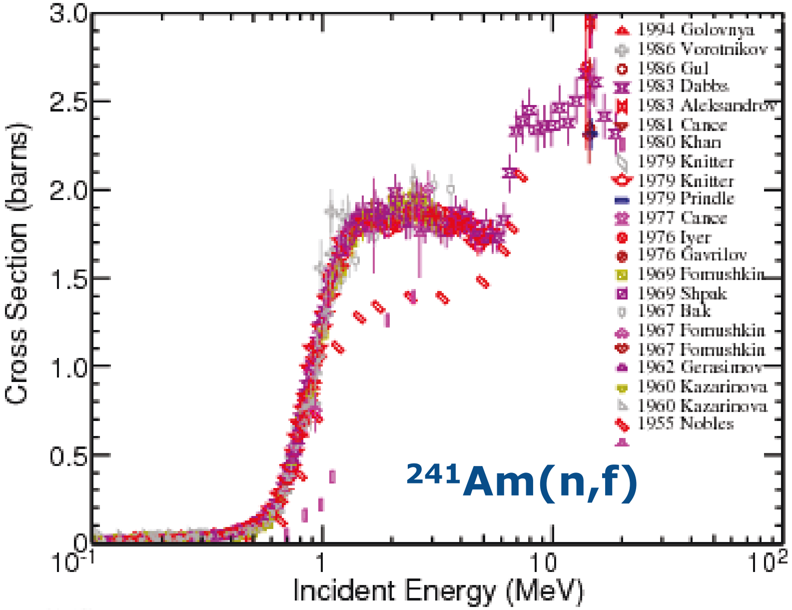
Many measurements claim 2% or better for the ratio

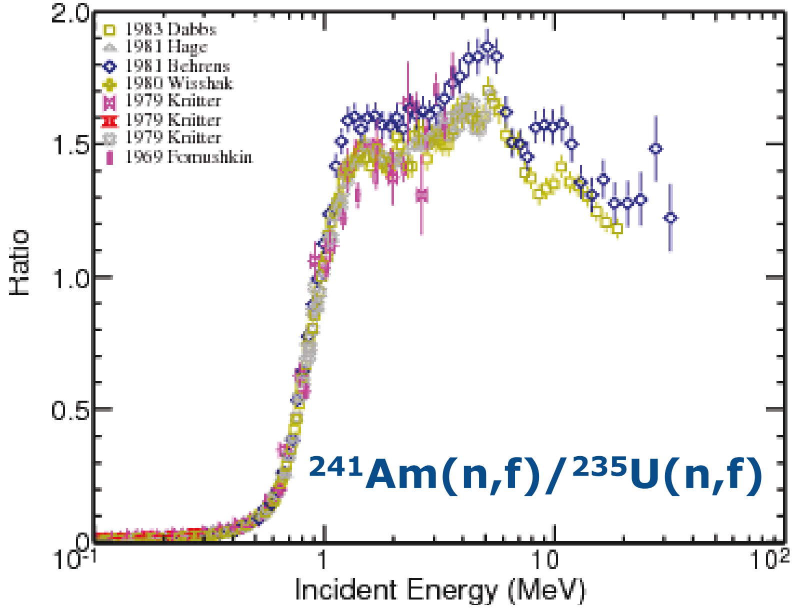
Many ratio data

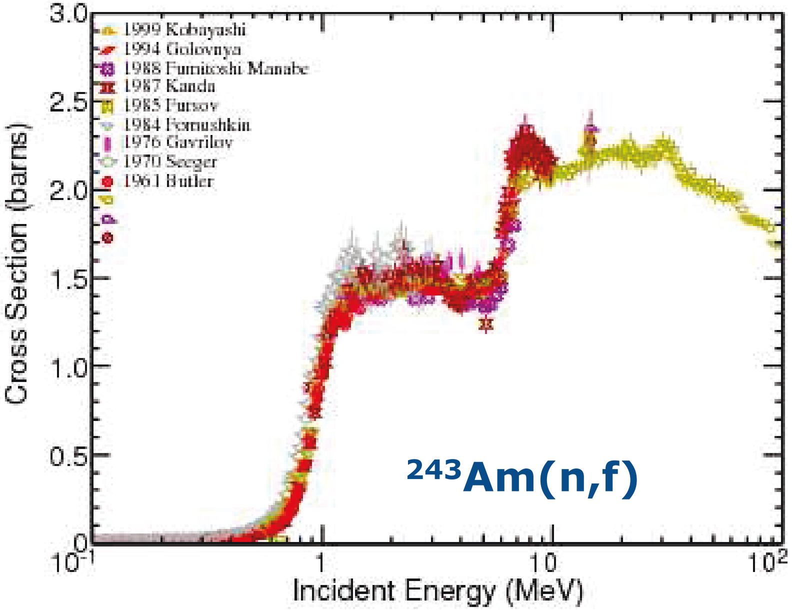
Almost all relative to ^{235}U(n,f)

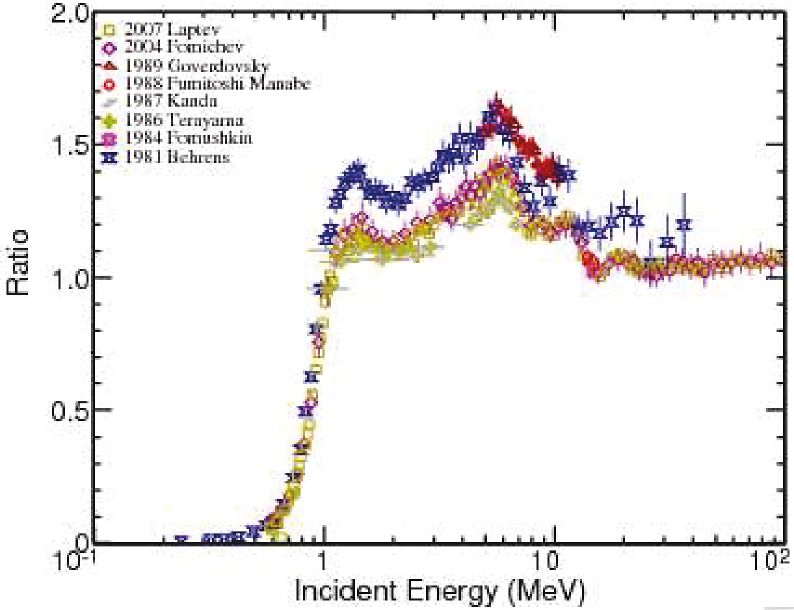
Lessons learned? What can we add?

Example: ^{241}Am (n,f) and ^{243}Am(n,f) status









Larger spread
Discrepancies
Fewer ratios to 235U for 241Am

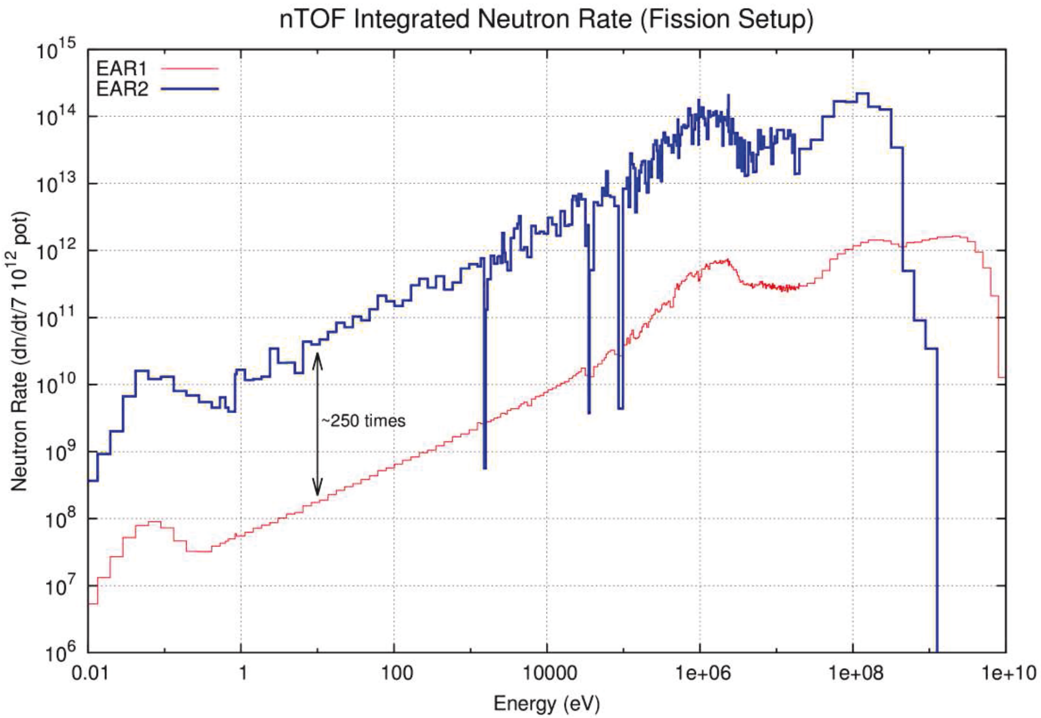
Experiments

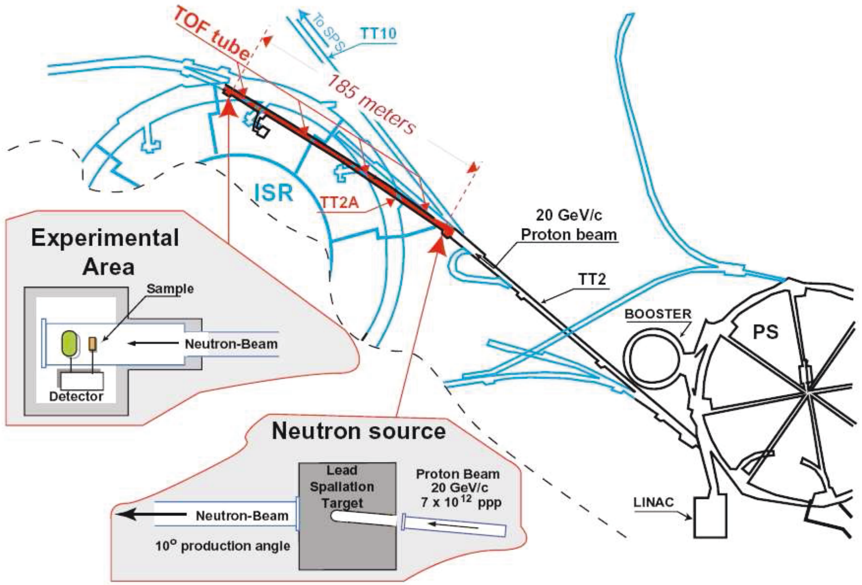
- Ingredients
  - Neutron source
  - Detectors for the process
  - Charactising flux or normalization
  - Sample material
- Characteristics differ strongly on the quantity studied
- % level cross sections require a lot of care for detail
- Important to know your experiment
- Important to learn from earlier experience
- Look for new technological developments

- Examples
  - Neutron sources
  - Cross sections
  - Neutron multiplicity
  - Neutron spectrum
  - Fission fragment yields
  - Kinetic energy, angular distributions
  - Prompt fission gammas
  - Delayed neutrons and gammas

CERN, LANL, JAEA, SNS, Nevis

Neutrons from spallation reactions (400 – 20 GeV)





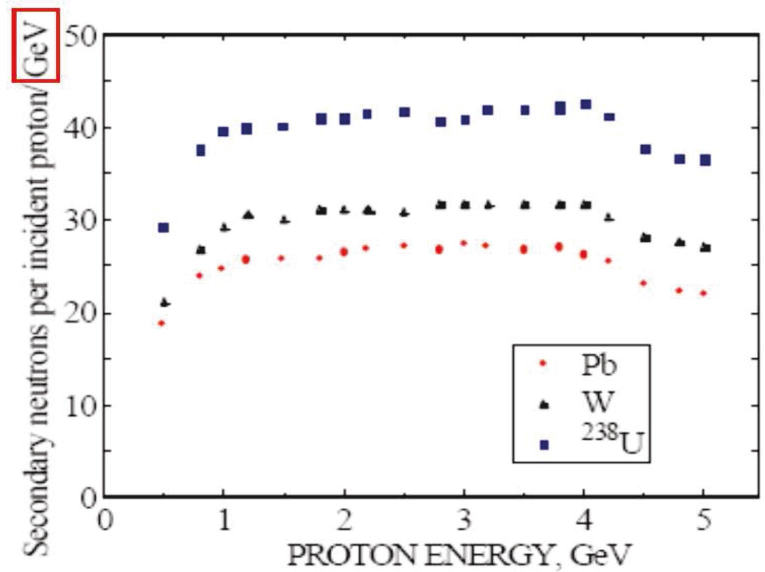
Neutron energies up to the proton energy

Neutrons/proton/GeV 25

Neutrons/pulse (nTOF) $2\ 10^{14}$

Neutrons/second (nTOF) $7\ 10^{13}$

Near future: EAR-2 at n_TOF, NFS @ SPIRAL2/GANIL



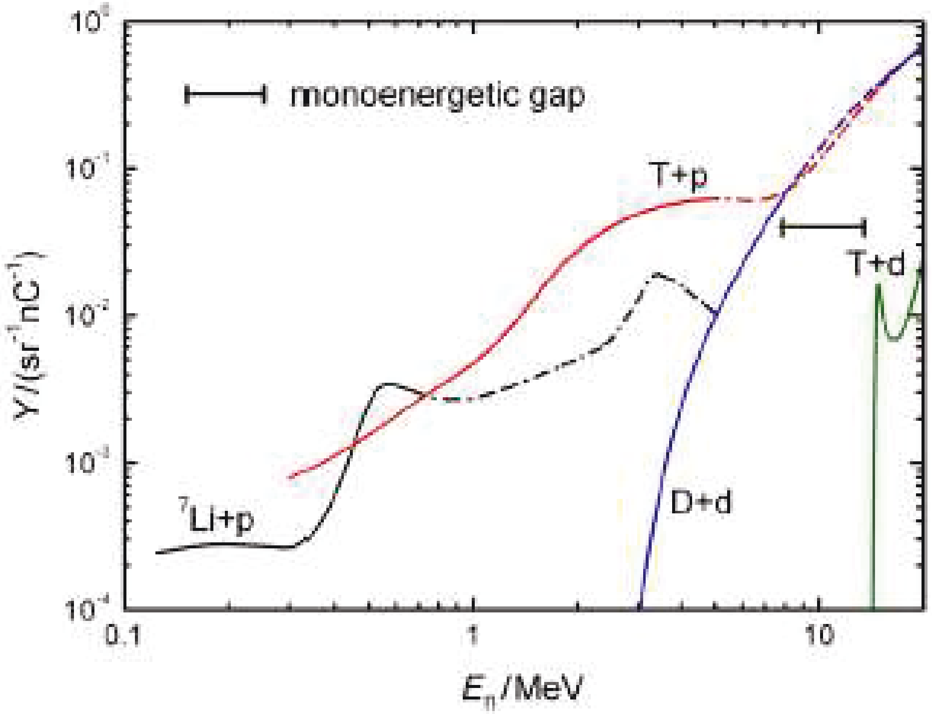
Quasi mono-energetic sources

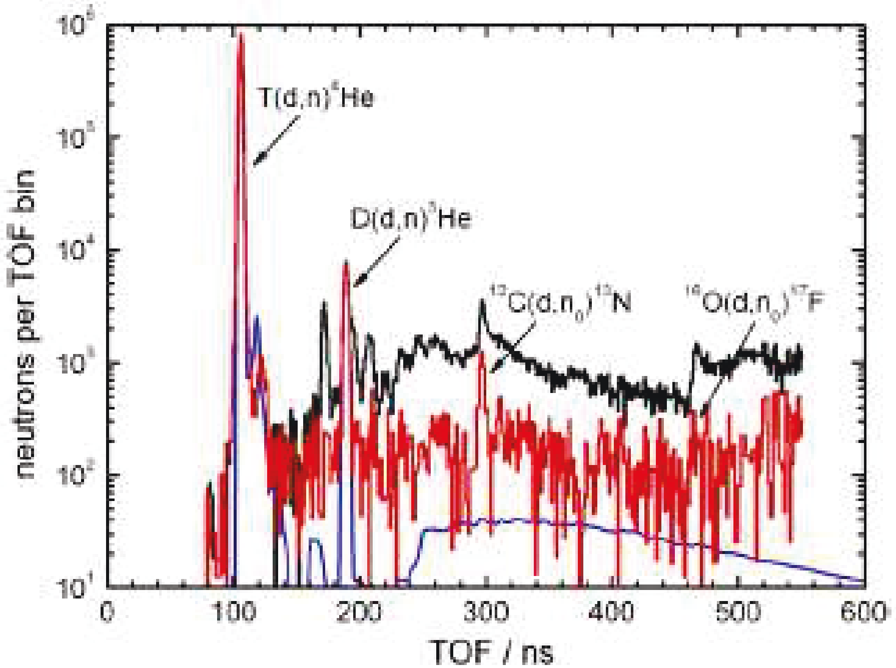
See presentation Nolte

- All flux at one energy
- Smaller cross sections
- Smaller samples

    E.g. Radioactive samples

- Charactised by reaction, energy loss, reaction kinematics, differential cross section
    - Primary neutrons, e.g. NeuSDesc
    - Background neutrons by TOF





Nolte and Thomas, Metrologia 48 (2011) S263
Birgersson and Lövestam, NeuSDesc, EUR 23794 EN (2009)

Cross sections

- Dominated by fission ionization chambers (FIC)
- Dominated by ratios to $^{235}U(n,f)$
- Important to know what is involved (FIC, ratios)
- Important to provide a good uncertainty analysis

- Other options?
  - Frisch-grid
  - PPAC
  - Solid state detectors
  - Normalization to hydrogen
  - Normalization to other fission

$$\sigma \equiv \frac{\text{Reaction rate}}{\text{flux}}(\text{per target nucleus}) = \frac{\dot{R}}{\Phi}$$

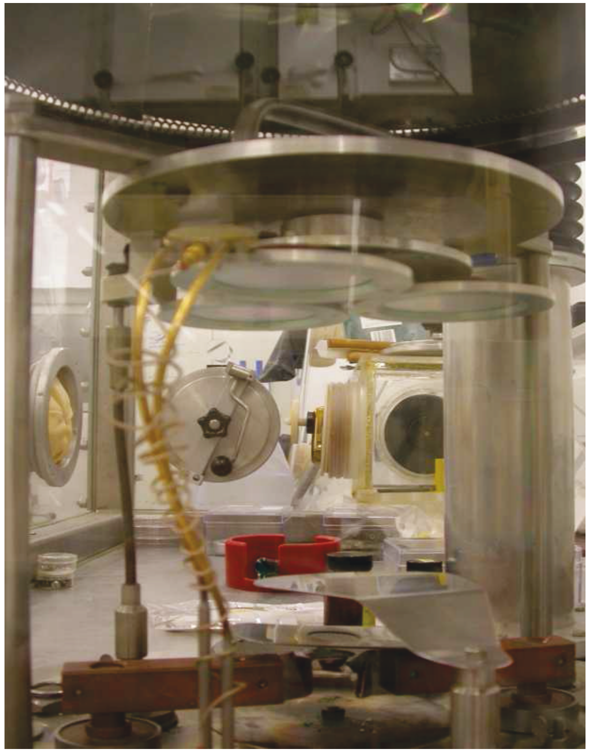
$$\sigma = \frac{\#\text{ reactions}}{\text{fluence}}(\text{per target nucleus}) = \frac{R}{\mathbf{F}}$$

$$\mathbf{F} = \Phi t, \mathbf{R} = \dot{\mathbf{R}}t, t = \text{measurement time}$$

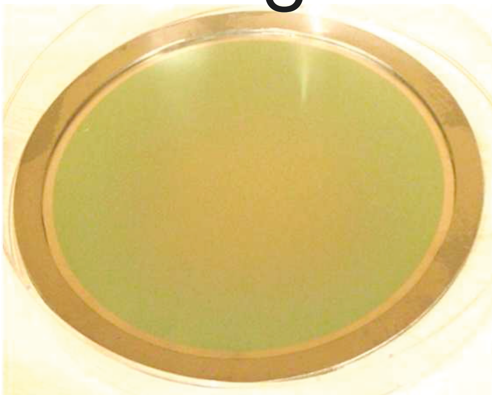
- Effects to consider
  - Sample quality
  - Selectivity
  - Detection efficiency
  - Background
  - Attenuation and scatter
  - Geometry
  - Source characteristics
  - Time resolution
  - …

Cross sections

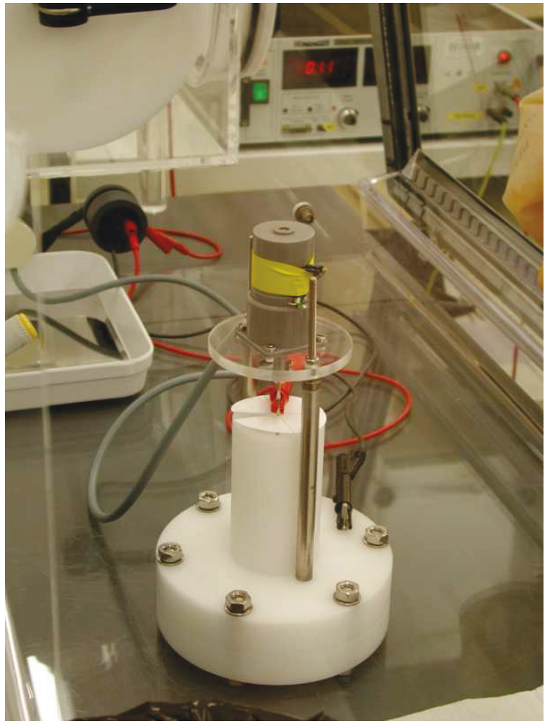
Sample preparation

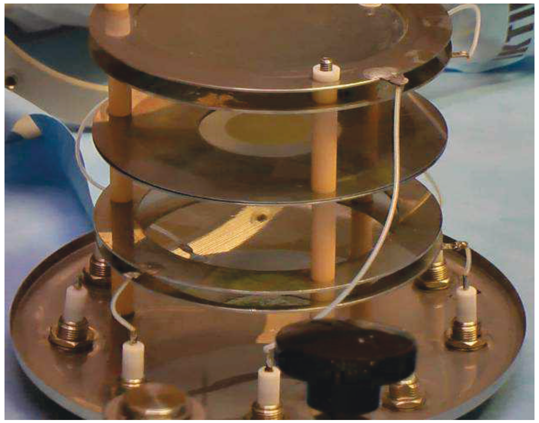


^{235}U

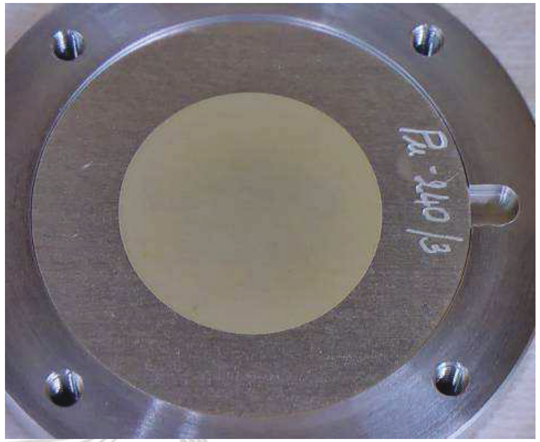


- **Evaporation**
  - **Best (^{235}U)**
  - **Inefficient**
- **Electrodeposition**
  - **Quality is less**
  - **Efficient**
  - **$^{240,242}Pu$**





^{242}Pu

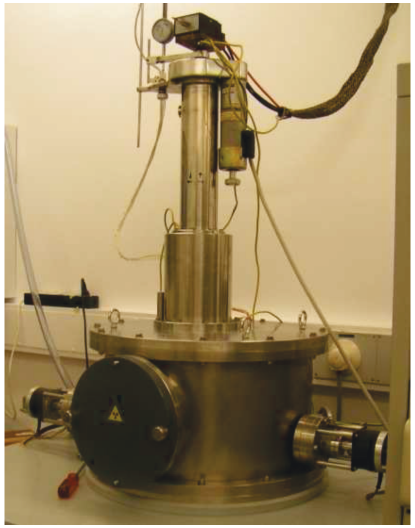


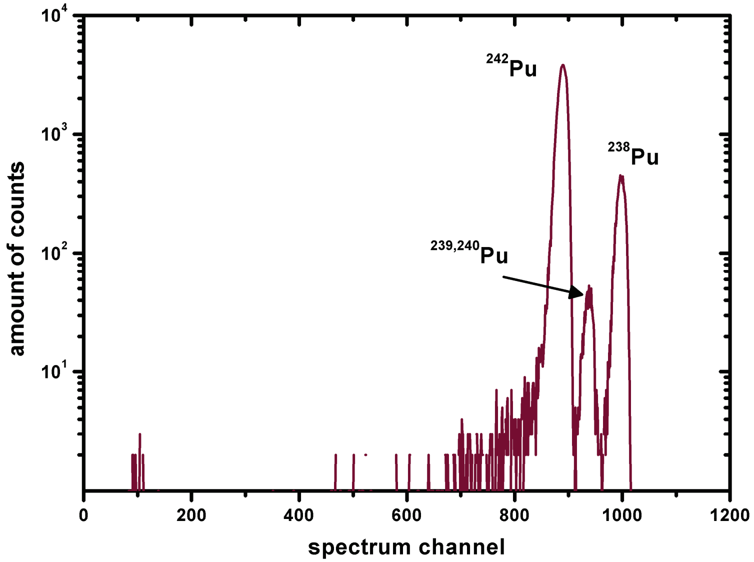
^{240}Pu

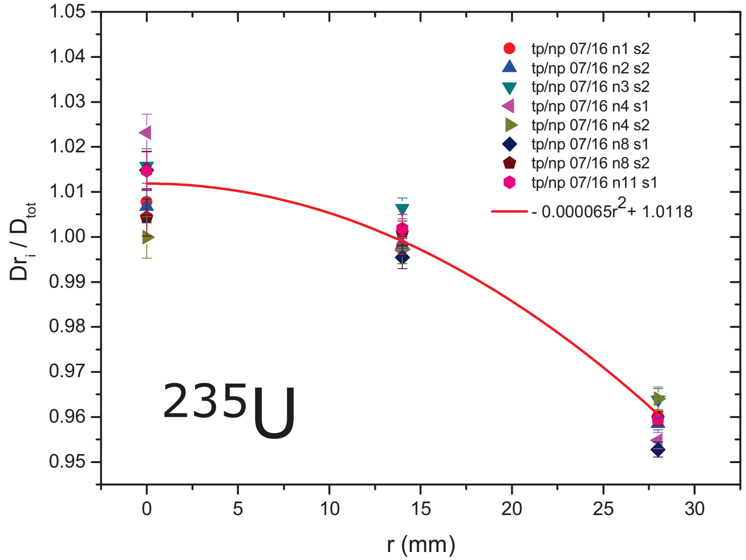
Cross sections

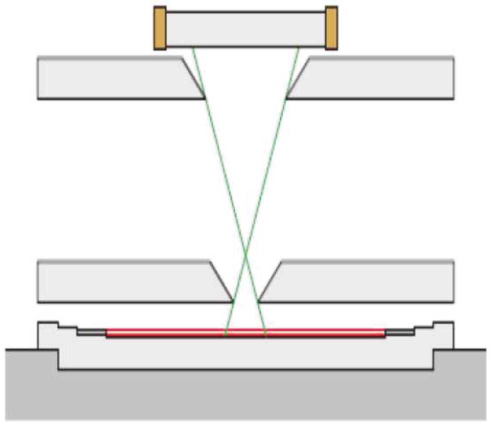
Sample Characterisation, activity and homogeneity

Inhomogeneity: +2% areal mass density
61 mm out of 70 mm









- **Defined solid angle counting (absolute)**
- **Homogeneity survey with alpha counting**
- **Note new findings by Vascon et al. (Mainz) for lanthanide molecular plating; bearing on actinides? See presentation Eberhardt.**

INTDS 2012, Heyse et al. Sibbens et al.

Cross sections

Characterisation, purity, isotopes, specific activity

Callet& De Bièvre 15 November 1985	Atom%	Acc (2s)
U-234	0.0626	0.0025
U-235	99.8266	0.0044
U-236	0.0365	0.0027
U-238	0.0739	0.0025

Activity to atoms
Mass spectrometry
65% activity: U-234

Quantity	Value (1s)
Decay constant	8.78(10) 10^{-17} U/s

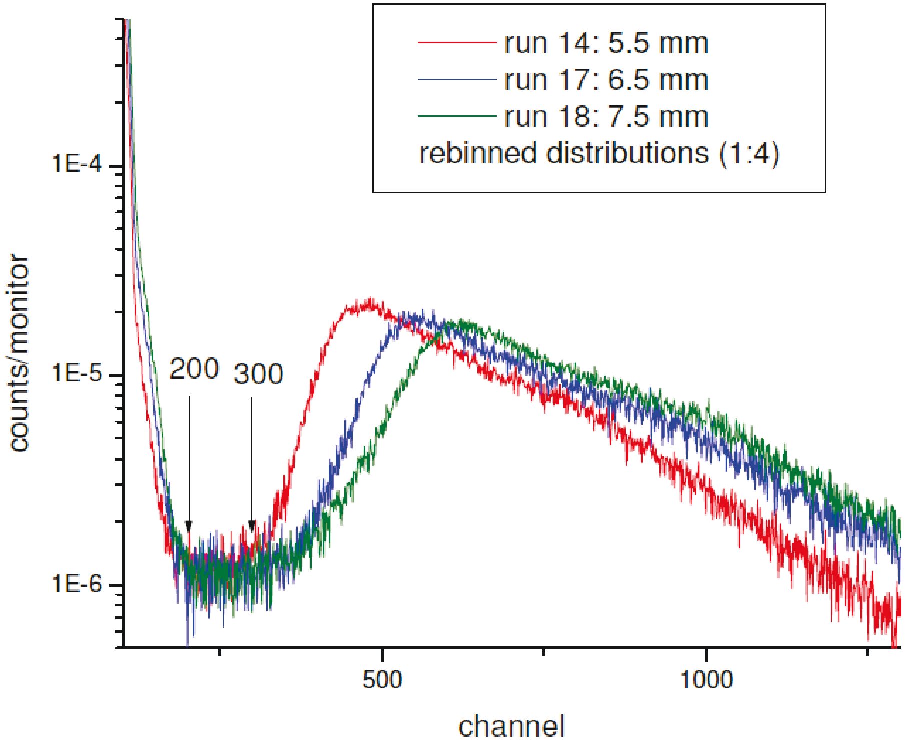
Richter 23 March 2009	Atom%	Acc (2s)
U-234	0.06389	0.00014
U-235	99.82275	0.00020
U-236	0.03768	0.00007
U-238	0.07568	0.00013

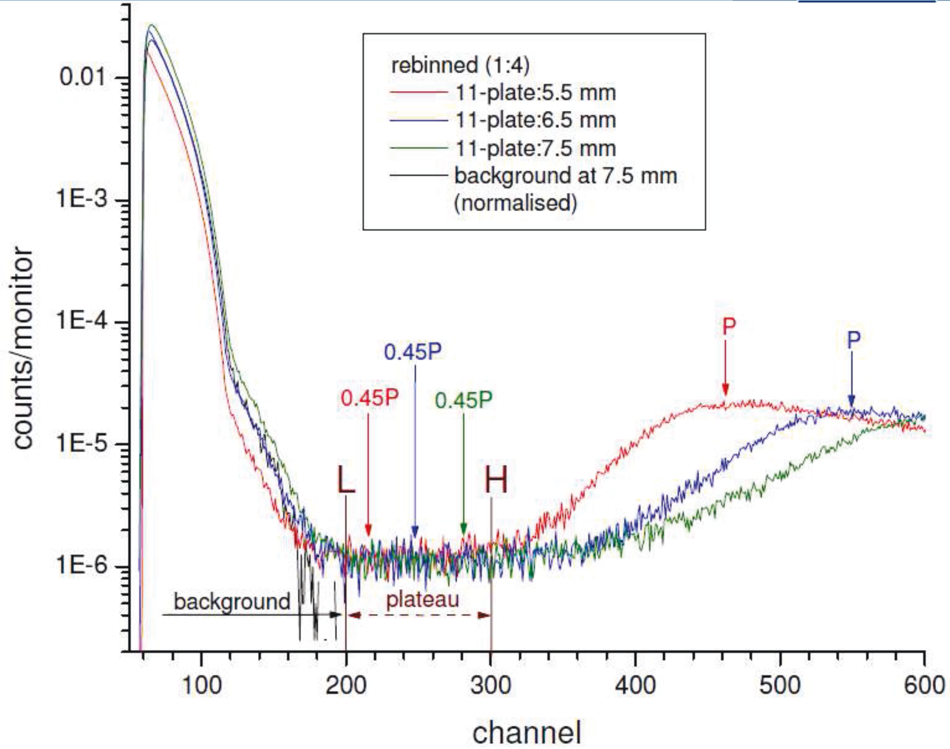
Quantity	Value (1s)
Decay constant	8.88(2) 10^{-17} U/s

More accurate new value: +1% and 0.3% uncertainty

Cross sections

Fission ionization chambers, pulse heights, selectivity, efficiency





Separation of alpha-particle+noise from fission fragments

Threshold criterium:

1) negligible/small alphas
2) small nr. of FF below threshold
3) calculable

gap	below-threshold	below+above
mm	%	1000*counts/monitor
5.5	3.4(1)	8.36(2)
6.5	3.7(1)	8.35(2)
7.5	4.1(1)	8.35(2)

- A larger gap facilitates 1).
- The fraction lost is small (475 µg/cm2 ^{235}U; form UF_4)
- The fraction lost is very calulable (to 0.25%).

Cross sections

Fission ionization chambers, expected behaviour at low energy

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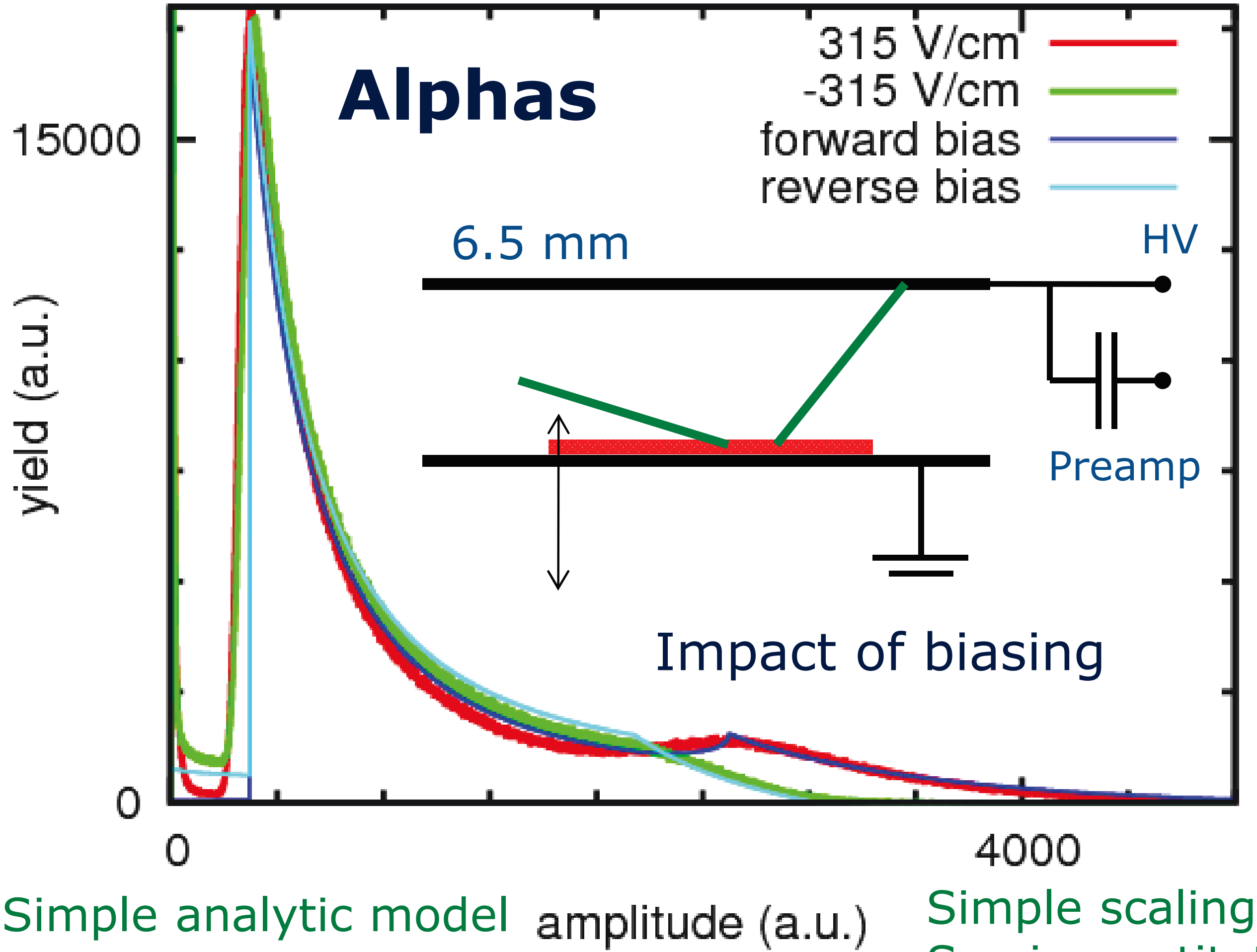
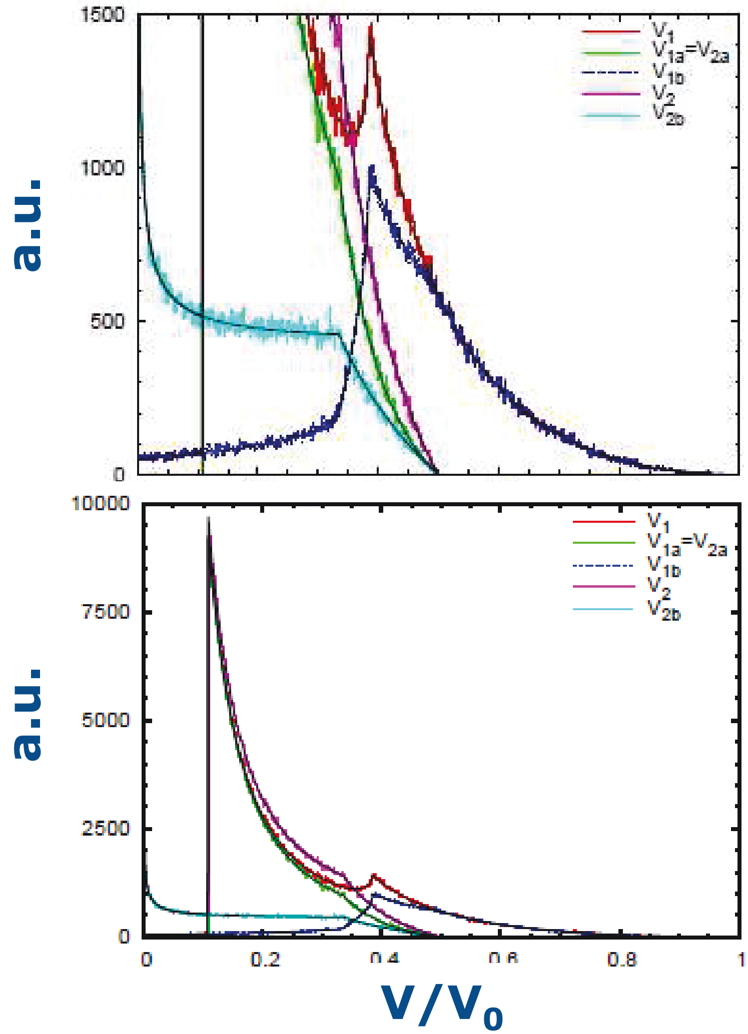
Parallel plate measured activitiy: 26.6(2) kBq (2009)
Defined opening angle counting: 26.88(5) kBq (2001)





Simple analytic model
One energy, one range
Uniform charge deposition

Simple scaling
Semi-quantitative agreement

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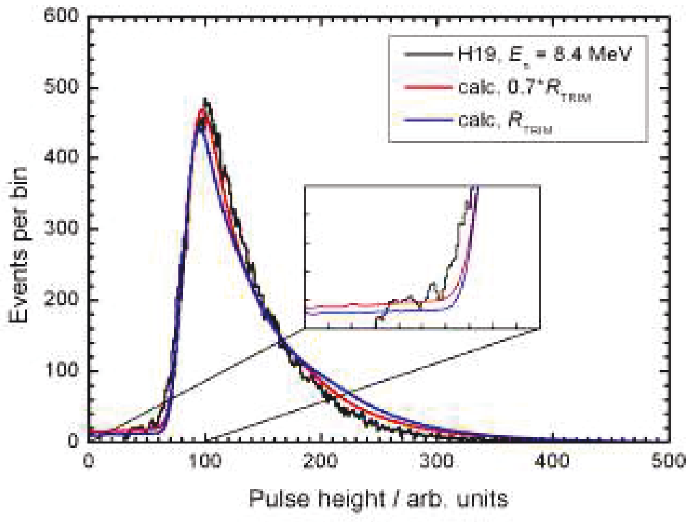
Cross sections

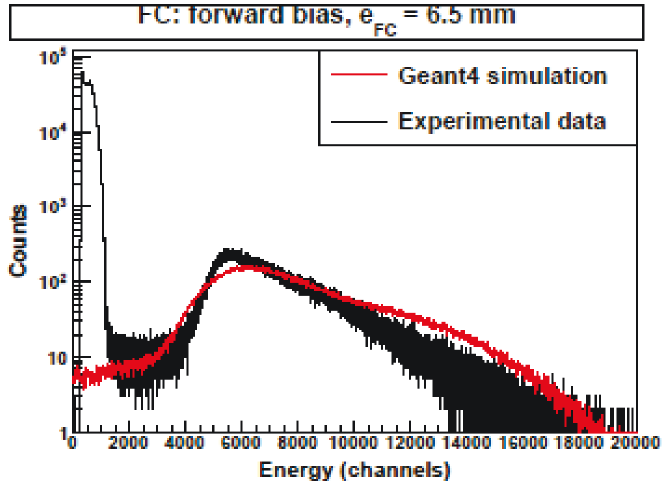
Ionization chambers: modeling the PH distribution

- **FIC U-235**



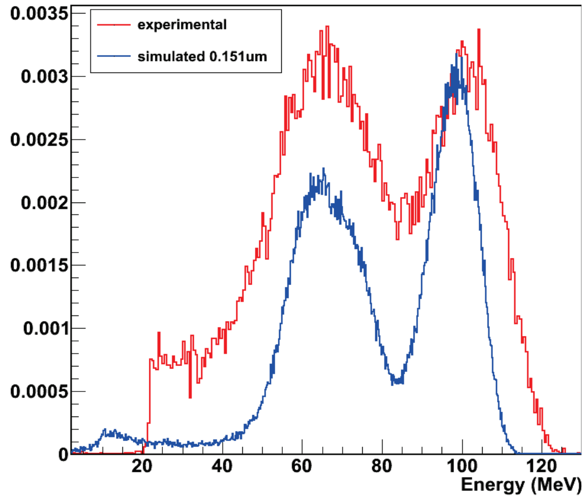
Mosconi et al. CERN EFNUDAT workshop 2010

FIC U-235



Thiry, PhD thesis, Strasbourg 2010

Frisch-grid

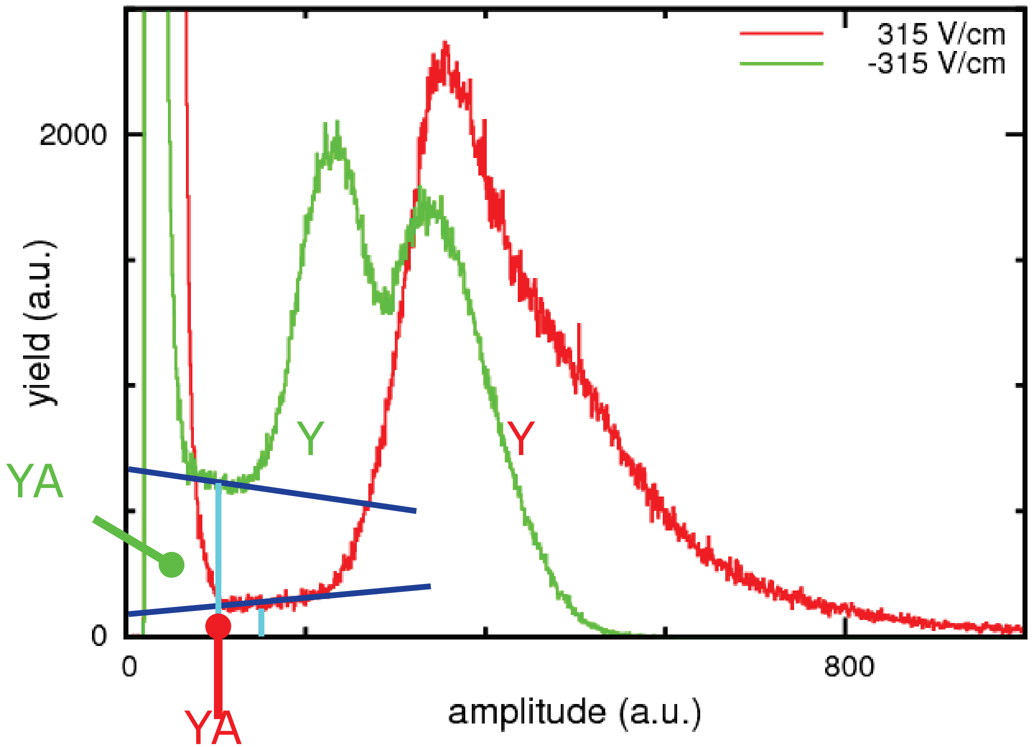


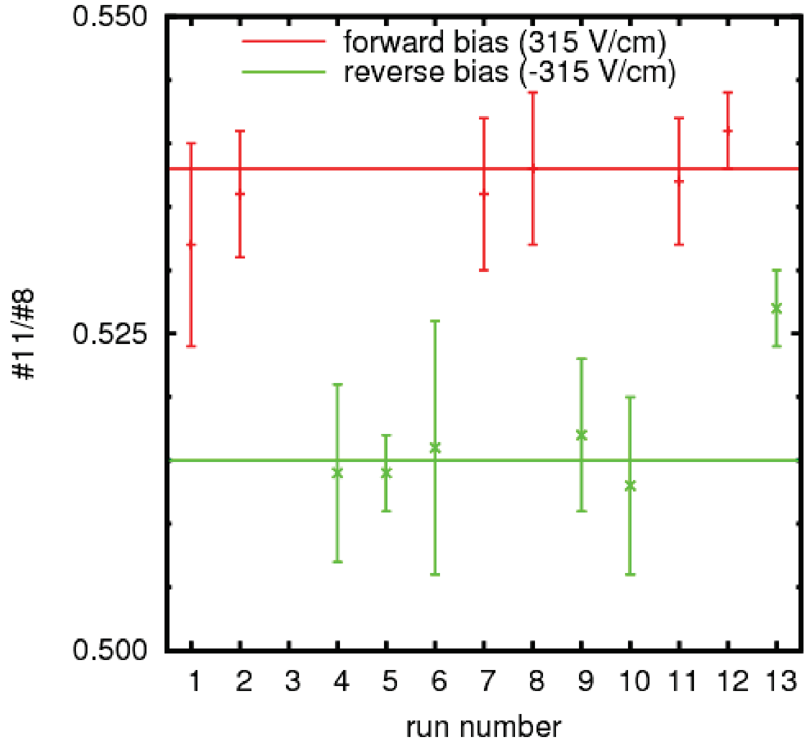
Salvador-Castineira, Theory-2, 2012

Here is where the impact of surface roughness and elemental composition need further investigation

Cross sections

Fission ionization chambers, forward and reverse bias





Efficiency = $Y / (Y+Y_A+Y_B)$

$Y+Y_A$ forward versus backward bias

4.4% effect for 0.475 mg/cm² ^{235}U in the form of evaporated UF_4

Y: yield above threshold, Y_A: yield below threshold (linear extrapolation)

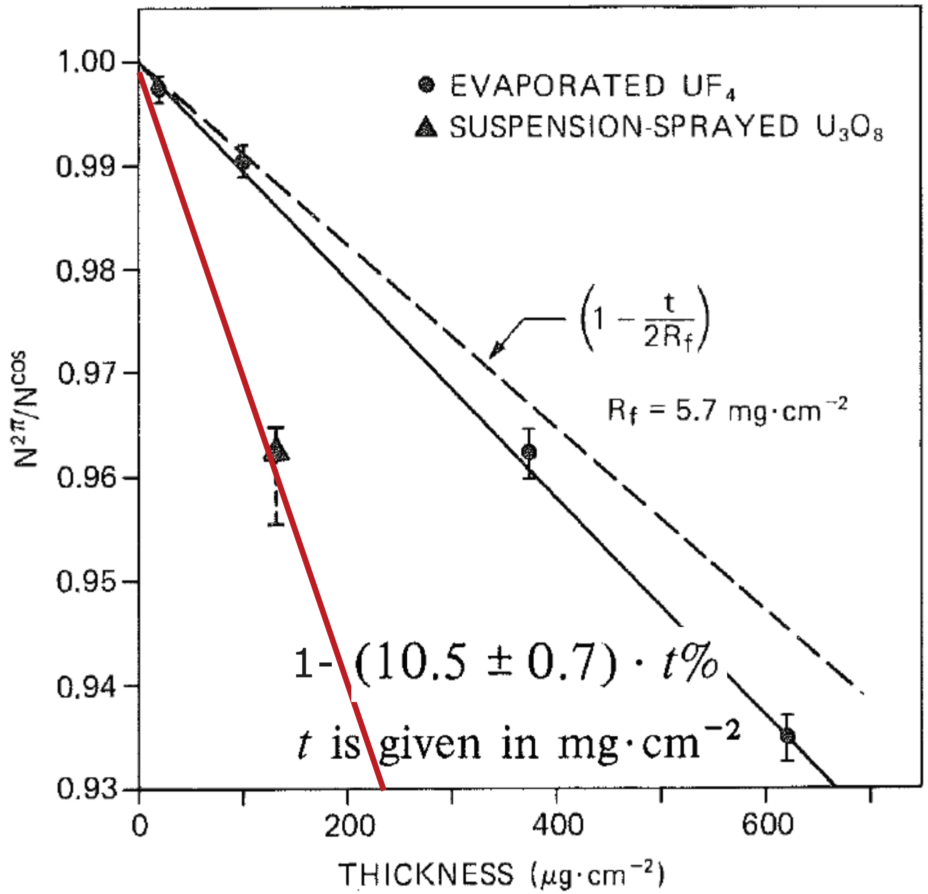
Y_B: fragments stopped in the deposit (not shown)

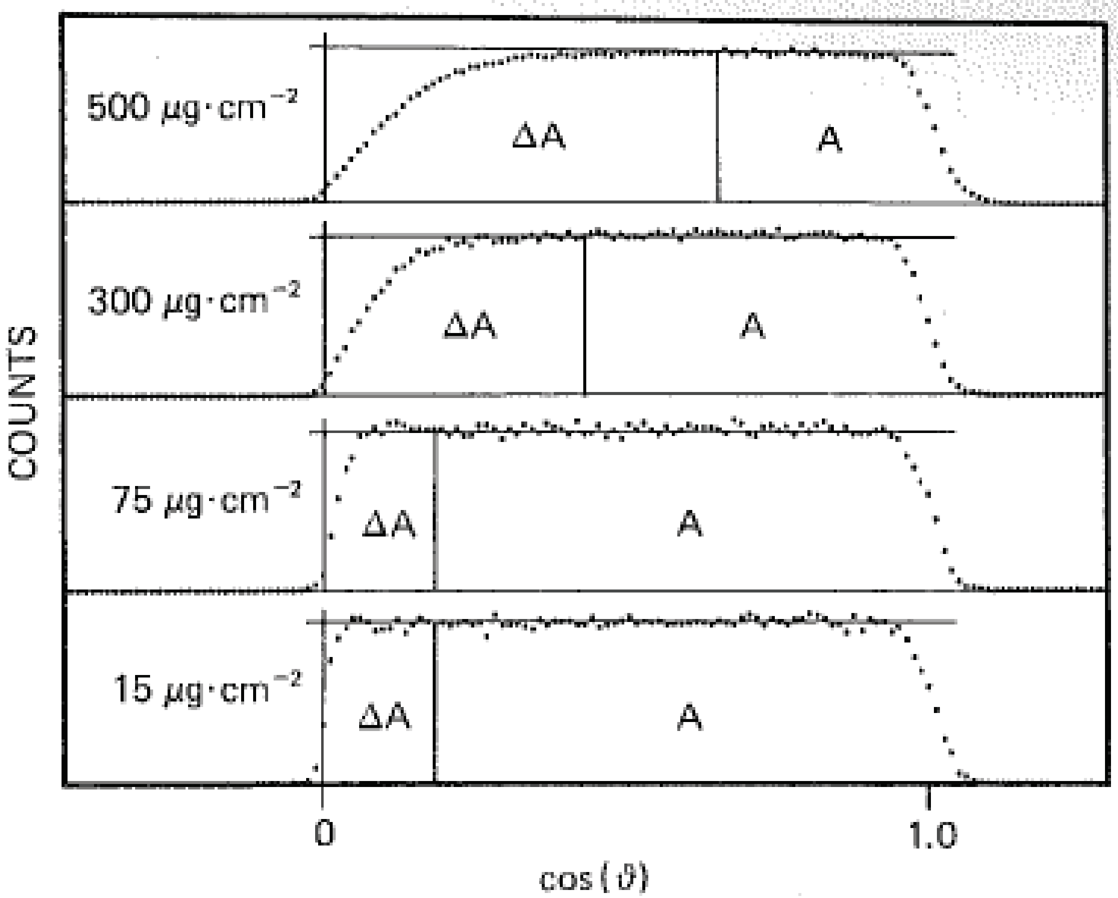
$Y_B/(Y+Y_A+Y_B)$ = 0.105(7) t/(mg/cm2)

Y_B: measured by Budtz-Jörgensen Nucl.Instrum.Meth. 236(1985)630

Cross sections

Loss in the deposit; FIC, Frisch-grids, PPAC, μ-pattern





Angular distribution measurements with isotropic fission (thermal neutrons, ^{235}U)

Ranges from SRIM2008 are too large! Too little loss in deposit by a 20%.

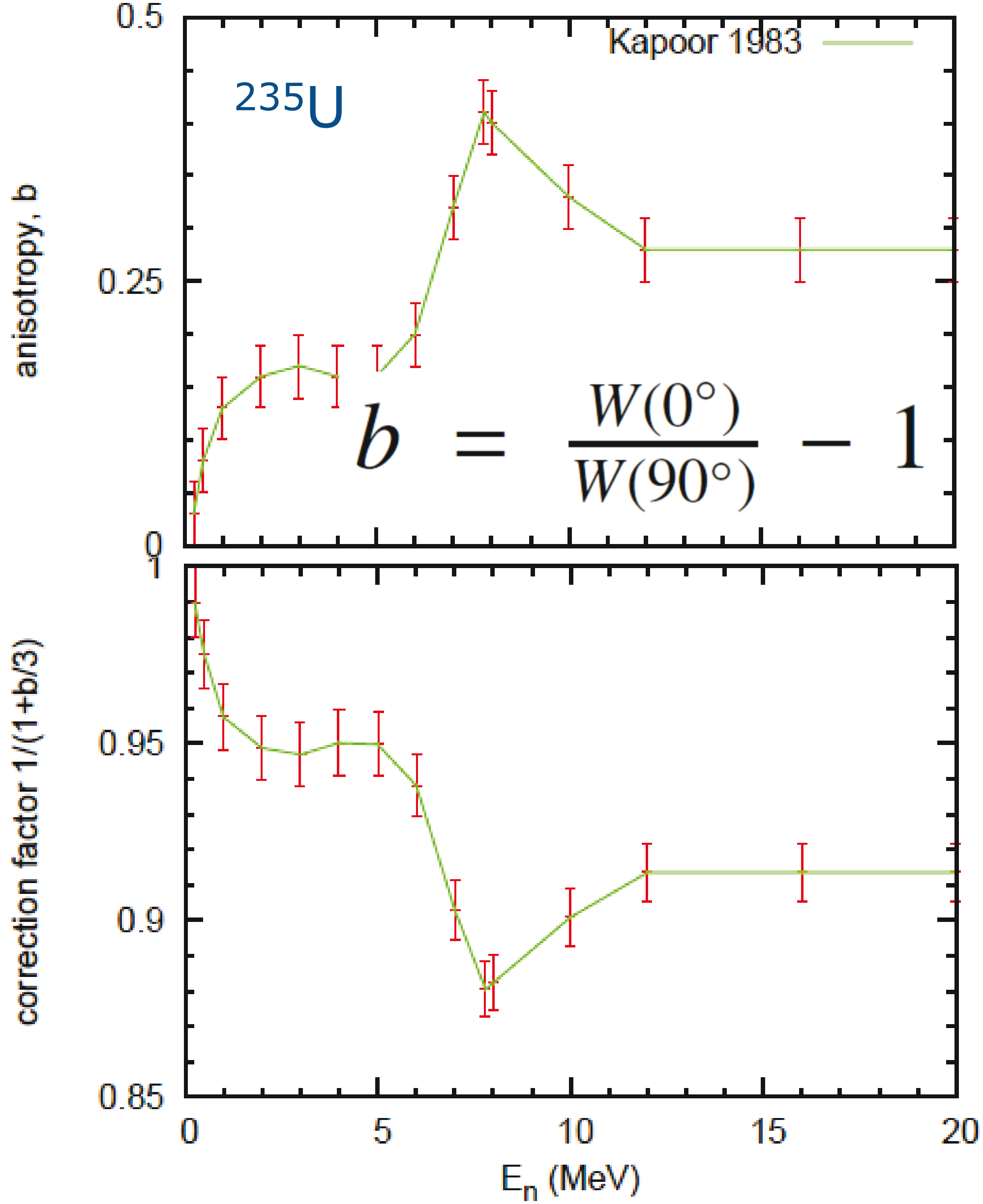
Situation appears much worse for U_3O_8 suspension spraying

What about other methods of preparation?

Budtz-Joergensen et al., NSE86(1984)10

Cross sections

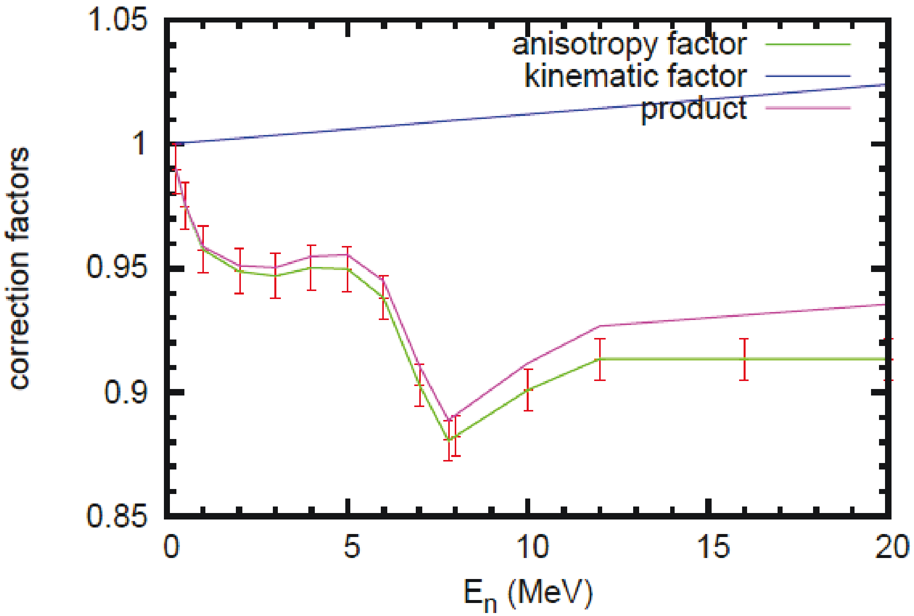
Role of anisotropy

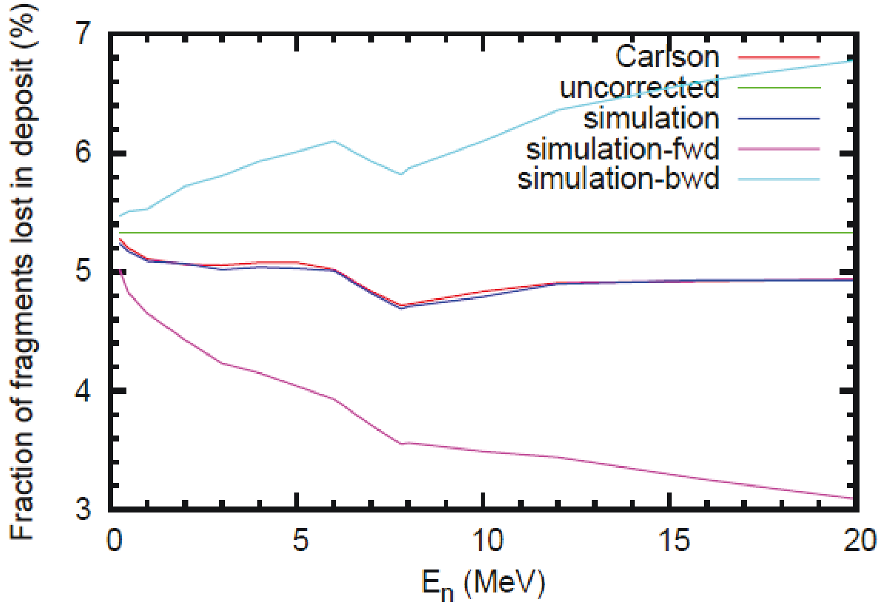


- **Reduced solid angle measurements need to account for anisotropy**
- **Already true for Ionization chambers**
  - **Depends on target thickness**

Cross sections

Role of kinematics





- **Chamber orientation dependent**
- **Orientations averaged: progressively important above 8 MeV**
- **Only one orientation: larger than 1% near 5 MeV**

Plompen et al. J. Kor.Phys.Soc. 59 (2011)1581; EUR 25208 EN (2011)

[1] D. B. Gayther, International intercomparison of fast neutron fluence-rate measurements using fission chamber transfer instruments, Metrologia 27 (1990) 221.

[2] G. W. Carlson, The effect of fission fragment anisotropy on fission-chamber efficiency, Nucl. Instrum. Methods Phys. Res. 119 (1974) 97.

[3] C. Straede, C. Budtz-Jörgensen, H. Knitter, ^{235}U(n,f) fragment mass-, kinetic energy- and angular distributions for incident neutron energies between thermal and 6 MeV, Nucl. Phys. A 462 (1987) 85.

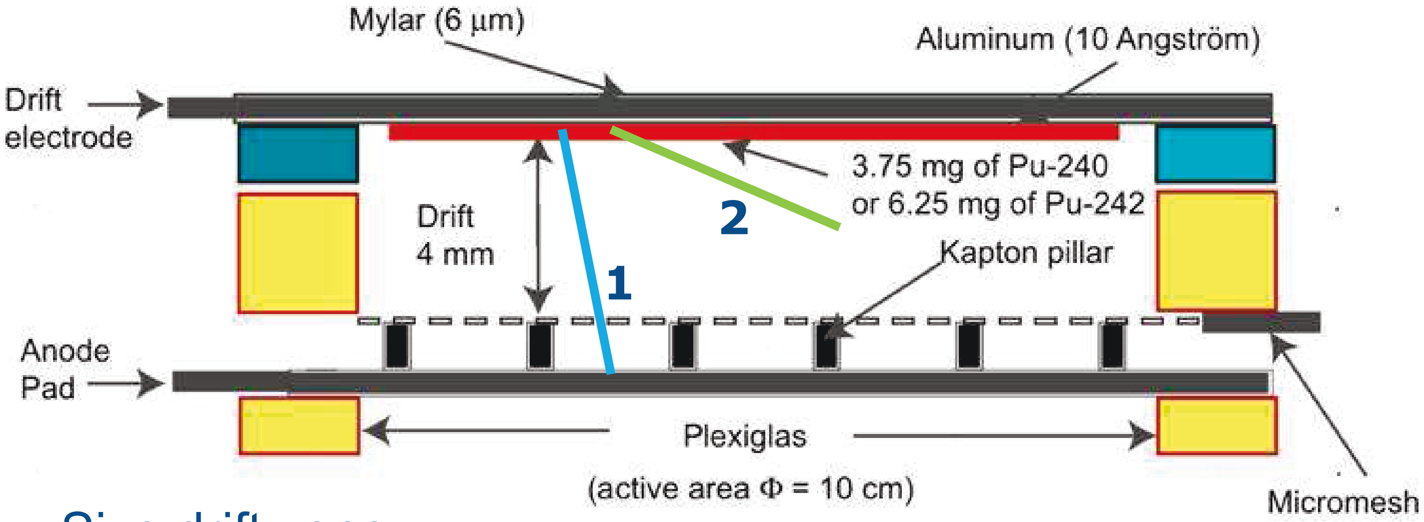
[4] S. S. Kapoor, The ^{235}U fission fragment anisotropies, Nuclear Data Standards for Nuclear Measurements, Technical reports series nr. 227, International Atomic Energy Agency, Vienna, ISBN 92-0-135083-X (1983).

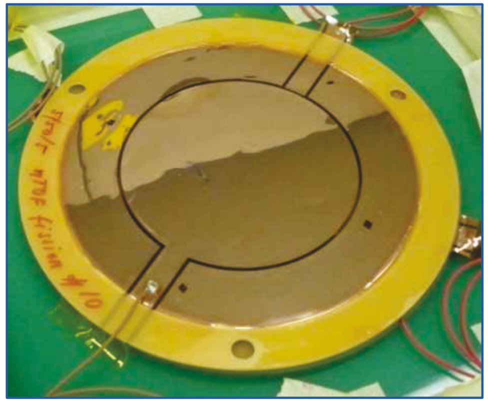
[5] J. F. Ziegler, J. Biersack, M. Ziegler, SRIM The stopping and range of ions in matter, www.srim.org, SRIM Co., Chester, MD 21619, USA, ISBN 0-9654207-1-X (2008).

[6] Budtz-Jörgensen, H. Knitter, Assaying targets for nuclear measurements with a gridded ionization chamber, Nucl. Instrum. Methods Phys. Res. 236 (1985) 630.

Detectors based on proportional amplification

PPAC, Micromegas, GEM

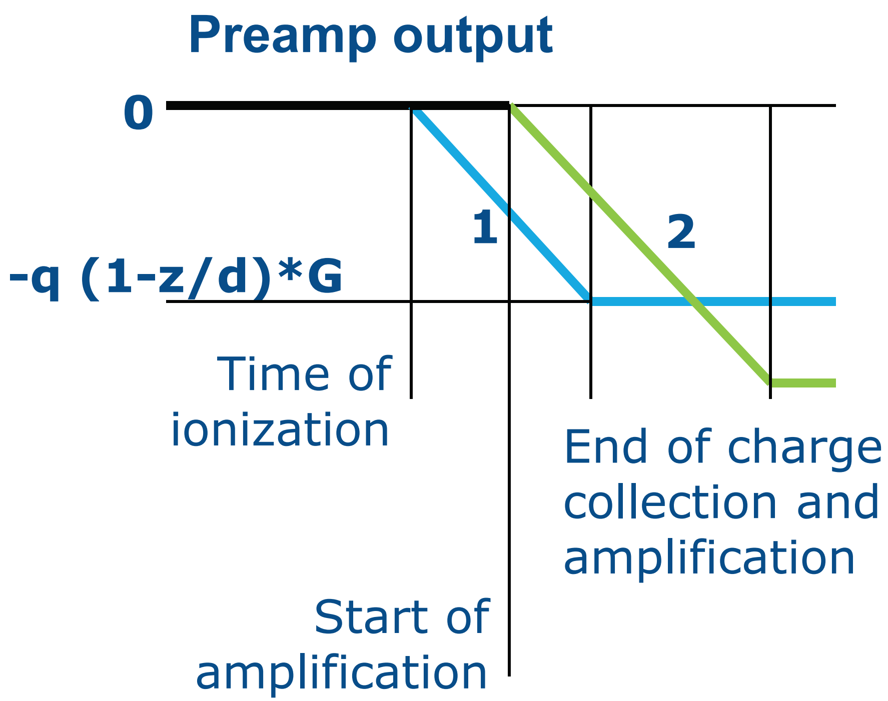




Size drift zone
determines minimum
charge deposited

High field zone
Charge is amplified
between mesh and
anode; gain G

Induced charge is
negligible (below
electronics threshold)



For good timing the
drift zone must be as
small as possible

Pictures: A. Tzinganis
ERINDA workshop JYU 2013

New approaches to fission cross sections

Solid state detectors and normalization to H(n,n)H

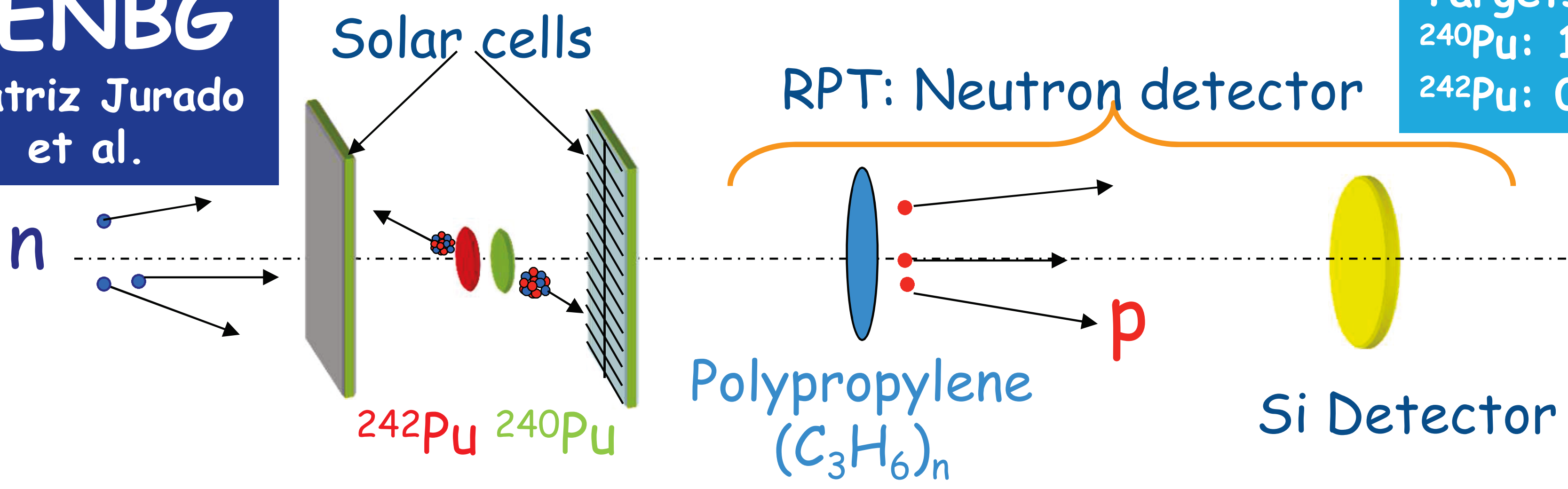
CENBG

Beatriz Jurado et al.

Targets from IRMM

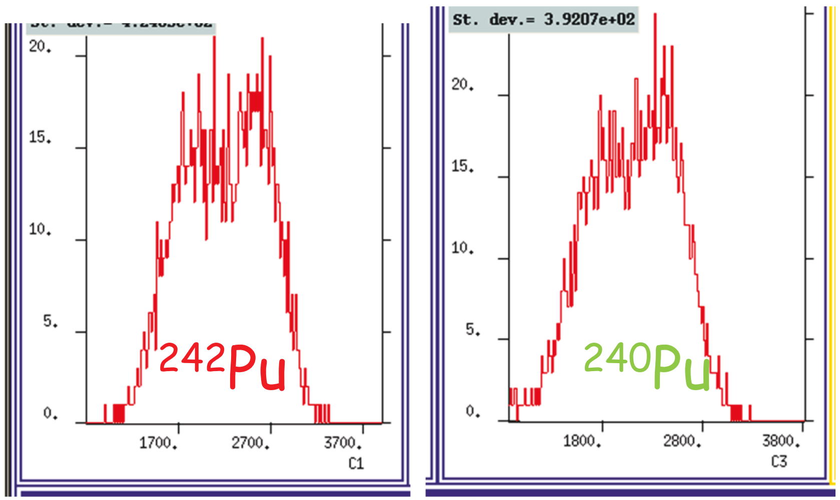
^{240}Pu: 14 Mbq (1.7 mg)

^{242}Pu: 0.2 Mbq (1.7 mg)

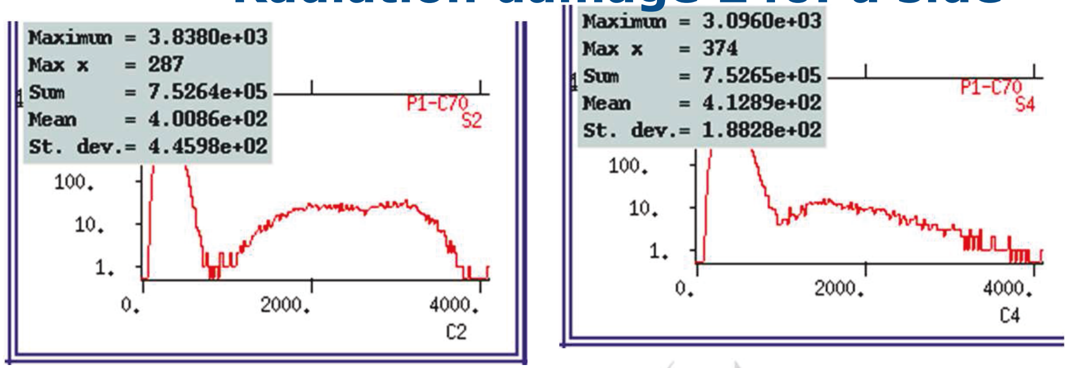


First run 6-14 December 2012 at Bruyeres le Chatel

Start of experiment
Very good α-FF separation



After experiment
Radiation damage 240Pu side



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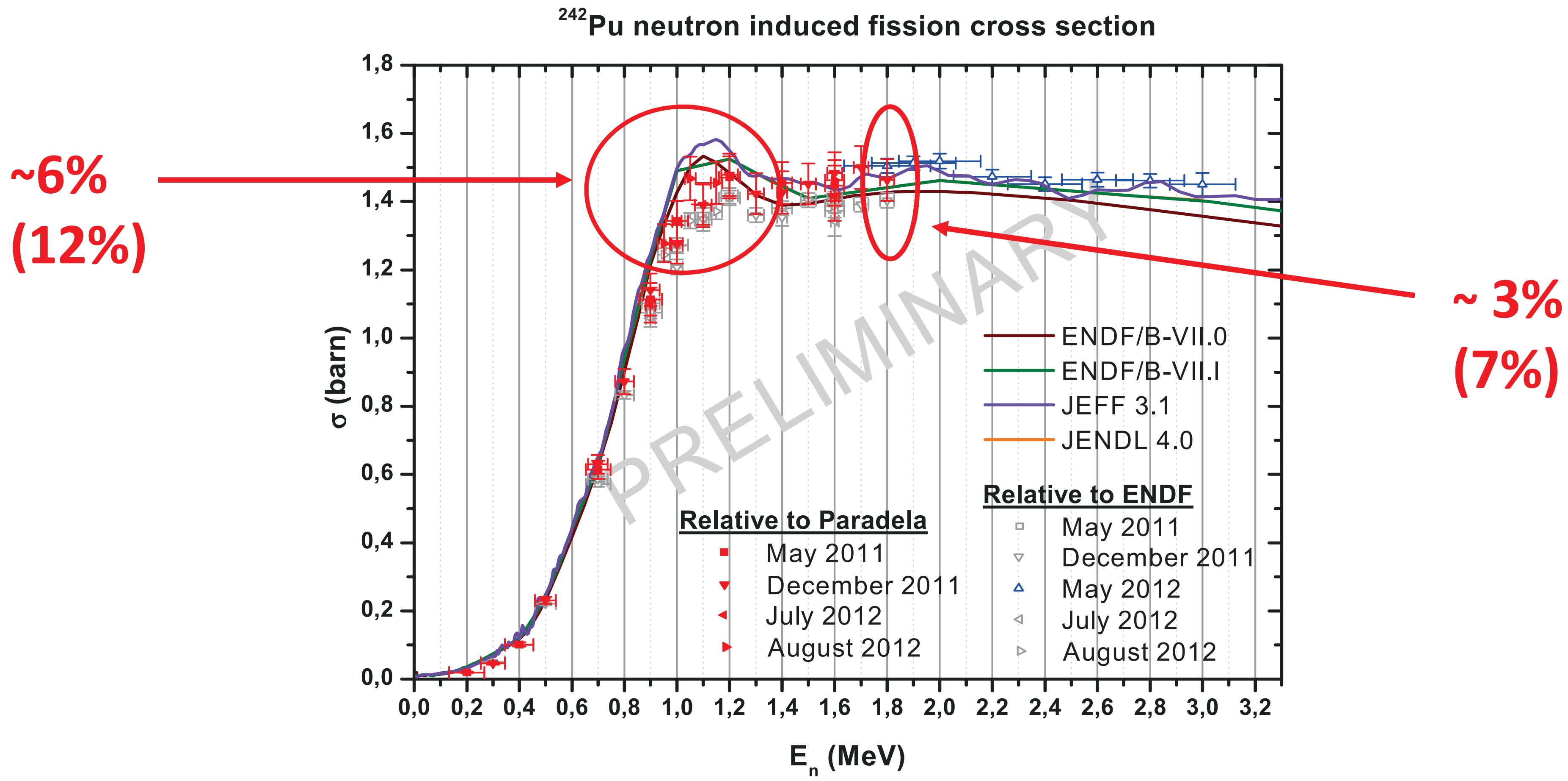
New approaches to fission cross sections

Frisch-grid and ^{237}Np and ^{238}U as reference

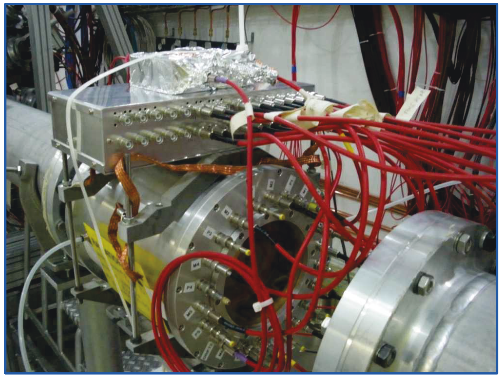
Poster session: Paula Salvador Castiñeira et al.

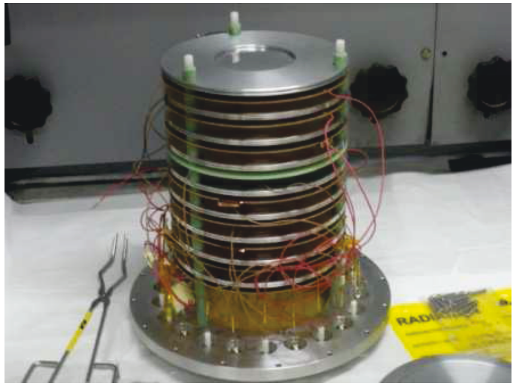
Neutron induced fission cross section ^{242}Pu

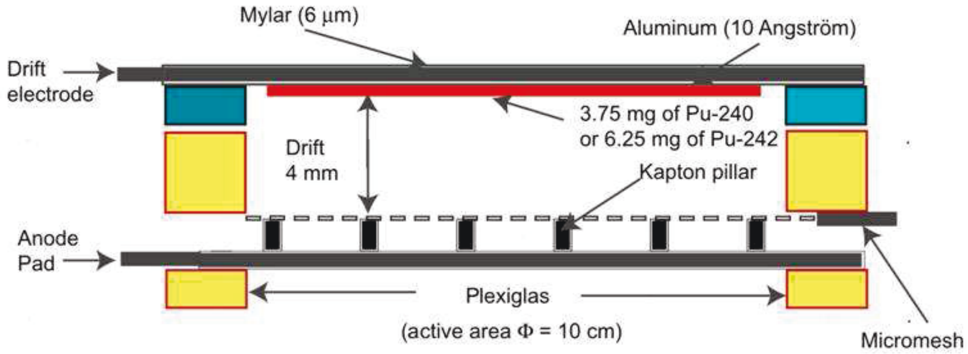


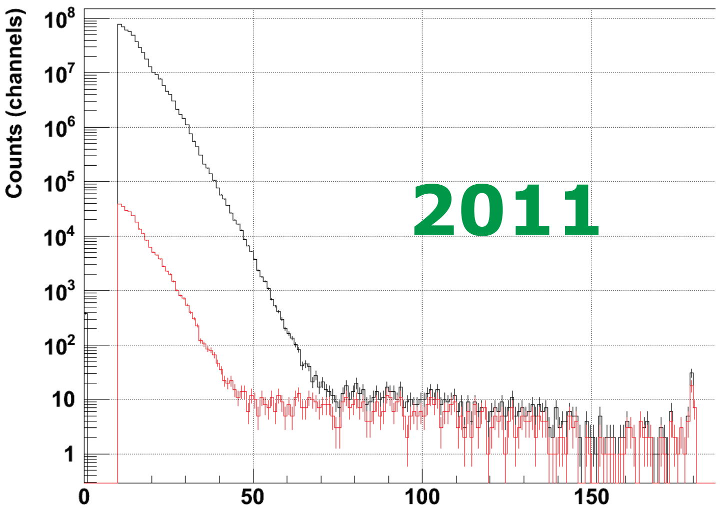
New approaches to fission cross sections

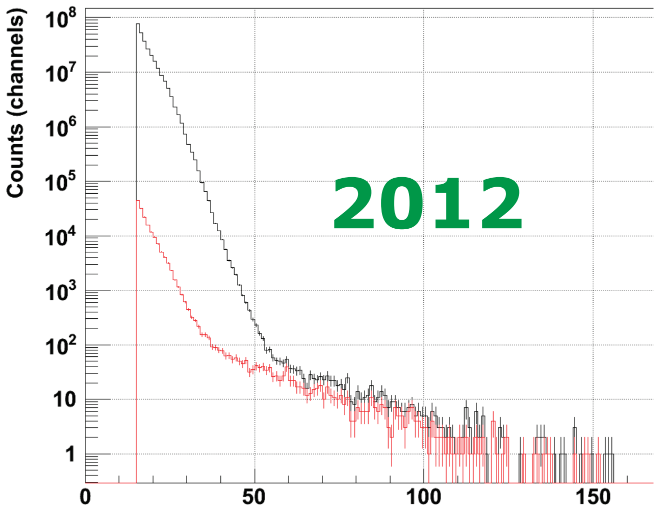
Micromegas detectors, relative to ^{235}U(n,f)







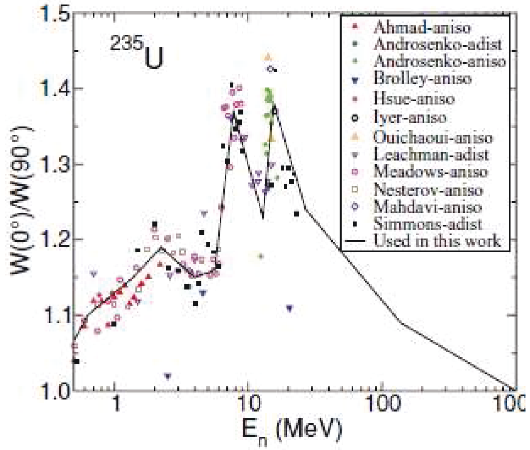


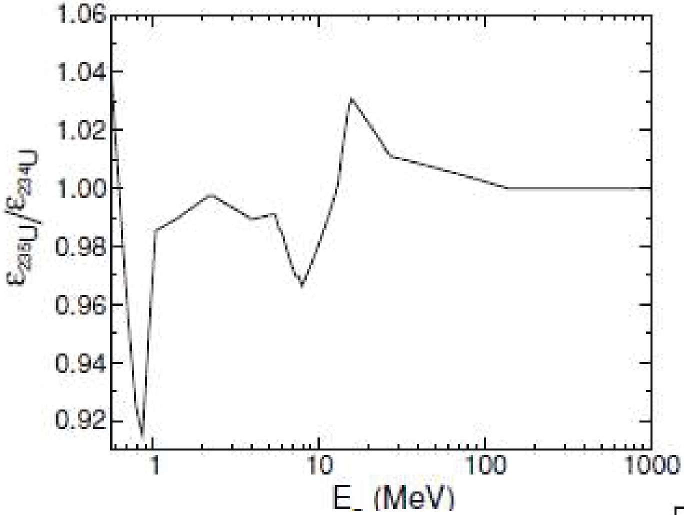


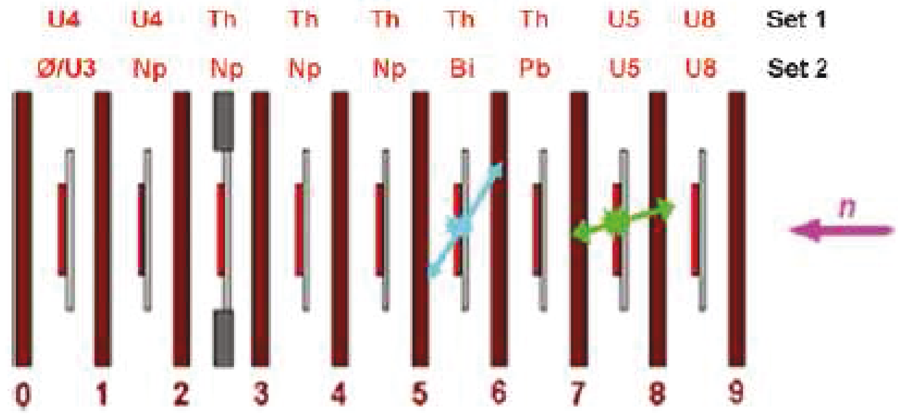
A. Tsinganis (NTUA), et al., 2nd ERINDA Progress Meeting Jyvaskyla, January 8-11, 2013

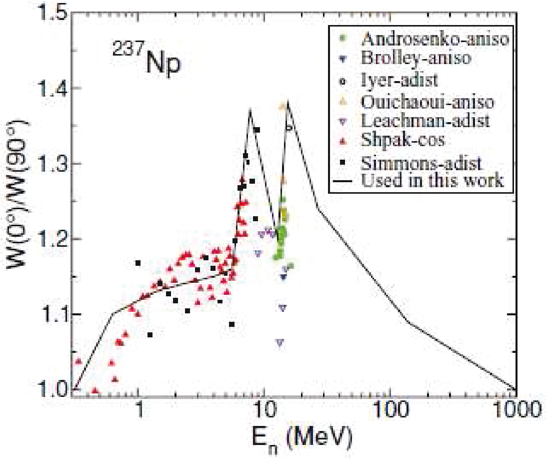
New approaches to fission cross sections

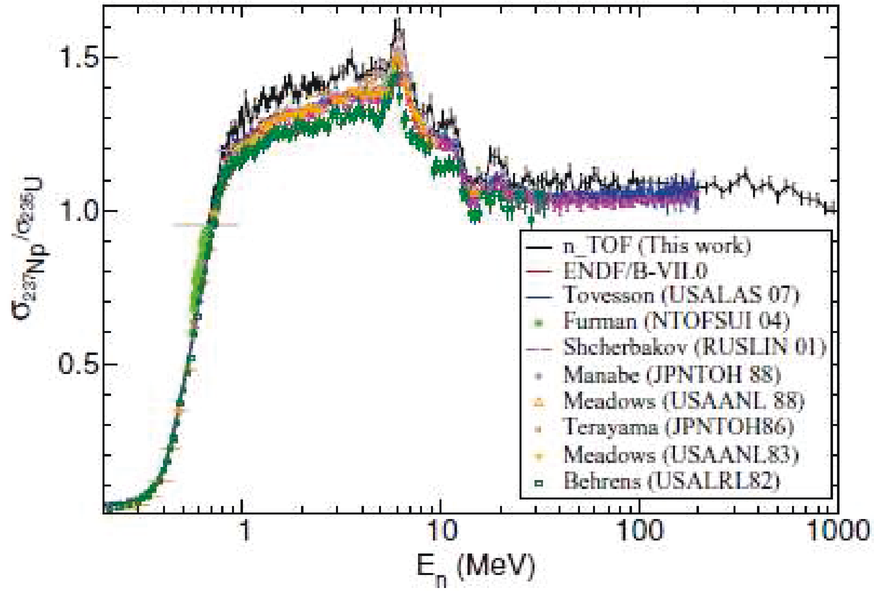
PPAC relative to ^{235}U(n,f); See Audouin and Leong









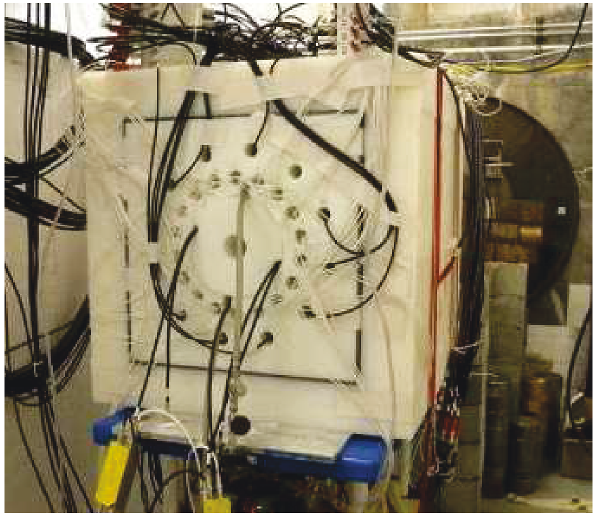


Paradela et al. PRC82(2010)034601

Neutron multiplicity

- **Number of neutrons per fission event**
- **Fission trigger**
- **Emitted neutron number**
  - Low efficiency detector
  - **High efficiency detector (CEA@LANL)**

  T. Granier, JEFF-GEDEPEON workshop 2011



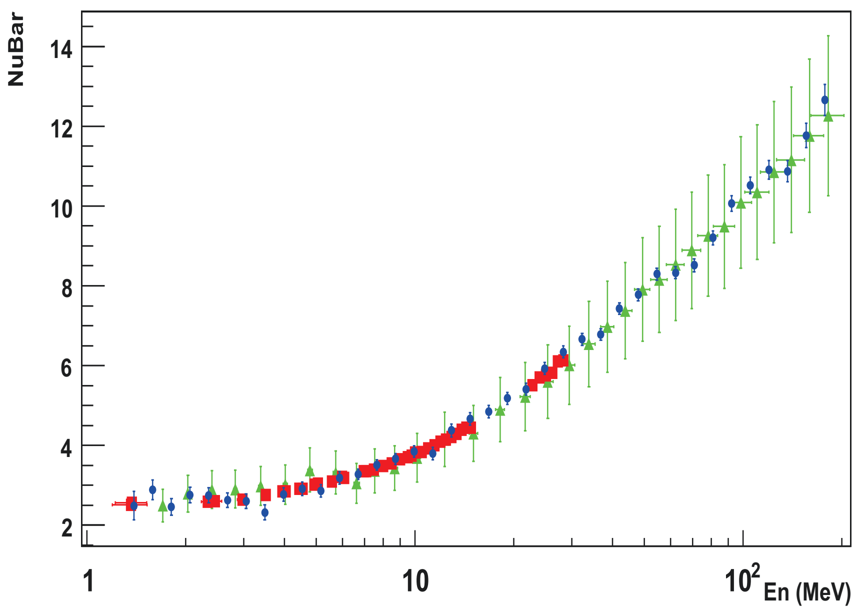
FC 1 mg ^{238}U

30 ^{3}He counters in PE moderator

PE+^{10}B shield block room return

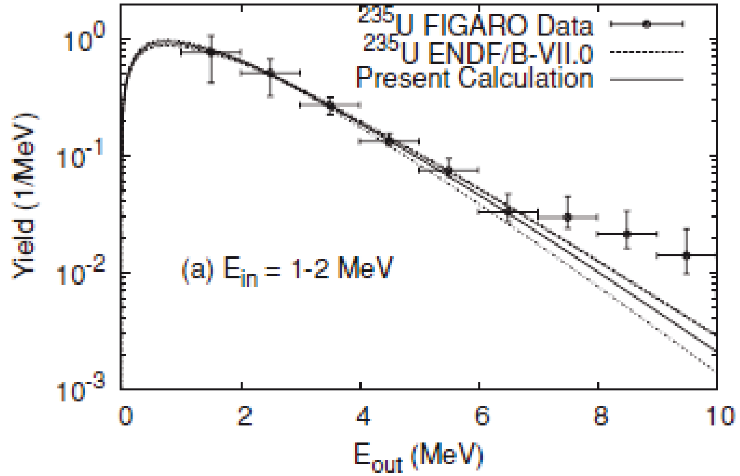
Neutron pulses up to 150 μs

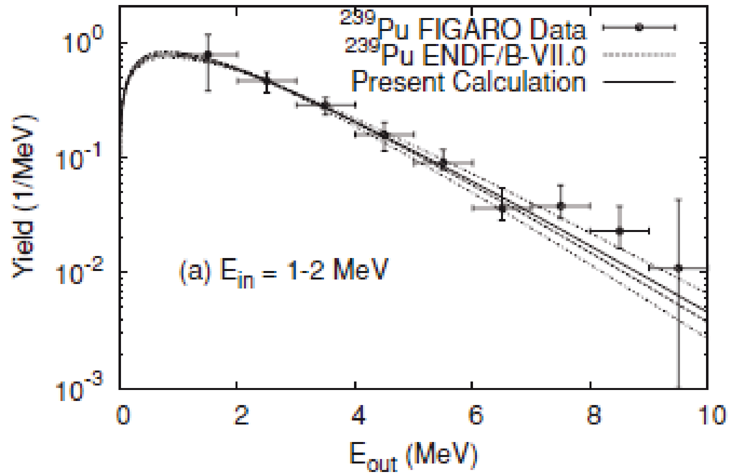
Calibration by $^{252}Cf(sf)$ (eff ≈ 25%)



Prompt fission neutron spectrum

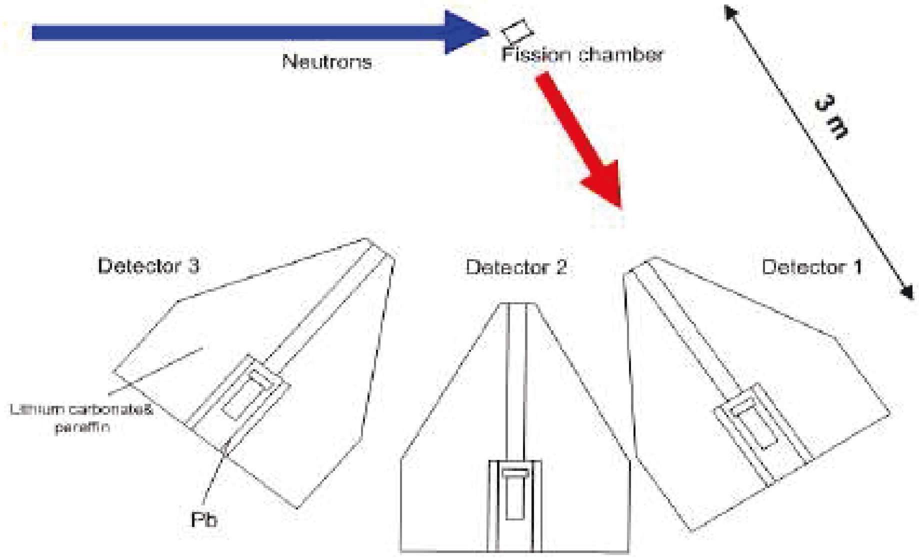
- FC: ^{235}U (100 mg), ^{239}Pu (80 mg), 11+11 deposits on 0.125 mm Pt
- LANL 800 MeV p + W, pulsed 200 ps
- E_n 1-8 MeV, 22.74 m FP, Pb+PE filters
- Double TOF: E_n and E_n'
- FC – 20 liq.scintillators 12.5x5 cm

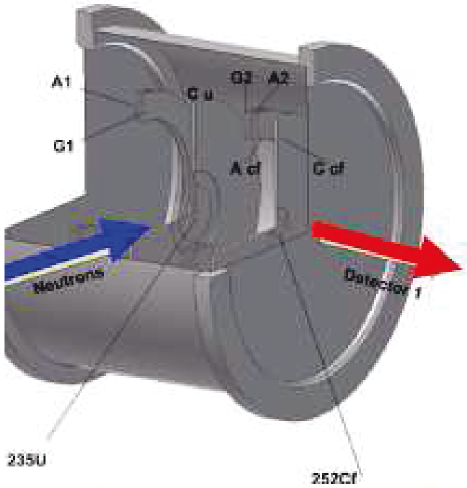


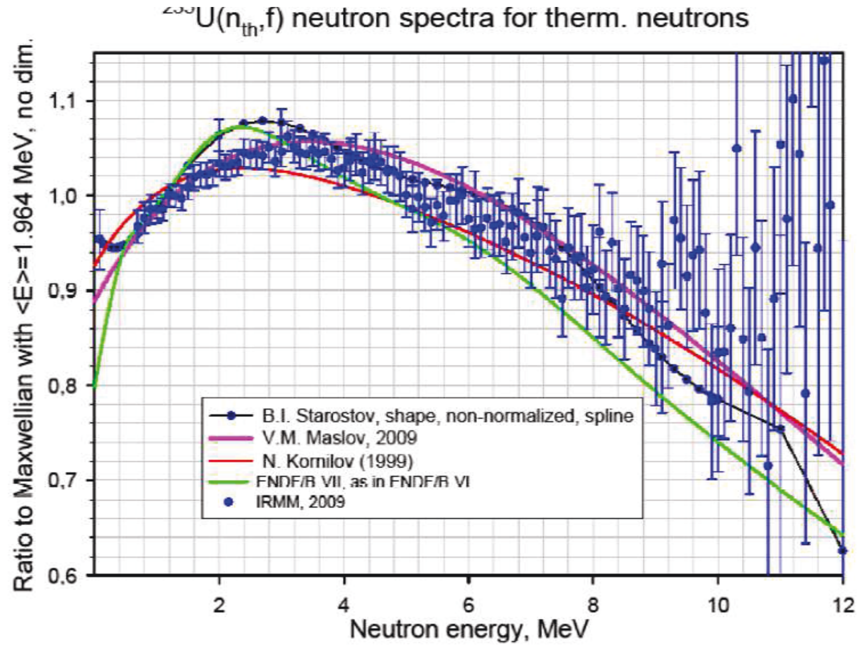


European Commission

Prompt fission neutron spectrum







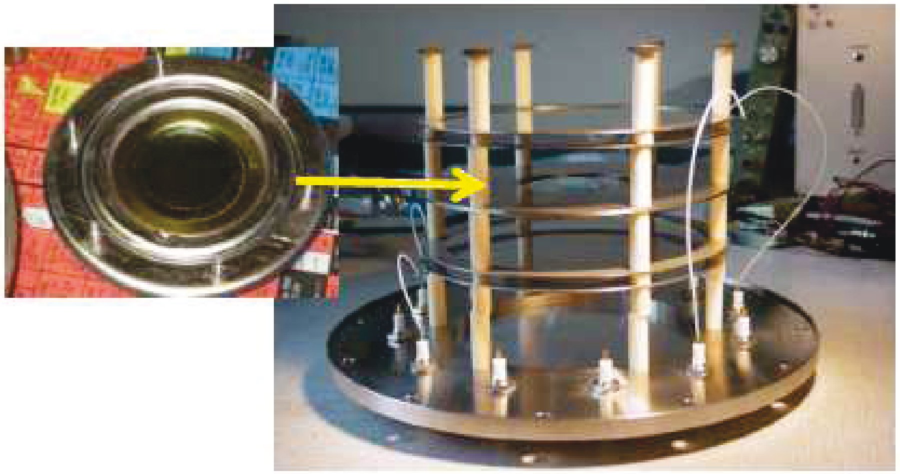
Jefdoc-1388 Hambsch et al. (2011)

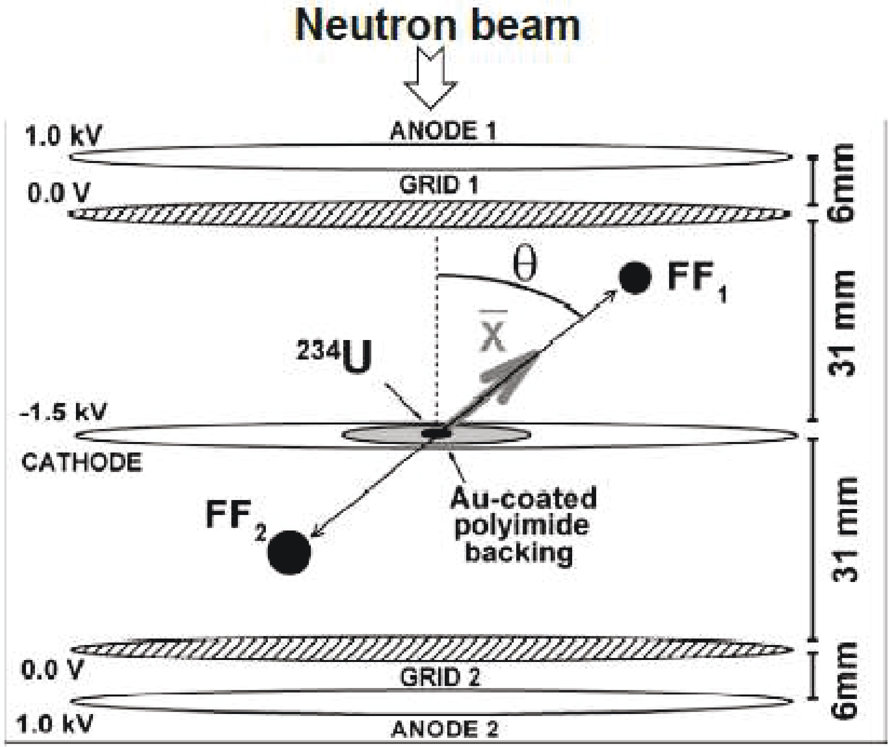
Hambsch et al.
Budapest
reactor beam
Liquid scintillator
LS301, 4”x2”
Frisch-grids, TOF
^{235}U, 112 μg/cm^2
^{252}Cf(sf) efficiency
97.7% enr
FF eff. 98%

Changes imply significant reactivity impacts: -300 to 800 pcm

FF mass, kinetic energy and angle

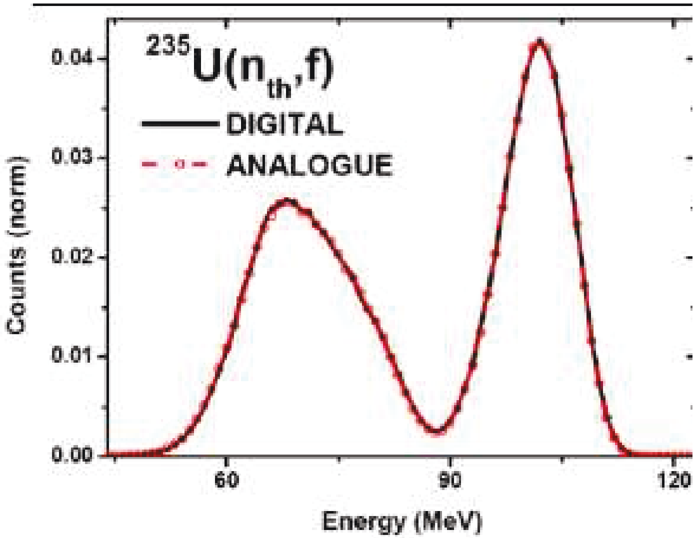
- Energy and angle are measured for both FF (summing and drift-time methods).
- Double E method used for fragment mass calculations (energy and momentum conservation).
- Precision pulse generator used for signal-drift monitoring.
- Counting gas: P-10 (90%Ar+10% CH_4).
- 1.05 bar gas pressure.
- 0.1 l/min gas flow.

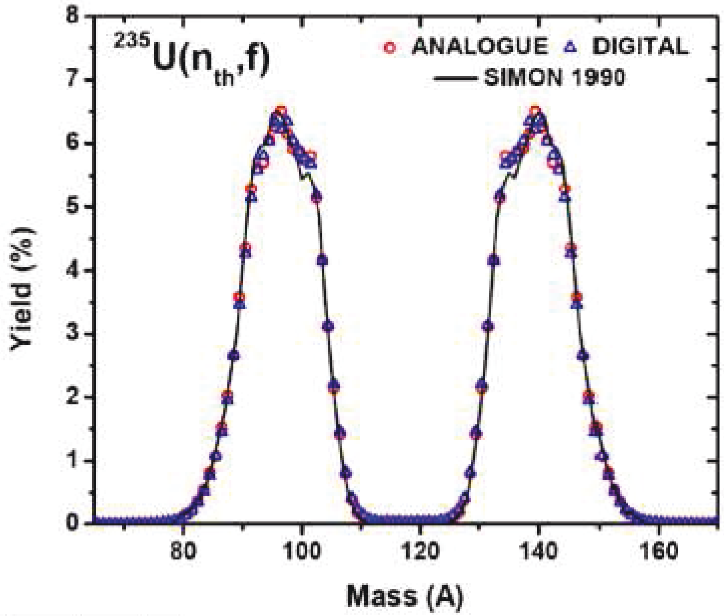


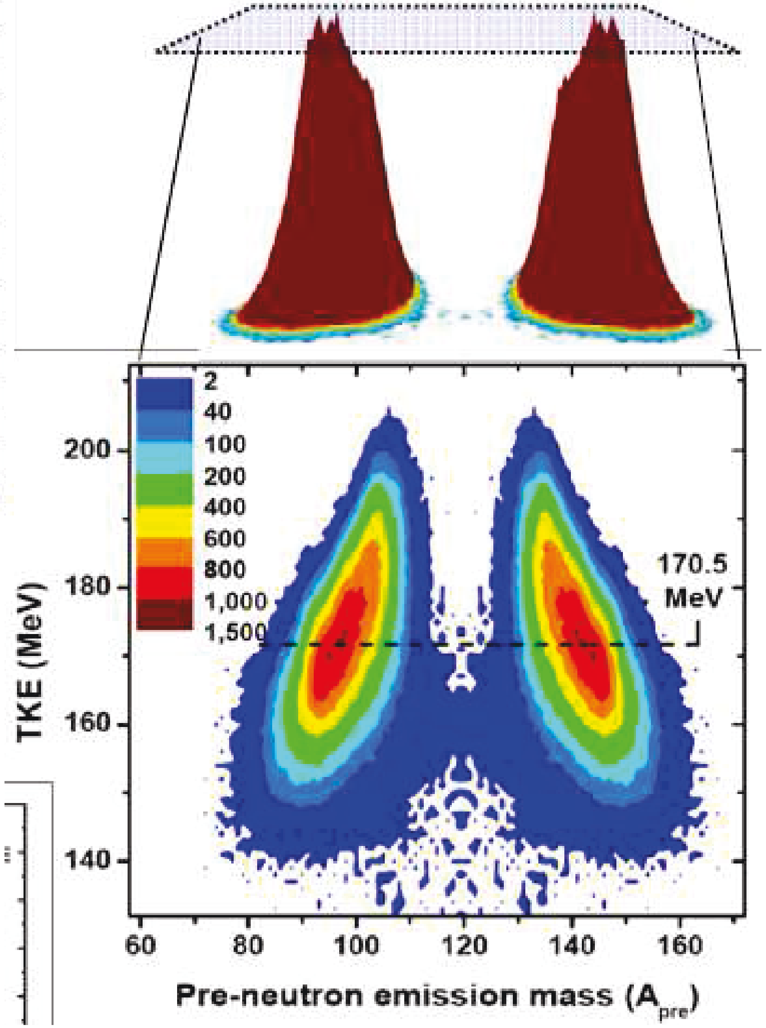


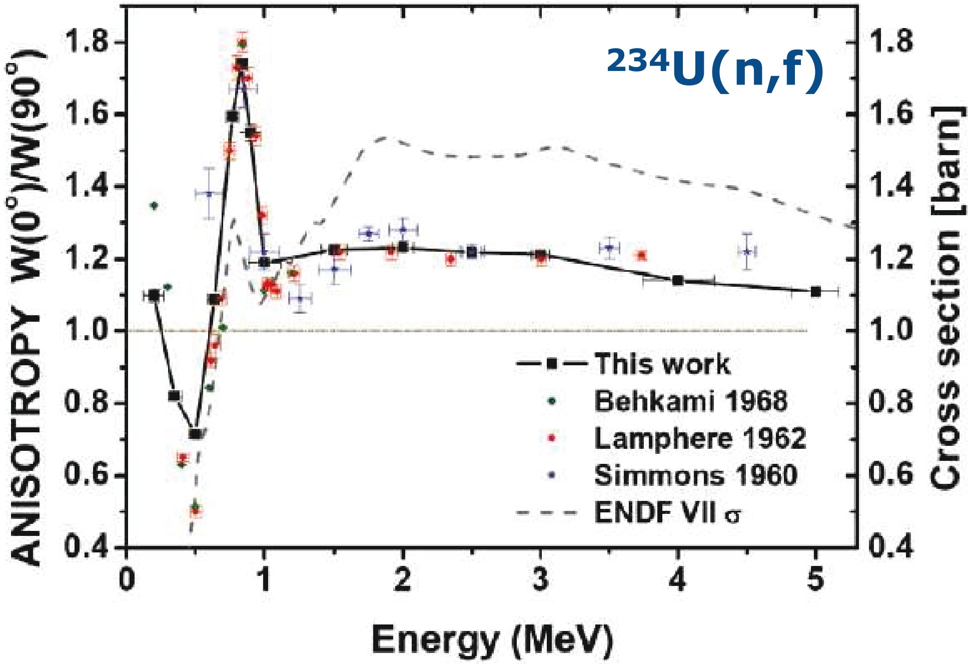
Budtz-Joergensen et al., NIMA258(1987)209
Buneman et al., Can.J.Res.A27(1949)191

FF mass, kinetic energy and angle







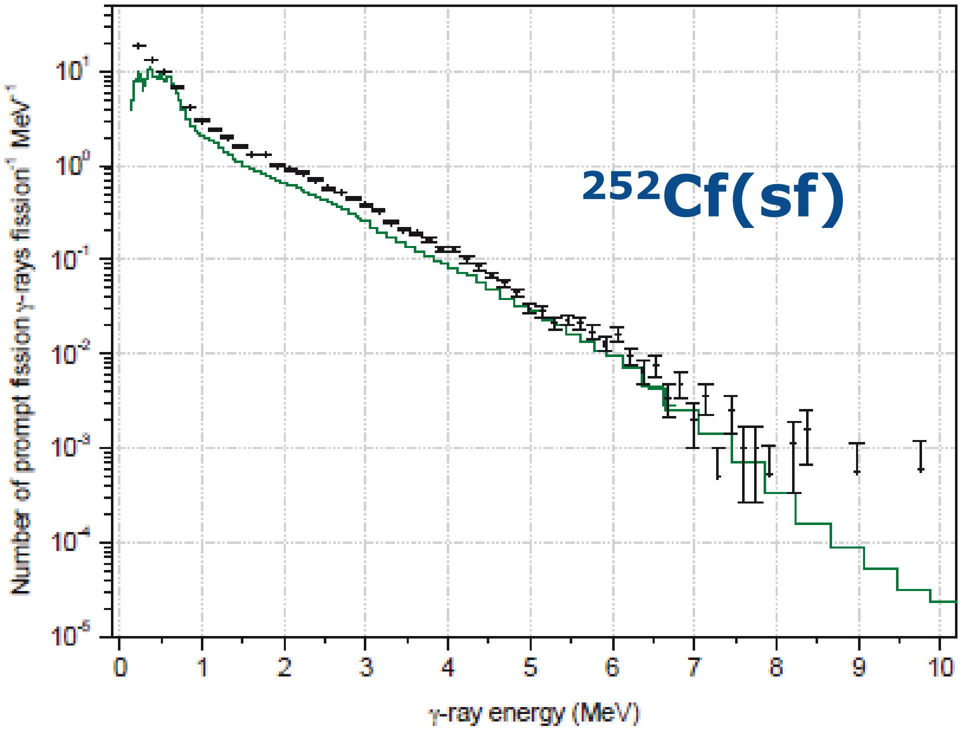


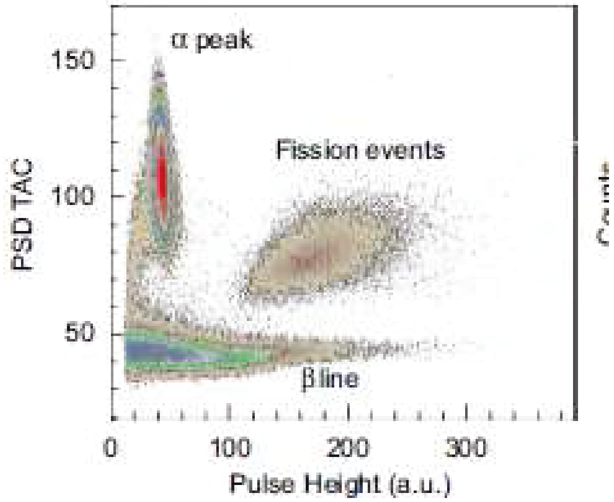
Prompt fission gammas

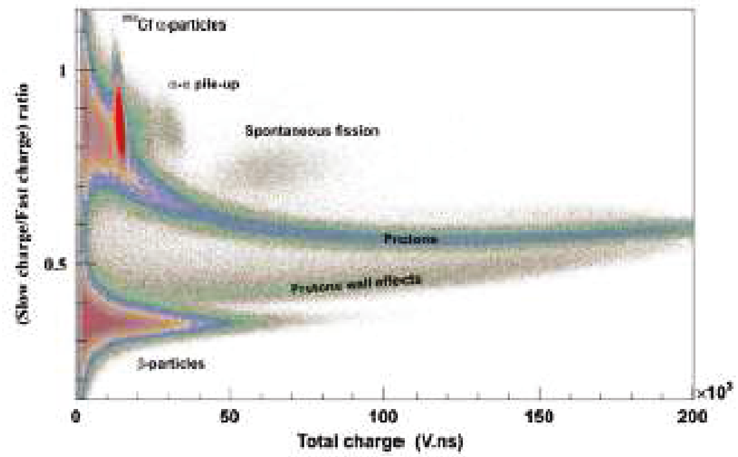
CEA DAM DIF (BRC); van de Graaff (right)

TOF for neutron gamma discrimination

Active-target (FC/LS) and BGO gamma-det.







Liquid scintillator
With target disolved in it

Laborie et al., Phys.Proc.31(2012)13

Bélier et al., Nucl.Instrum.Meth. A664(2012)341

Prompt fission gammas

New gamma-ray detectors: $LaBr_3$, $LaCl_3$, $CeBr_3$

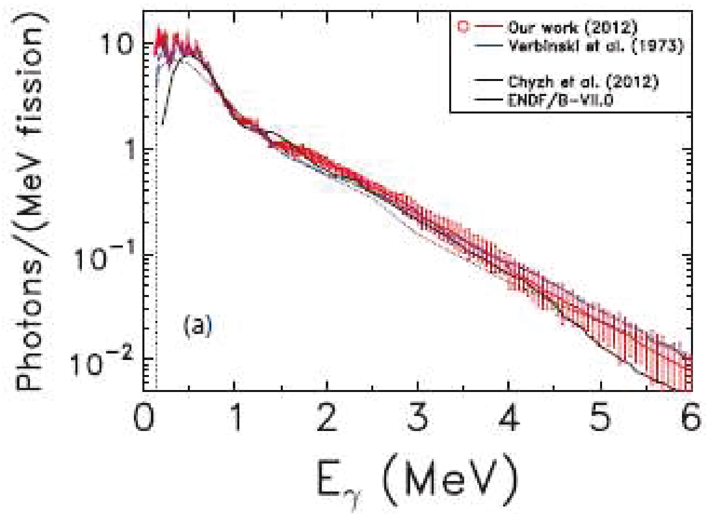
Testing and characterisation

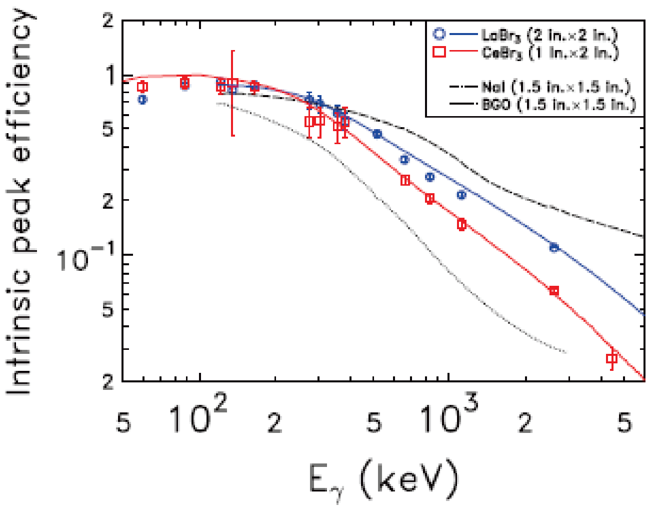
First demonstration ^{252}Cf

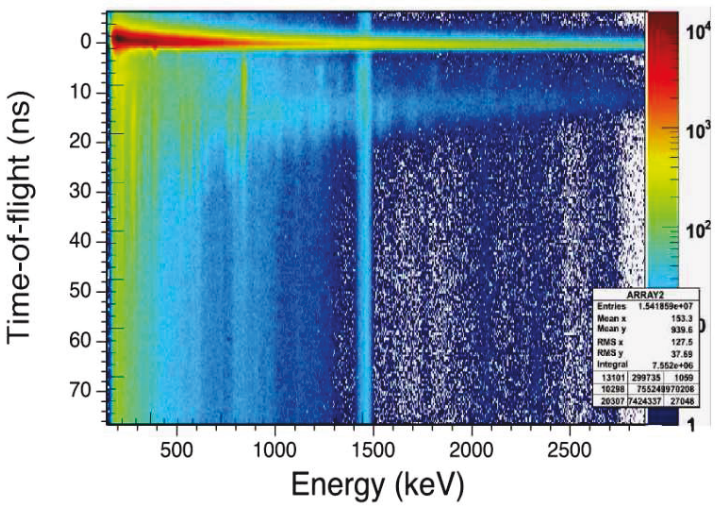
Ongoing/nearly completed ^{235}U

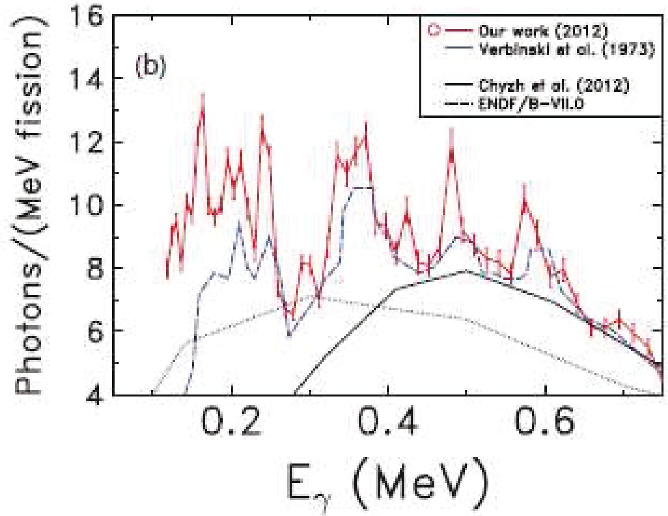
TOF, FIC vs gamma detector

Neutron-gamma separation









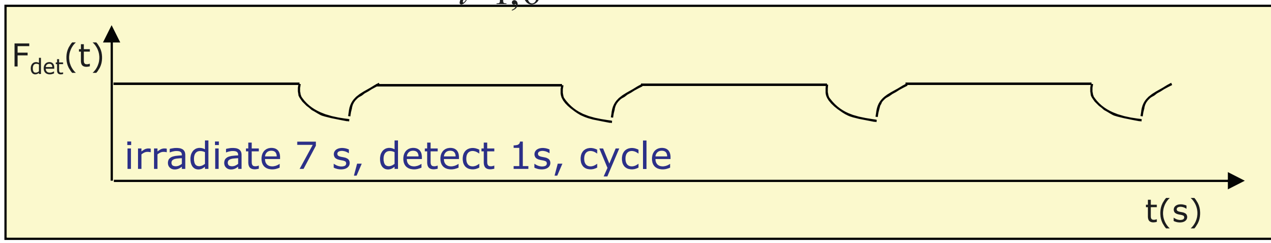
PRC87(2013)024601 Billnert et al.

Delayed neutrons

- **232Th**
- **PTB VdG**

$$F_{det}(t) = n_{fissions}\,\varepsilon\,Y_d(t)$$

$$Y_d(t) = \upsilon_d \sum_{i=1,6} a_i \exp(-\lambda_i t)\left(1-\exp(-\lambda_i t_{irr})\right)$$



Neutron detector :

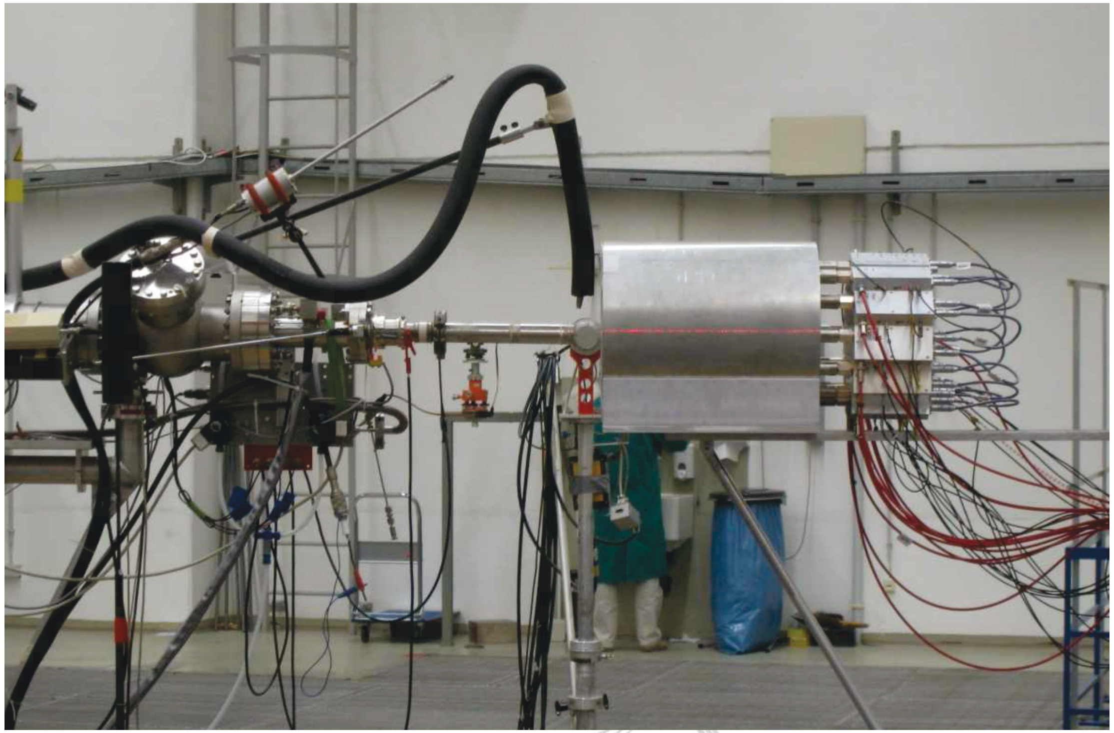
- Cylinder of CH2
  Φin=6 cm, Φout=16 cm, L=37cm)
- 12 tubes 3He
- efficiency ε > 20% (sample in centre)
- constant efficiency between 0.1 and 1 MeV
- not sensitive to gamma
- Also used for photofission studies

Thorium sample :

- Cylinder (r=1.275 cm, h=3 cm)
- m=182 g

Efficiency :

- MCNPX simulations
- Measurements with Cf-252 sources

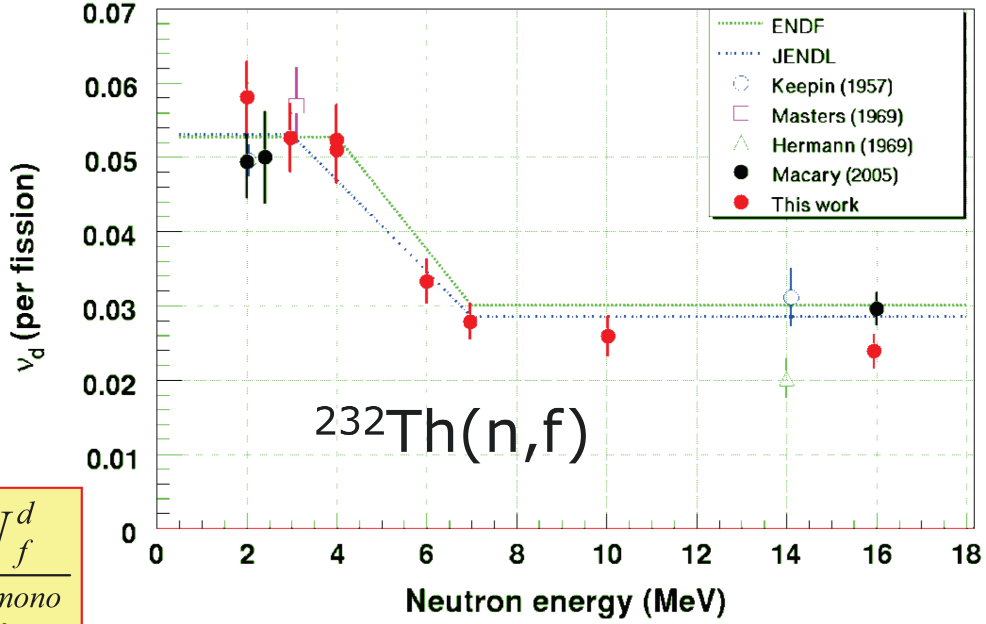


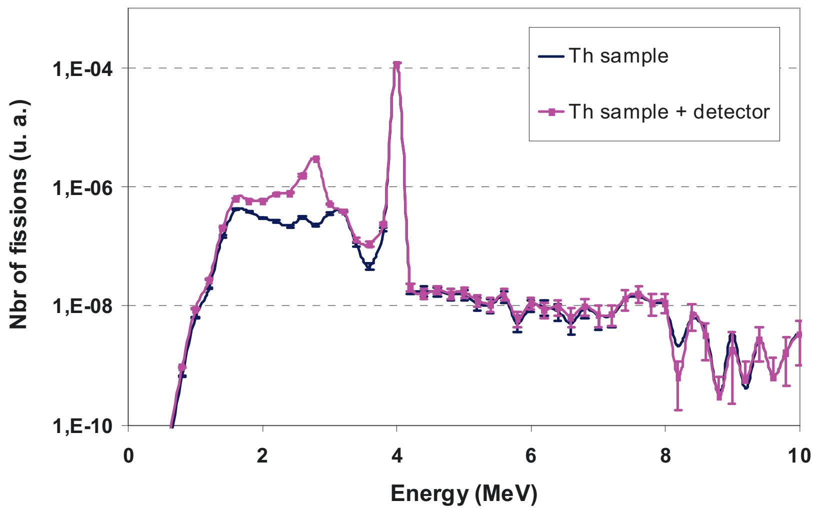
X. Ledoux et al. JEFF-GEDEPEON 2011

Delayed neutrons (Ledoux et al.)

- **Effective spectrum**
  - **Neutron source**
  - **Scattering in sample**
  - **Fission neutrons**
  - **Two background zones**
  - **to subtract**

$$\upsilon_d^{mono} = \frac{F_{det}(0)}{\varepsilon\, m\ \ N_f^{mono}\ Y_{\theta=0^\circ}\ K} - \upsilon^u \frac{N_f^u}{N_f^{mono}} - \upsilon^d \frac{N_f^d}{N_f^{mono}}$$





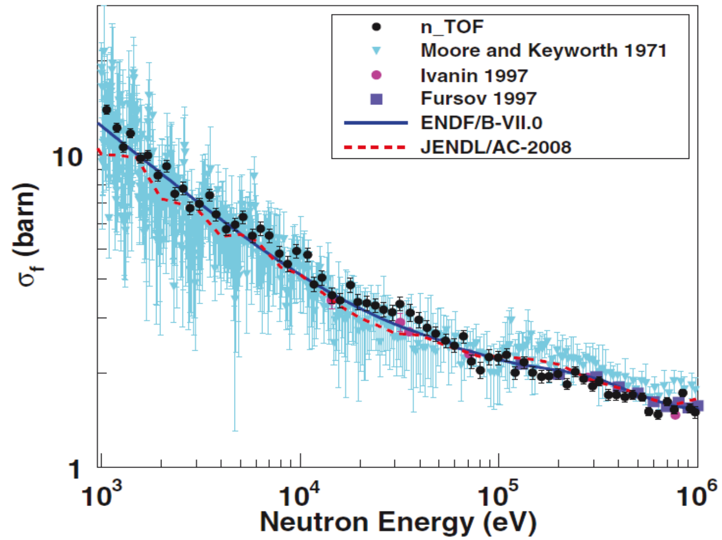
International collaboration

European projects acting on HPRL

ANDES
ERINDA
EUFRAT
GEDEPEON (NEEDS…)
TRAKULA
Subgroup-31
Subgroup-33

New: CHANDA
Broader: file projects, WPEC, IAEA

ANDES Example: ^{245}Cm(n,f), required: 3%
Calviani et al.: 5% PRC85(2012)034616



Belloni et al., ^{241}Am(n,f) EPJA49(2013)2
Belloni et al., ^{243}Am(n,f) EPJA47(2011)160
Belloni et al., ^{233}U(n,f), EPJA47(2011)2

Summary

- Fission is the process for energy production, safety, waste
- Many different aspects, all needed for a certain application
- Target uncertainty for cross sections and nubar are very tight
- For cross sections there is a remarkable bias for technique and normalization (FIC, 235U(n,f))
- Investigations in new technical options should be promoted
- There is considerable work ongoing that adds diversity with clear benefits (and also some learning points)
- Learn from past lessons
- International collaboration, in and beyond value is Europe, is a must

- Good new work is needed with new technology and enthusiastic new people