REACTION CROSS SECTIONS, FISSION YIELDS AND PROMPT-NEUTRON EMISSION FROM ACTINIDE TARGETS

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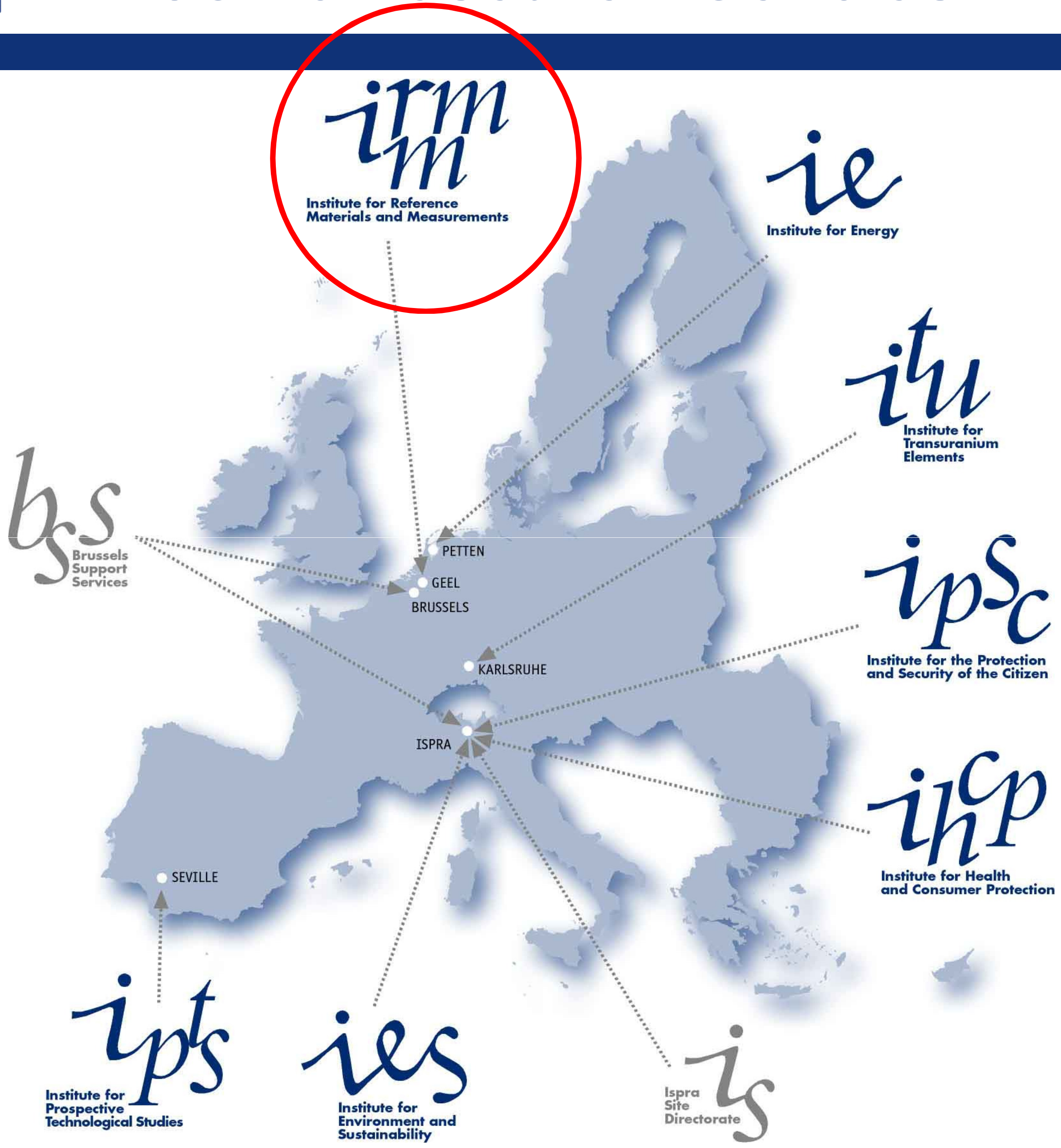
- ^{241}Am transmission and capture cross sections
- Conclusions

Joint Research Centres



Introduction

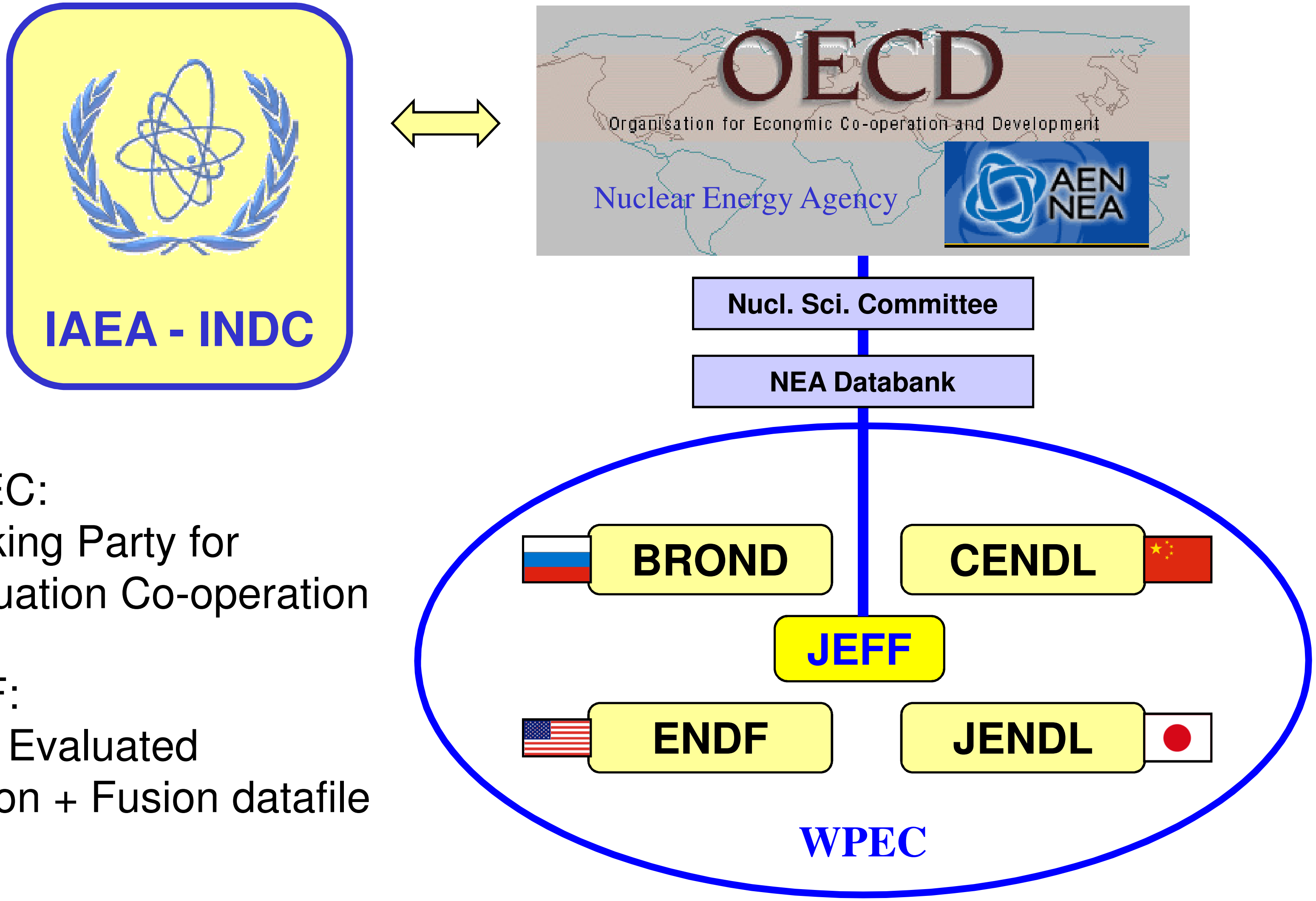
Mono-energetic neutron source (MONNET)

- 7 MV Van-de-Graaff accelerator
  - ^{7}LiF(p,n)^{7}Be, TiT(p,n)^{3}He, D_2(d,n)^{3}He, TiT(d,n)^{4}He
  - DC ($I_{p,d}$ < 50 μA), pulsed beam available
  - 4 + 1 non-T beam line
- $\Phi_n < 10^9$ /s/sr
- *NEPTUNE* isomer spectrometer
- ionisation chambers, NE213 neutron/gamma-ray detectors, BF_3 counters, HPGe detectors
- Bonner spheres
- fast rabbit systems ($T_{1/2}$ > 1s) for activation studies

GELINA neutron TOF spectrometer

- 70 - 140 MeV electron accelerator
- repetition frequency: 40 - 800 Hz
- neutron pulse: 2 μs - 1 ns @ FWHM
- $\Phi_n = 3.4\ 10^{13}$/s @ 800 Hz
- 12 different flight paths with a length between 8 and 400 m
- ionisation chambers, C_6D_6 detectors
- high-resolution γ-ray detectors
- fission chambers for flux monitoring

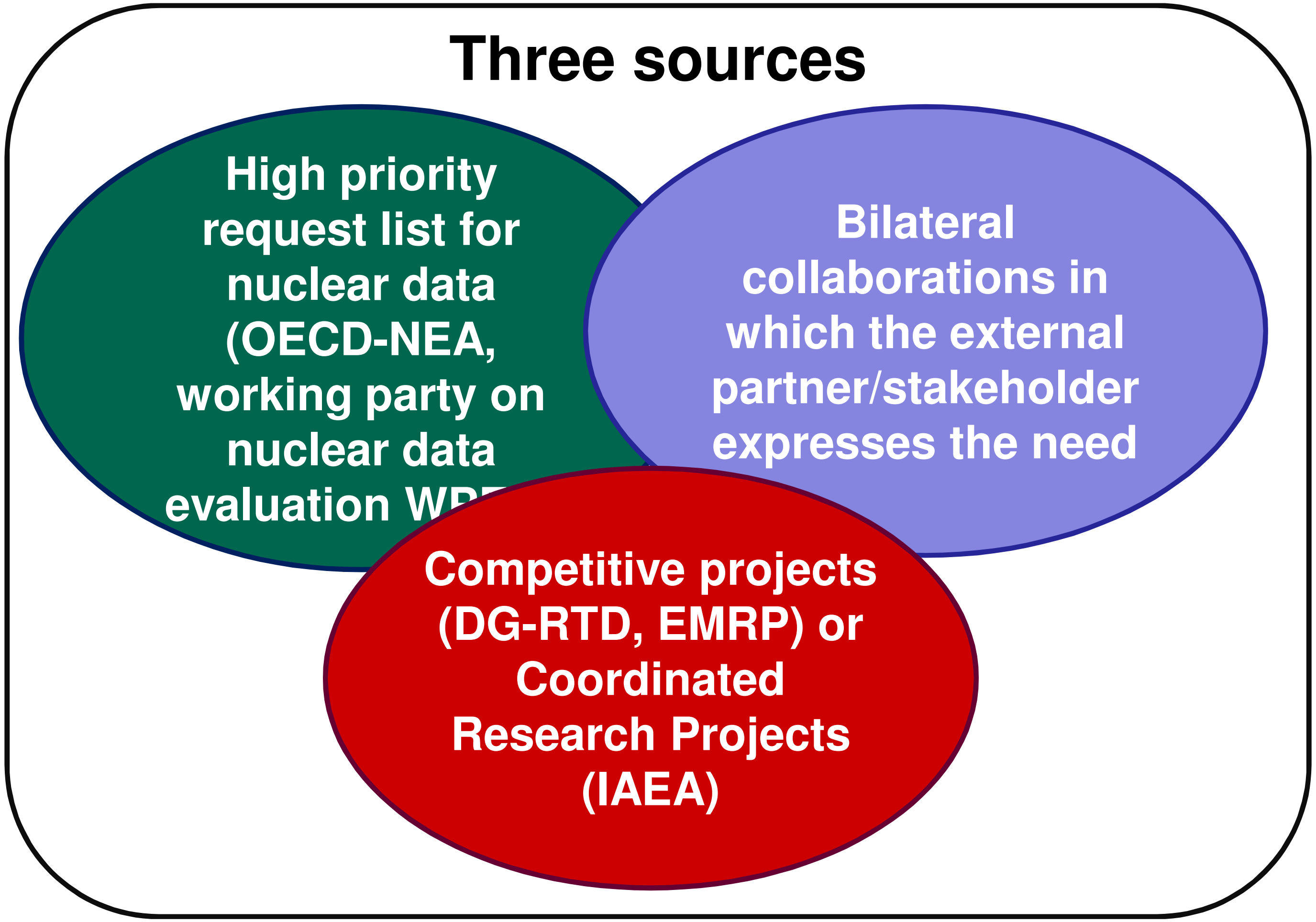
The OECD nuclear data network



WPEC:
Working Party for
Evaluation Co-operation

JEFF:
Joint Evaluated
Fission + Fusion datafile

Measurement needs derived from applications



Required Nuclear Data Accuracy for Fission Energy Applications

- **Reactor design parameters and uncertainties**

	A priori uncertainty (1 σ) associated to calculations of **classical SFRs** (SPX) Using unadjusted JEFF-3.1 data	***Targeted uncertainty*** (1 σ) for **innovative FR** calc., "performance" phase
k_{eff} (BOC)	1600 pcm	300 pcm
Power max core BOC	3%	2%
Power local (away from singularities)	5%	3%
Internal BG	± 0.06	± 0.02
EOC nuclide inventory, heavy nuclides and FPs	5% for major U & Pu isotopes 10-20% for other actinides	2% for major U & Pu isotopes 10% for other actinides
Control rod antireactivity	16% (single rod) 5% (bank)	10% (single rod) 2% (bank)
Coolant void	16% for both central and leakage components	7% for both central and leakage components
Doppler effect	10%	7%

Source: R. Jacqmin Uncertainties in reactor parameters due to nuclear data uncertainties

Similar lists for all Generation-IV systems were obtained by Subgroup-26 of the OECD-NEA working party on evaluation cooperation (WPEC)

The role of nuclear data

Required Nuclear Data Accuracy for Fission Energy Applications

- Typical uncertainties to be achieved in nuclear data to meet the requested performance (from SG26, very partial list)

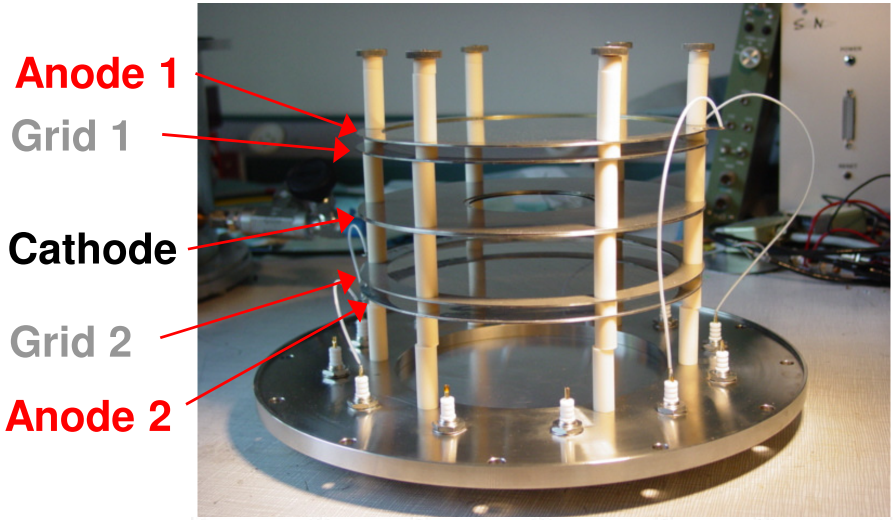
	Energy interval	Current uncertainties	Required uncertainties
U-238 inelastic	0.5 – 6 MeV	10-20%	4-5% (SFR), 2% (GFR)
U-238 capture	9 keV – 25 keV	9%	3-4% (SFR), 1.5% (GFR)
Pu-239 capture	2 keV – 67 keV	7-15%	6% (SFR), 3% (GFR)
Pu-240 capture	9 keV – 67 keV	10-11%	6-7% (SFR)
Pu-241 fission	9 keV – 1.35 MeV	9-20% (!?)	3% (SFR, GFR)
Na-23 inelastic	0.5 – 1.35 MeV	20-30%	4-8% (SFR)
Fe inelastic	0.5 – 1.35 MeV	15-25%	3-8% (SFR)
C elastic	0.5 – 1.35 MeV	5% (?)	2% (GFR)
Si-28 inelastic	1.35 – 2.2 MeV	50%	6% (GFR)

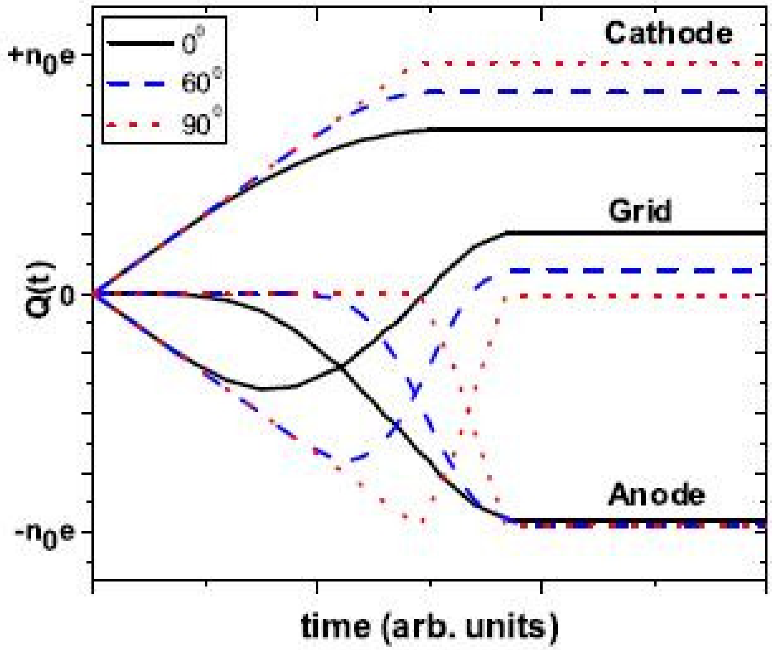
Subgroup-26 final report (OECD-NEA/WPEC M. Salvatores, R. Jacqmin):
www.nea.fr/science/wpec/volume26/volume26.pdf

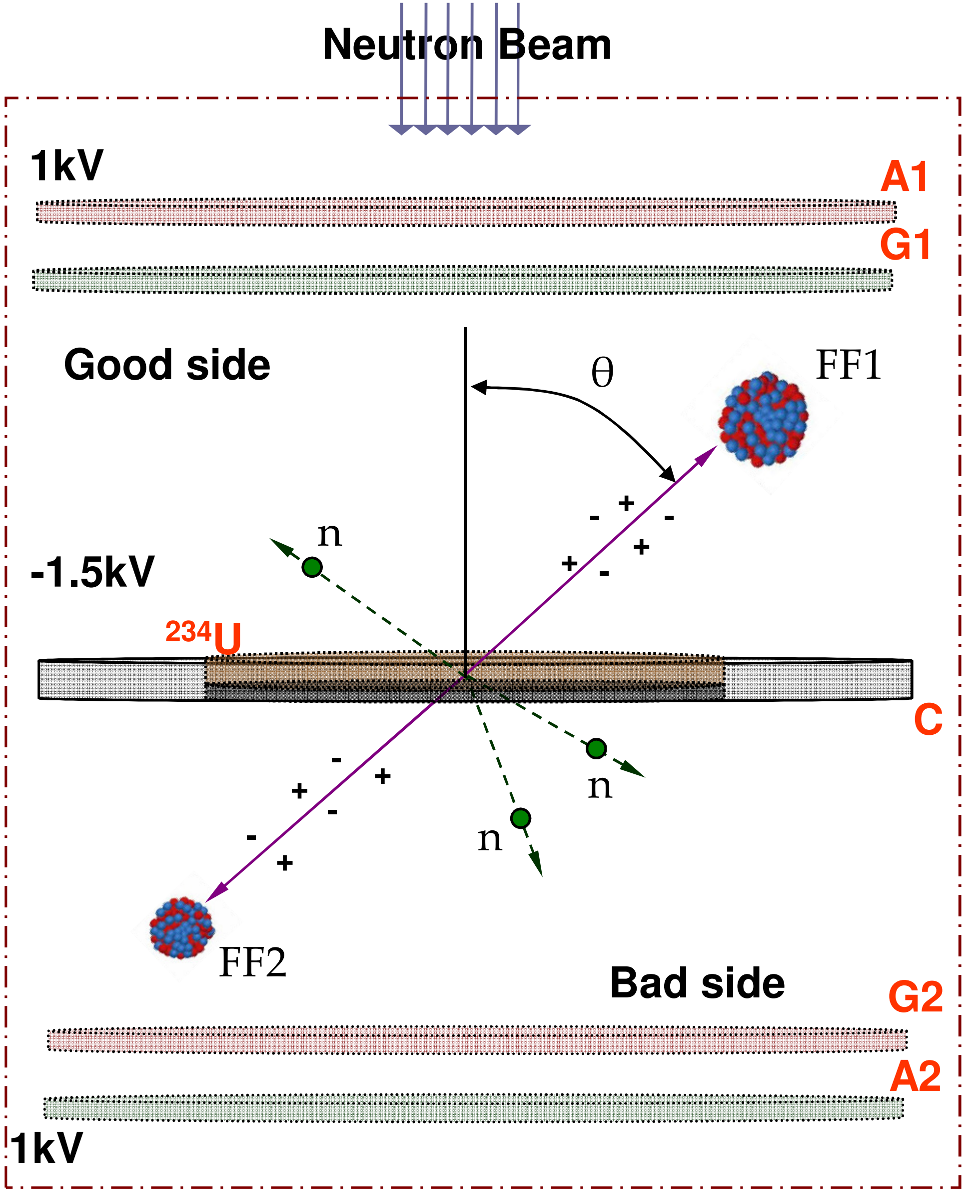
High priority request list for nuclear data
(OECD-NEA/WPEC Subgroup-C, A. Plompen)
www.nea.fr/html/dbdata/hprl

Neutron induced fission reaction (^{234}U)

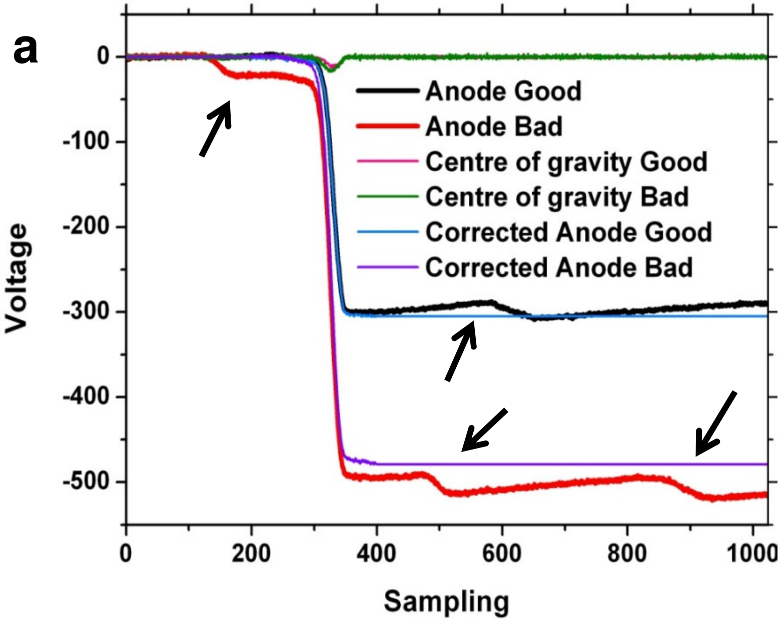
Ionisation chamber

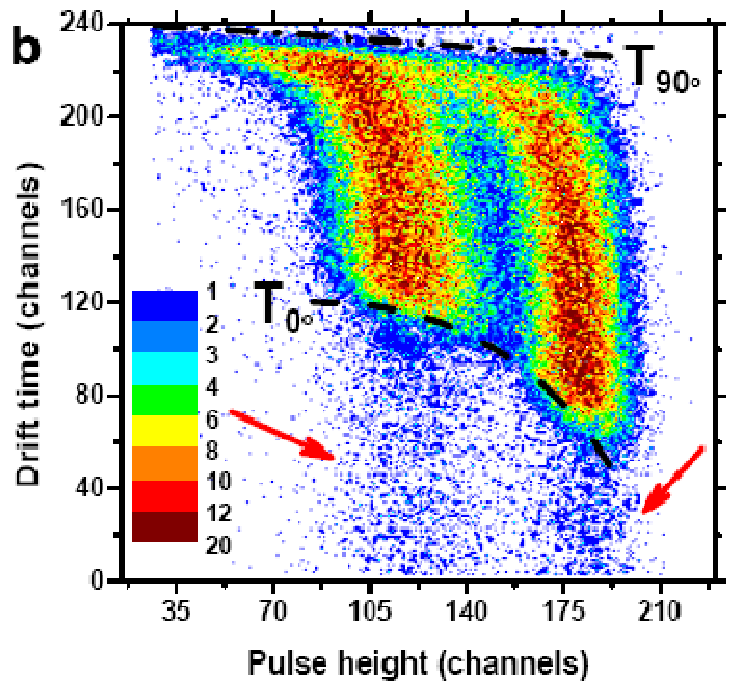


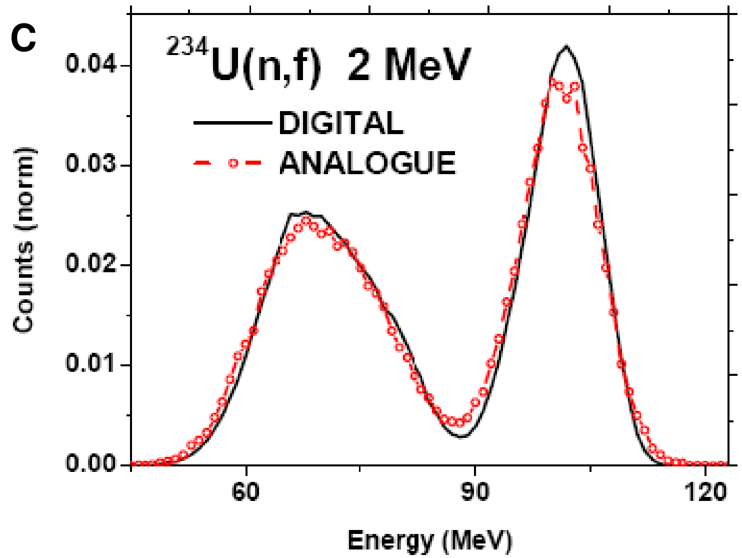




Digital data acquisition in fission reactions



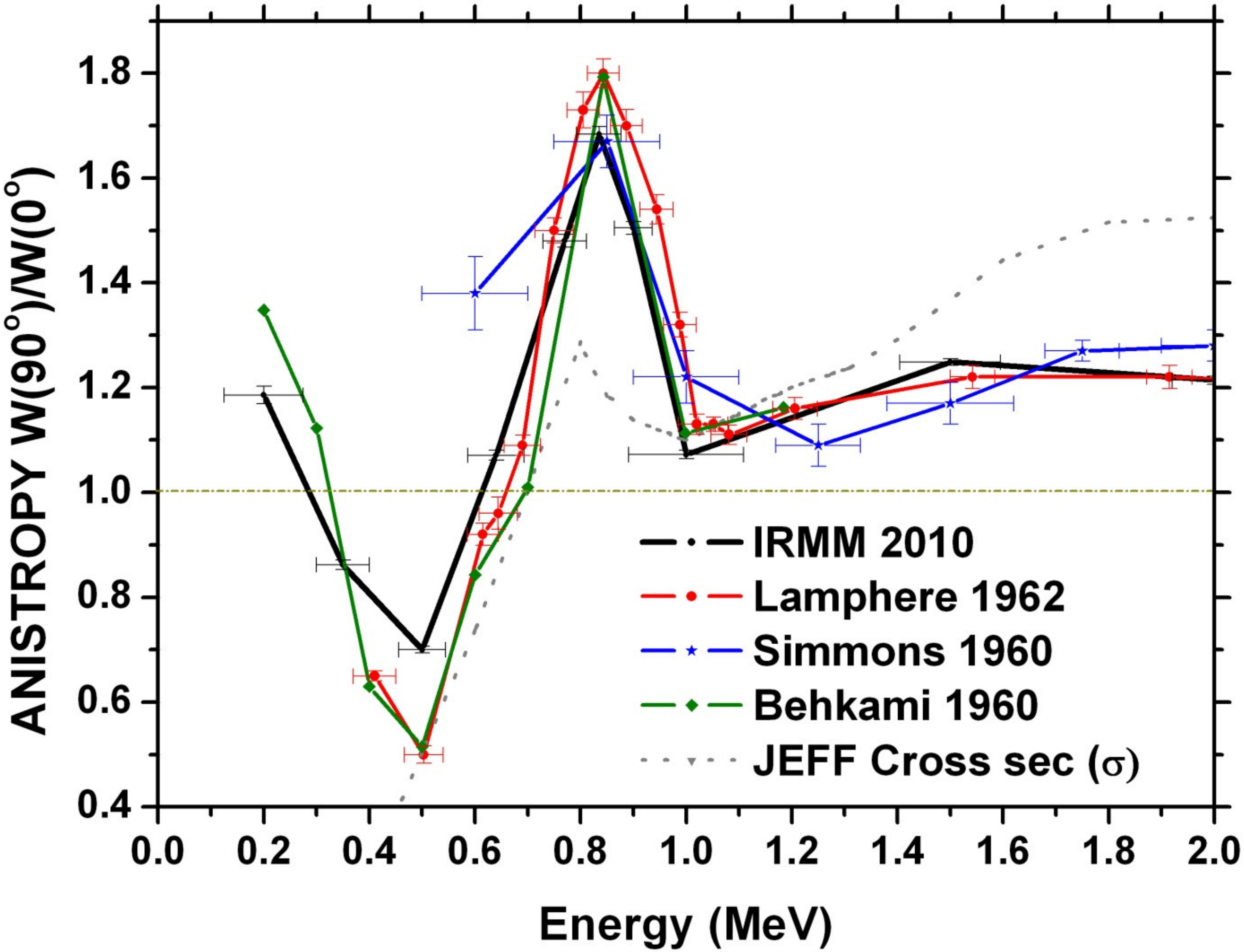




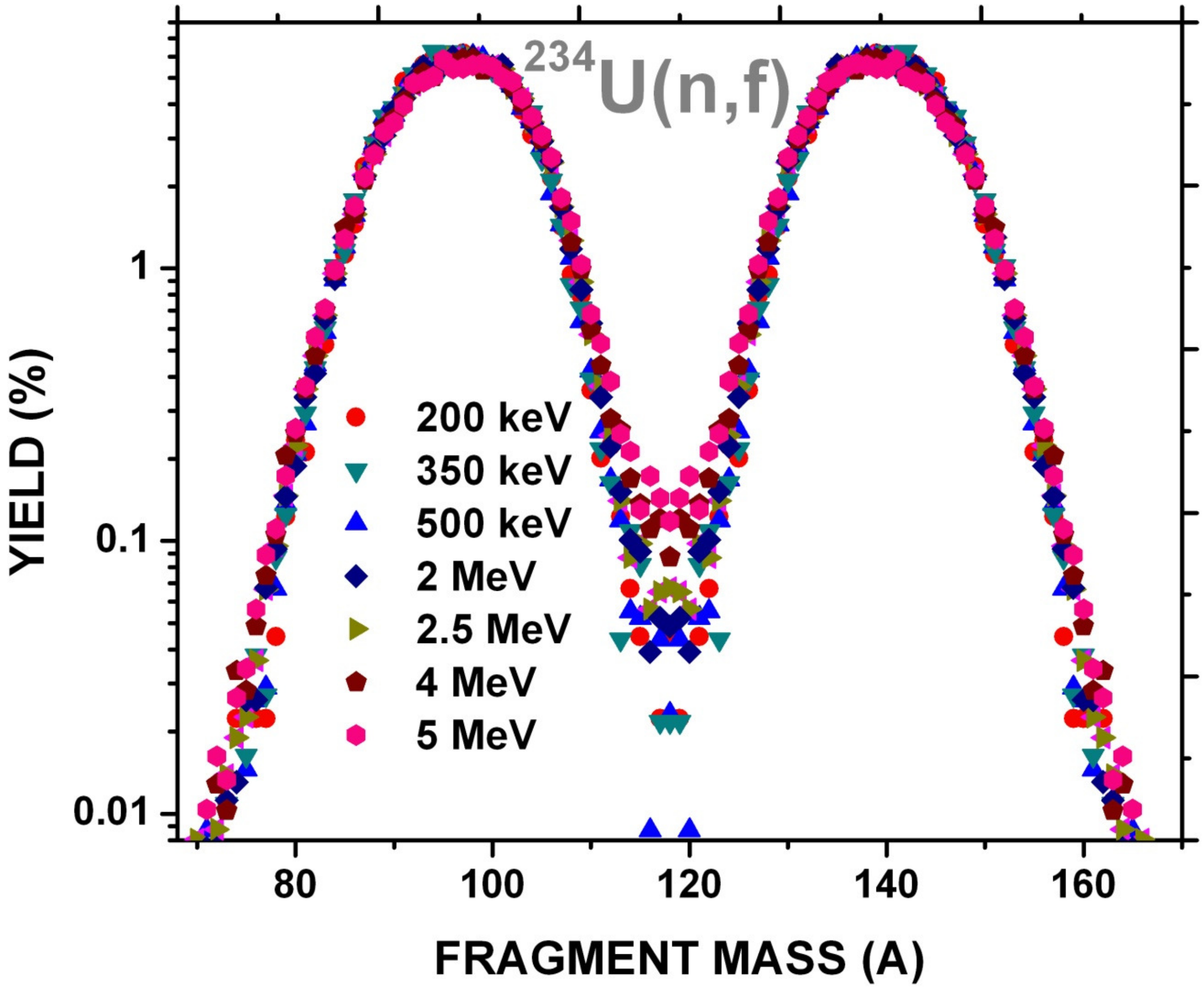
- a) Efficient Pile-up rejection
- b) Identification of false triggering
- c) Improved pulse height resolution

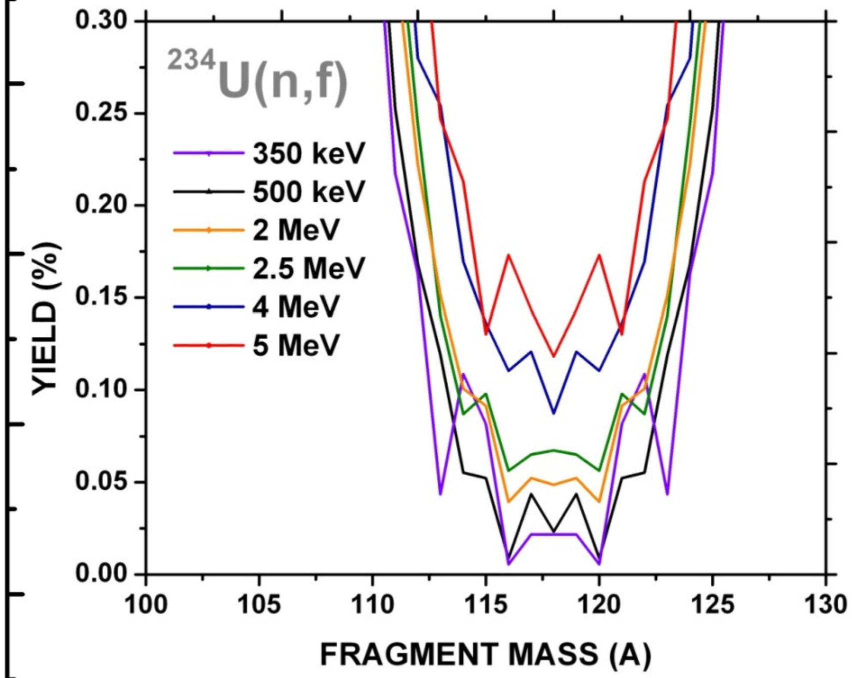
=> Accurate fission yield data

Angular anisotropy



Mass distributions





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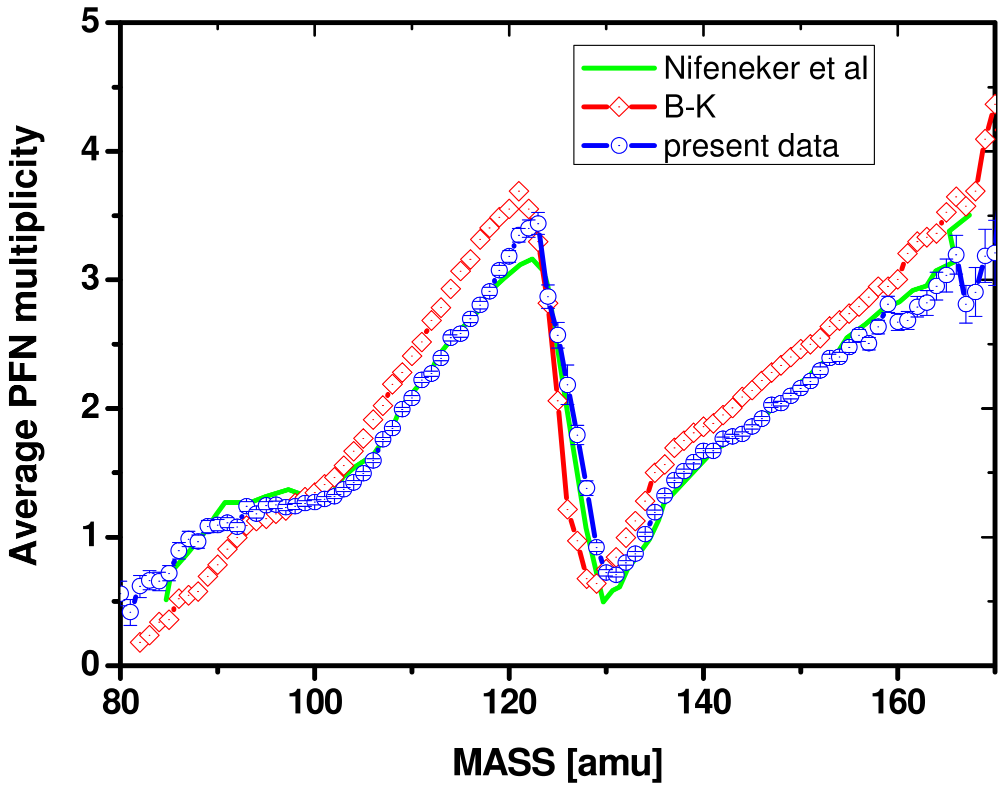
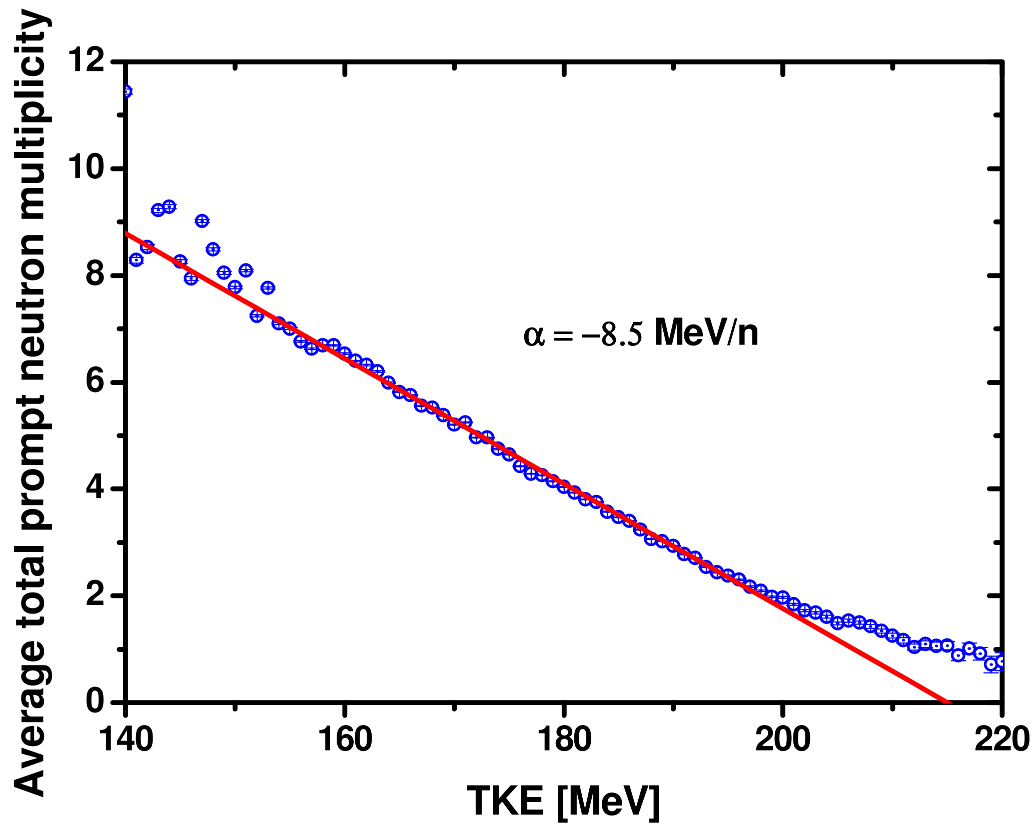
Neutron emission in fission of $^{252}Cf(SF)$

Neutron emission in fission

- Important nuclear data for understanding of the fission process and for nuclear applications
- Scarce available experimental data
- Quality of experimental data ??

Dependence of ν_{bar}





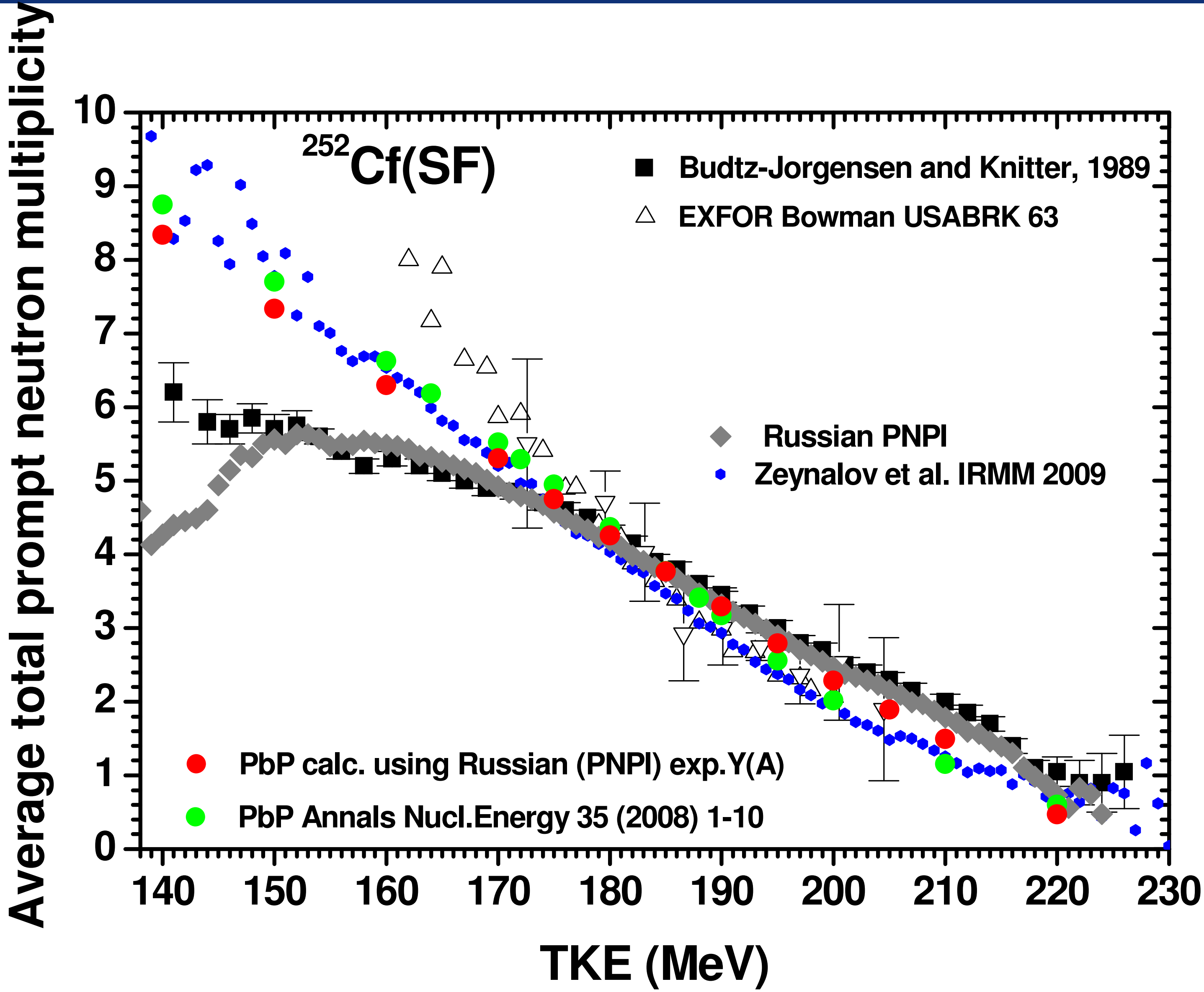
$$\nu(A) = \frac{\int_0^\infty \nu(A,TKE) * Y(A,TKE)\,dTKE}{\int_0^\infty Y(A,TKE)\,dTKE}$$

$$\nu(TKE) = \frac{\int_0^\infty \nu(A,TKE) * Y(A,TKE)\,dA}{\int_0^\infty Y(A,TKE)\,dA}$$

$$\bar{\nu} = \int_0^\infty \nu(A)Y(A)\,dA = 3.763$$

$$\bar{\nu} = \int_0^\infty \nu(TKE)Y(TKE)\,dTKE = 3.763$$

First time no strong reduction of ν_{bar} at low TKE

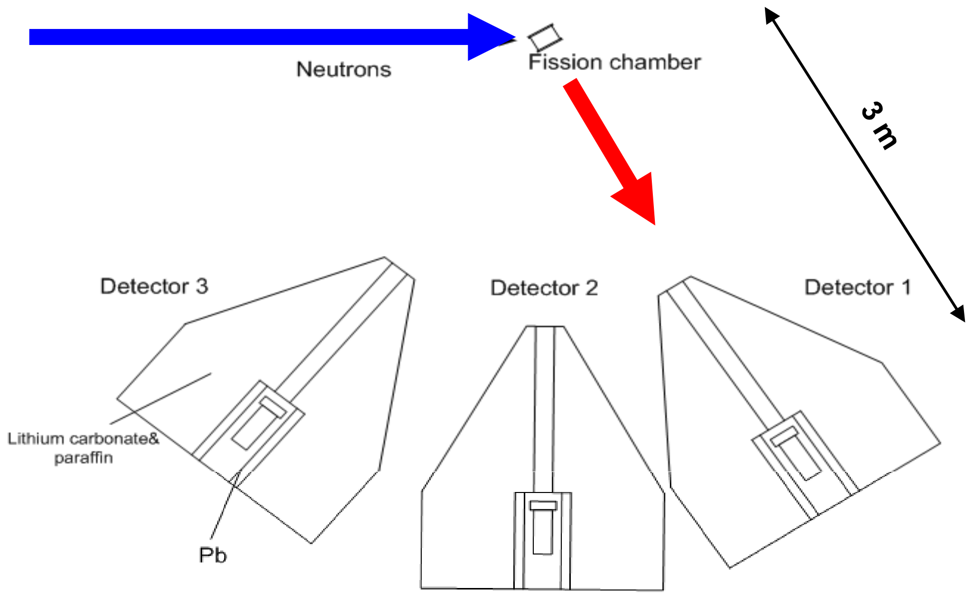

252Cf(SF)
Budtz-Jorgensen and Knitter, 1989
EXFOR Bowman USABRK 63
Russian PNPI
Zeynalov et al. IRMM 2009
PbP calc. using Russian (PNPI) exp.Y(A)
PbP Annals Nucl.Energy 35 (2008) 1-10
Average total prompt neutron multiplicity
TKE (MeV)
0
1
2
3
4
5
6
7
8
9
10
140
150
160
170
180
190
200
210
220
230

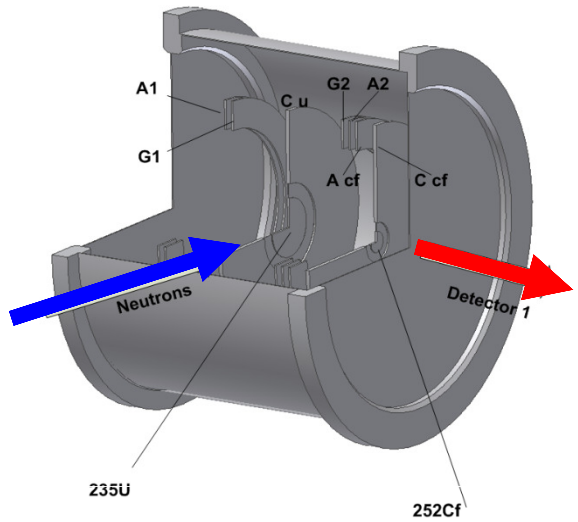
$^{235}U(n_{th},f)$ prompt fission neutron spectrum

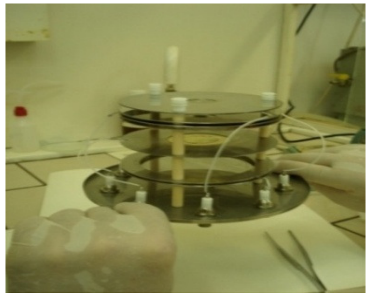
Neutron spectrum of $^{235}U(n_{th},f)$

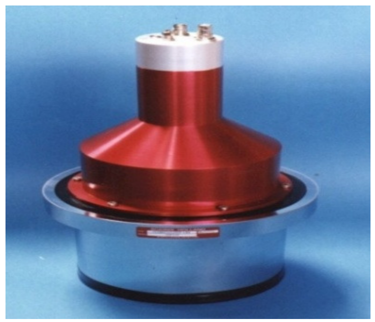
- Requested by subgroup 9 of WPEC
- Persisting discrepancies between macroscopic (integral) and microscopic data

Experiment: Setup





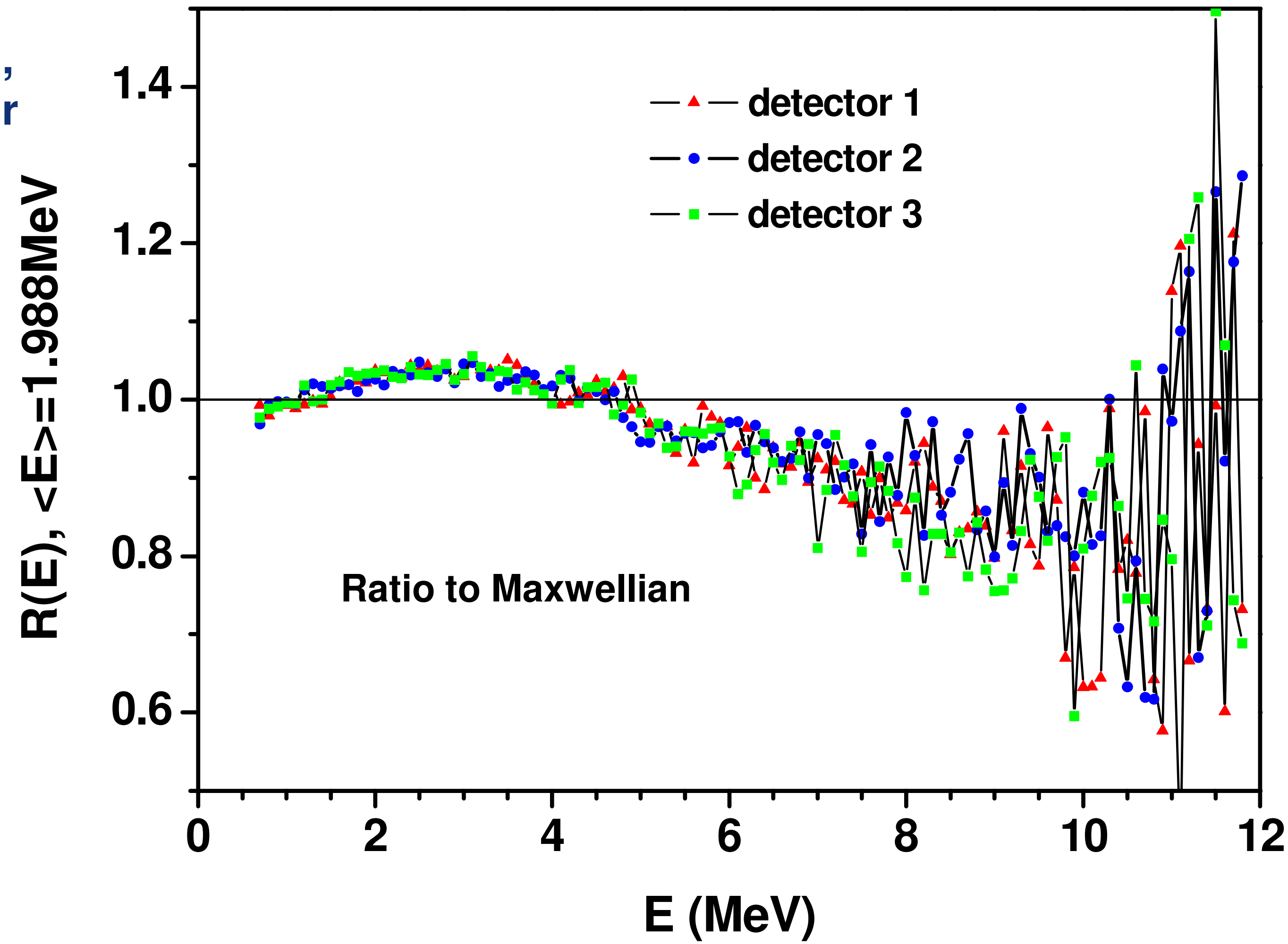




- TOF measurement technique used (L = 3 m)
- 3 neutron detectors LS301 (NE213 equivalent, size: 4'' x 2'' =10.16 x 5.08 cm) SCIONIX in heavy shielding
- Thin ^{235}U (97.7%) target 112 μg/cm^2 at centre of Ionisation chamber, fission count rate 50.000 /sec
- ^{252}Cf target placed simultaneously into the same chamber shifted 5 cm relative to ^{235}U target (20.000 fissions/s)
- High Fission Fragment counting efficiency 98%

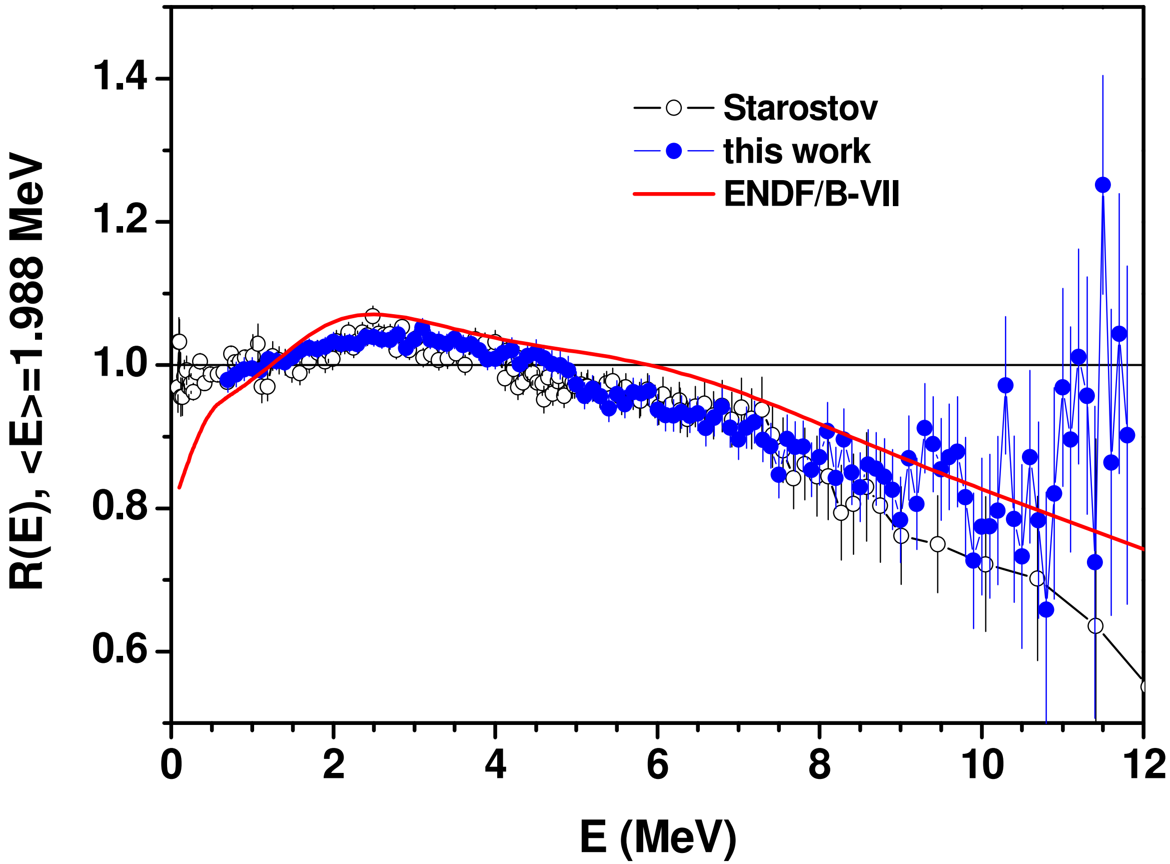
Neutron detector spectra

- Since 3 detectors were used, they can be checked for reliability of results
- Each Run was analyzed separately to check for systematic errors
- No angular effect



Excellent agreement of 3 individual neutron detectors

Comparison to Literature Data



Our Data

Starostov et al. 1984 (EXFOR)

ENDF/B-VII

Ratio to Maxwellian

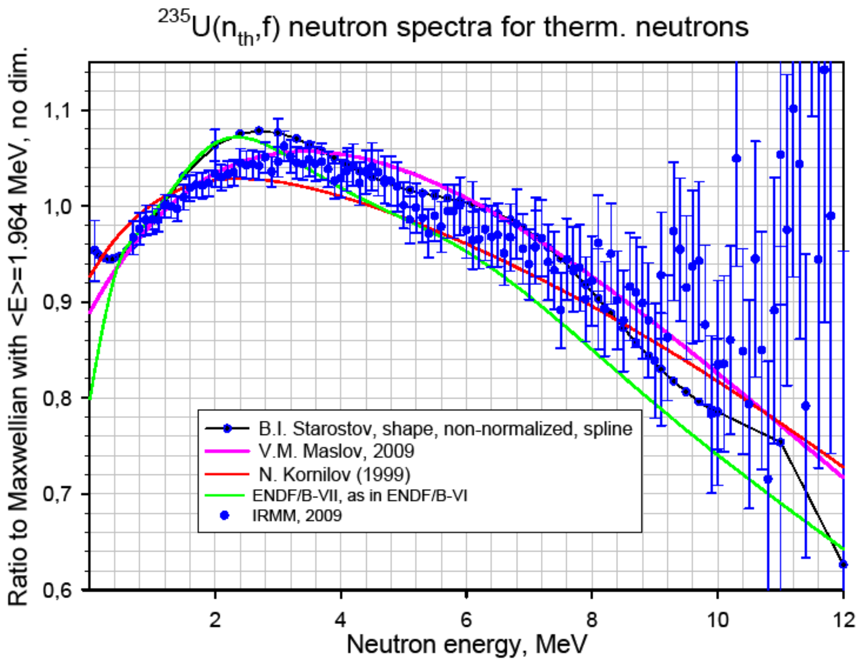
- Starostov et al.: Gas-scintillation-ionization detector + ^{235}U, IC, Reactor, relative to ^{252}Cf
- Excellent agreement with Starostov et al. over full energy range
- Our data and Starostov et. al. contradict ENDF/B-VII evaluation and the Los Alamos Model (Madland Nix)

Prompt fission neutron spectra

- **impact benchmarks k_{eff} as strongly as cross sections:**
  - *+ 500 pcm* for solutions (unique amongst all libraries)
  - *- 300 pcm* for thermal U but *+ 300 pcm* for fast U
  - *+ 800 pcm* for thermal Pu but *- 300 pcm* for fast Pu

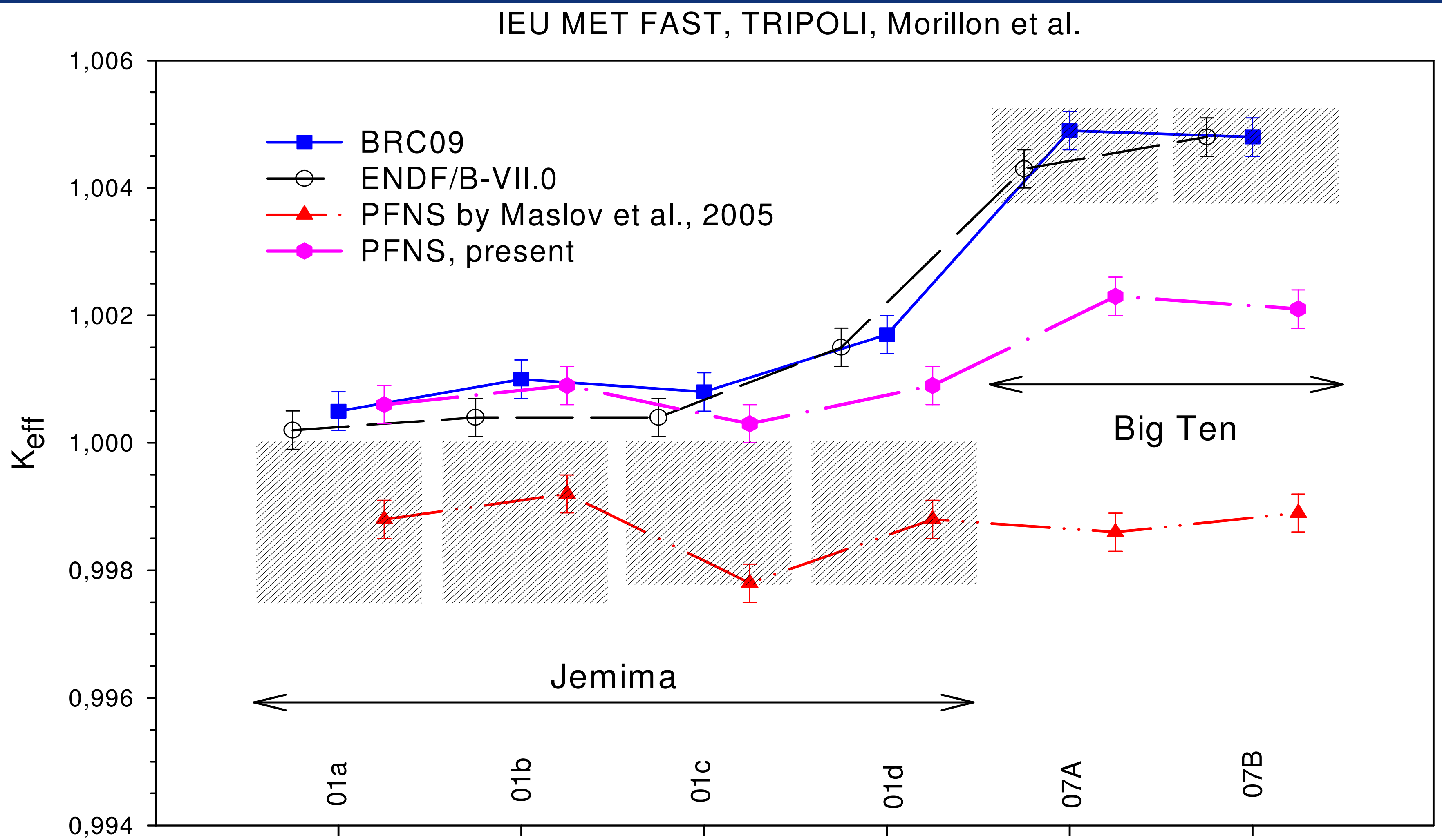
- **are as important as cross sections or angular distributions**

=> IAEA CRP on Prompt fission neutron spectra



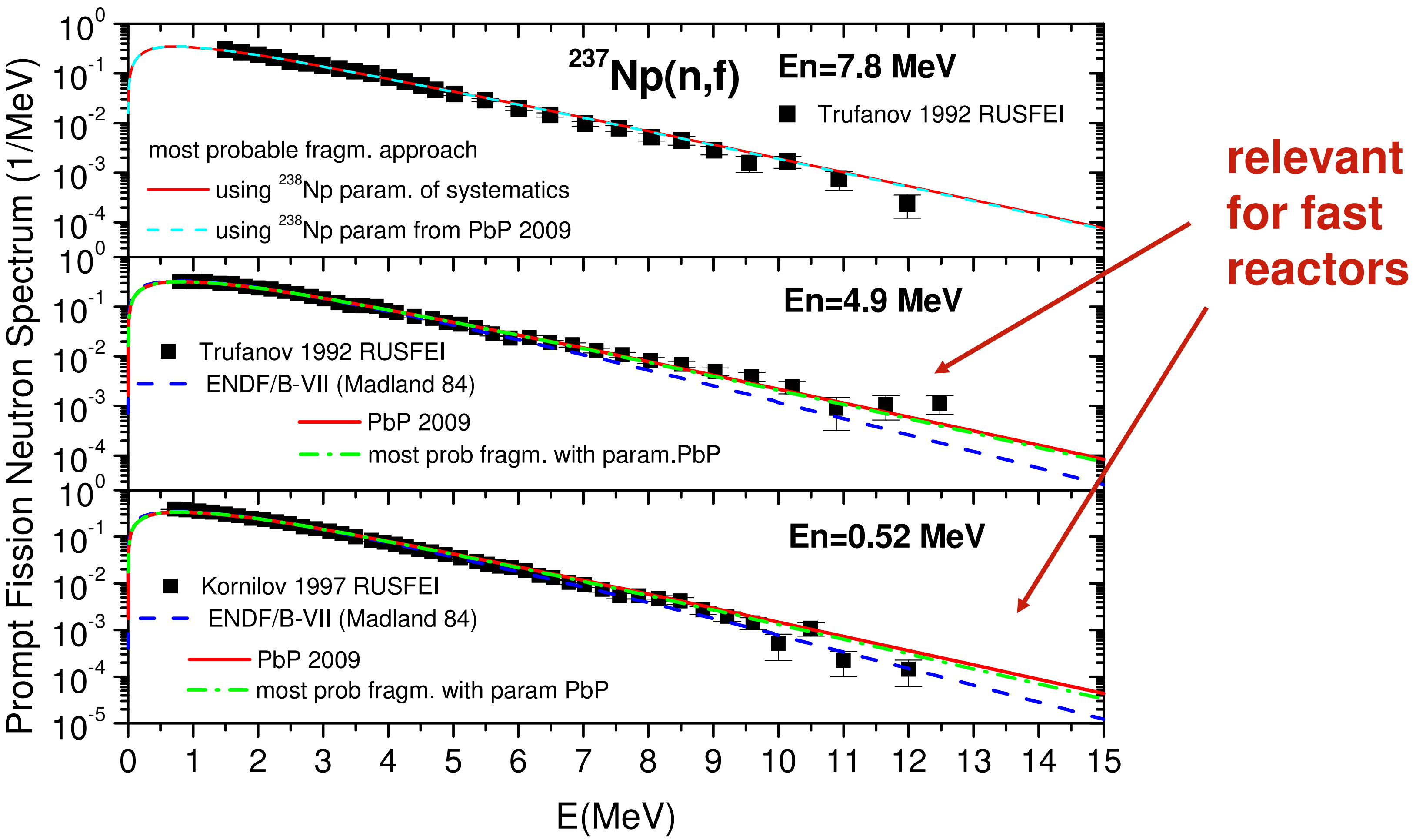
- ✓ Recent measurements performed by IRMM @ reactor in Budapest (EFNUDAT project)
- ✓ previous data from IPPE Obninsk confirmed
- ✓ Disagreement with the Los Alamos model (up to now still accepted reference)
- ✓ (new data adopted in most recent ENDF/B-VII library)
- ✓ New efforts for an improved theoretical description in collaboration with LANL and JINER, Minsk (ISTC project)

Benchmarking critical assemblies



K_{eff} very sensitive to mean energy and shape of PFNS

Prompt fission neutron spectrum



Successful modelling needs high quality input data: mass yield and kinetic energy of fission fragments

^{241}Am capture and transmission

Resonance parameters for $^{241}Am + n$ Literature data

Transmission

– Derrien and Lucas **(EXFOR)**
- Saclay, LINAC (17 m and 53 m)
- AmO_2 powder
- $^{10}B(n,\alpha_1)$, 478 keV with NaI

– Kalebin et al. **At. Energ. 40 (1976) 303**
- Chopper
- AmO_2 powder
- BF_3 proportional counters
- Dimension target in beam

  ~0.8 and 0.4 mm

Capture

– Weston and Todd **NSE 61 (1976) 356**
- ORELA (20 m and 85 m)
- AmO_2 + S powder
- Total energy detection + WF (C6F6)
- Normalization: $\sigma(n_{th},\gamma) = 582$ b

– Jandel et al. **PRC 78 (2008) 034609**
- LANSCE (20 m)
- ^{241}Am electroplated on Ti
- Total absorption (4π)
  $M\gamma = 4$ and $3.75 < E\gamma_{tot} < 5.4$ MeV
  $\varepsilon n,\gamma = 12.5 \pm 1.0$ %
- Normalization at 4.9 eV of $^{197}Au(n,\gamma)$
  RP for 4.9 eV (not specified)
  no limits on $M\gamma$ and $E\gamma_{tot}$
  $\Rightarrow \sigma(n_{th},\gamma) = 655 \pm 33$ b

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Impact of target properties $^{242}PuO_2$ powder diluted in carbon powder at 300 K

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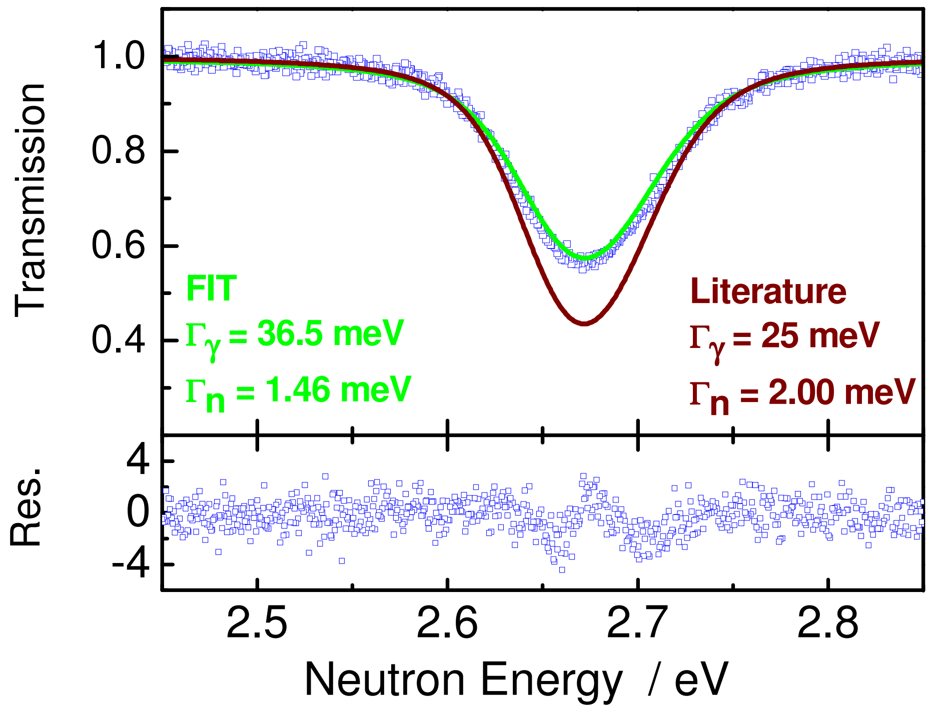
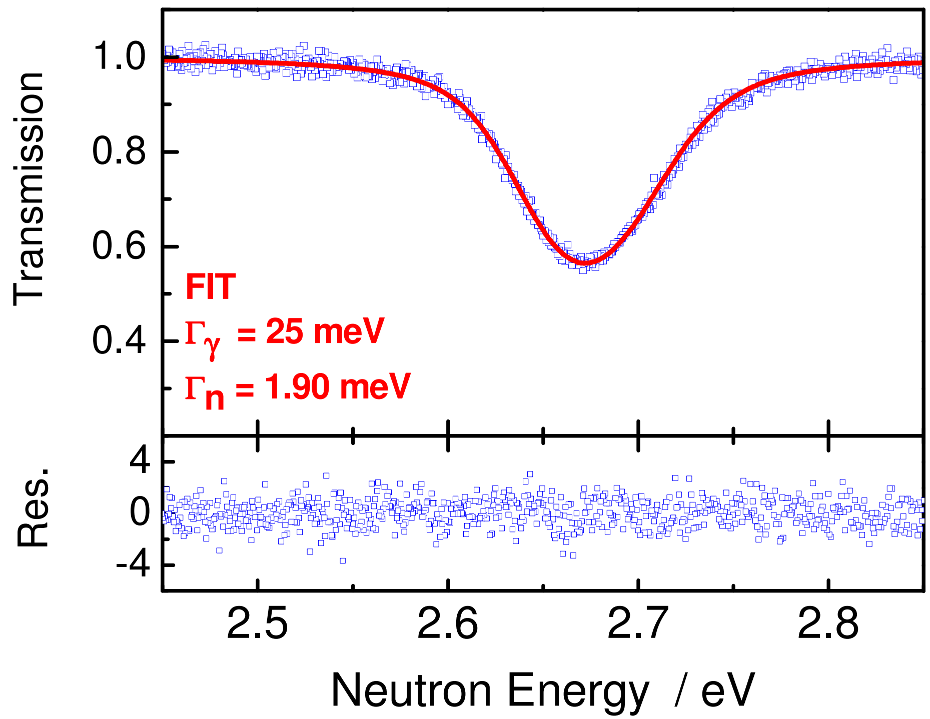
Homogeneous sample



REFIT: accounting for the powder grain size

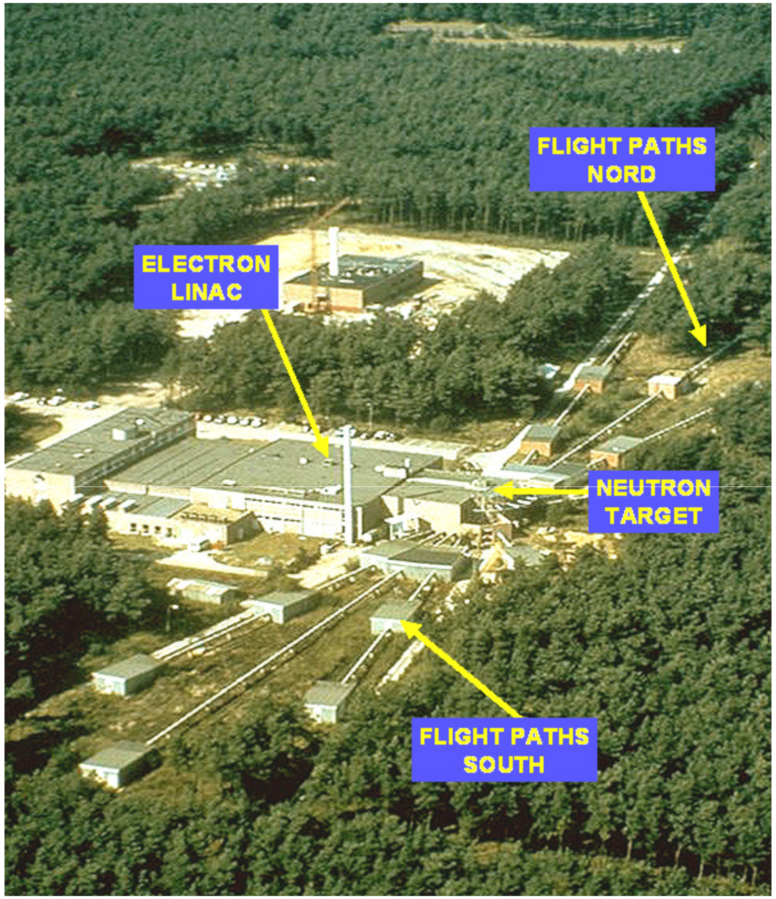


For inhomogeneous target

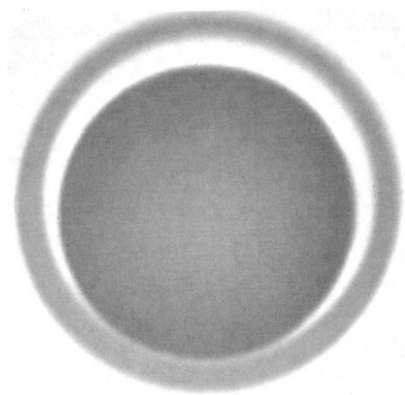
For $\Gamma_n < \Gamma_\gamma$

- **Γ_n underestimated**
- **Γ_γ overestimated**

TOF - experiments at GELINA



- **Sample (JRC-ITU Karlsruhe)**
  - AmO_2 homogeneously diluted in a Y_2O_3 matrix (solgel method)
  - Ø = 22.1 mm
  - Homogeneity verified by X-ray radiography
  - Impurities : mass spectrometry
  - ~ 325 mg ^{241}Am (40 GBq) by γ - spectroscopy (calorimetry planned)
- **Transmission at 25 m**
  - ^{6}Li-glass scintilllators
- **Capture at 12.5 m**
  - Total energy detection
  - C_6D_6 detectors + WF (validated by exp.)
  - Flux : $^{10}B(n,\alpha)$ IC
  - Normalization
    Internal : Γ_n from transmission
    External : 4.9 eV of $^{197}Au+n$ (saturated)



Comparison with LANSCE (Jandel et al.) E_r and Γ_γ

E_r / eV		Γ_γ / meV		
GELINA	LANSCE	GELINA	LANSCE	Weston and Todd
0.30605 ± 0.000014	0.3051 ± 0.0002	42.4	44.4 ± 0.3	46.9 ± 0.3
0.57387 ± 0.00026	0.5724 ± 0.0003	41.1	43.3 ± 0.5	47.3 ± 0.3
1.27106 ± 0.00058	1.2718 ± 0.0004	42.6	45.3 ± 0.7	49.2 ± 0.3

GELINA :

- Flight path length traceable to E_r = 6.6735 ± 0.0030 eV eV of ^{238}U + n (ORELA)
- Response function of GELINA in REFIT includes neutron storage term (Ikeda and Carpenter)

Conclusions

- Data needs for innovative systems (e.g. GEN IV) are summarized in HPRL and WPEC Subgroup 26 document.
- To structure our work we have strong collaboration with international organizations (NEA, IAEA) and participate in EU programs (e.g. EUROTRANS, EFNUDAT, ….).
- Theoretical modelling of reaction cross sections and the fission process strongly dependent on high quality experimental data as input to the codes.
- New IAEA CRP started due to the problems encountered with the PFNS. First result of new PFNS shows better agreement with benchmarks. Points to the importance of the PFNS.

= > Prompt neutron multiplicities and spectra are crucial nuclear data

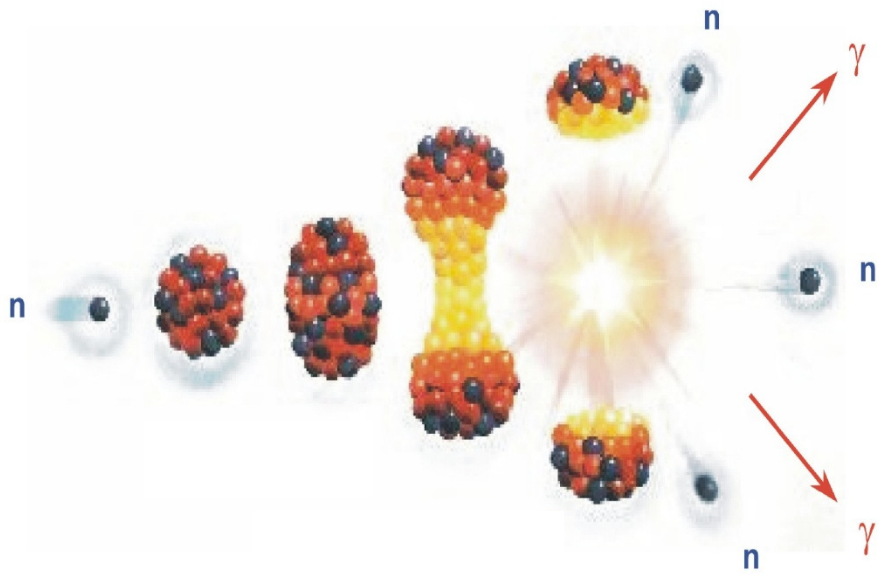
Conclusions and Outlook

- **Impact of target properties and importance of target characterization**
- **Transmission and capture yield (counts) are fully consistent**

  ⇒ in a simultaneous analysis of T_{exp} and Y_{exp} the application of a weighting function is in first approximation not required

- **Capture data at 400 Hz and 800 Hz (extension of energy region)**
- **Application of WF and verify normalization by 4.9 eV of $^{197}Au + n$**
- **Determination of ^{241}Am quantity by calorimetry ($\delta n/n < 1.0$ %)**

  ⇒ $\sigma(n_{th},\gamma)$ and (E_r, $g\Gamma_n$, $<\Gamma>$) up to ~ 300 eV



Thank you for your attention ☺