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

F.-J. Hambsch, S. Oberstedt, A. Al-Adili, P. Schillebeeckx, S. Kopecky, C. Sage, C. Lampoudis,

IRMM - Institute for Reference Materials and Measurements
Geel - Belgium

http://irmm.jrc.ec.europa.eu/
http://www.jrc.ec.europa.eu/
Content

- Introduction
- Neutron induced fission reaction ($^{234}$U)
- Neutron emission in fission of $^{252}$Cf(SF)
- Prompt fission neutron spectrum of $^{235}$U($n_{th}$,f)
- $^{241}$Am transmission and capture cross sections
- Conclusions
Introduction

Mono-energetic neutron source (MONNET)

- 7 MV Van-de-Graaff accelerator
  - $^7\text{LiF}(p,n)^7\text{Be}$, $\text{TIT}(p,n)^3\text{He}$, $\text{D}_2(d,n)^3\text{He}$, $\text{TIT}(d,n)^4\text{He}$
  - DC ($I_{p,d} < 50 \mu\text{A}$), pulsed beam available
  - 4 + 1 non-T beam line
- $\Phi_n < 10^9 /\text{s}/\text{sr}$
- NEPTUNE isomer spectrometer
- ionisation chambers, NE213 neutron/gamma-ray detectors, BF$_3$ counters, HPGe detectors
- Bonner spheres
- fast rabbit systems ($T_{1/2} > 1\text{s}$) for activation studies

GELINA neutron TOF spectrometer

- 70 - 140 MeV electron accelerator
- repetition frequency: 40 - 800 Hz
- neutron pulse: 2 $\mu$s - 1 ns @ FWHM
- $\Phi_n = 3.4 \times 10^{13}/\text{s} @ 800 \text{ Hz}$
- 12 different flight paths with a length between 8 and 400 m
- ionisation chambers, $\text{C}_6\text{D}_6$ detectors
- high-resolution $\gamma$-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

IAEA - INDC

BROND
CENDL
JEFF
ENDF
JENDL

Nucl. Sci. Committee
NEA Databank

WPEC
Measurement needs derived from applications

Three sources

- High priority request list for nuclear data (OECD-NEA, working party on nuclear data evaluation WPEC)
- Bilateral collaborations in which the external partner/stakeholder expresses the need
- Competitive projects (DG-RTD, EMRP) or Coordinated Research Projects (IAEA)
The role of nuclear data

Required Nuclear Data Accuracy for Fission Energy Applications

- Reactor design parameters and uncertainties

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

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)
### 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)

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

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**Subgroup-26 final report (OECD-NEA/WPEC M. Salvatores, R. Jacqmin):**


**High priority request list for nuclear data**

(OECD-NEA/WPEC Subgroup-C, A. Plompen)

Neutron induced fission reaction ($^{234}\text{U}$)
Ionisation chamber

Anode 1
Grid 1
Cathode
Grid 2
Anode 2

Neutron Beam

Good side

-1.5kV

Bad side

1kV

234U

FF1

FF2

-1.5kV

1kV
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

ANISOTROPY $W(90^\circ)/W(0^\circ)$

Energy (MeV)

IRMM 2010
Lamphere 1962
Simmons 1960
Behkami 1960
JEFF Cross sec ($\sigma$)
Mass distributions
Neutron emission in fission of $^{252}\text{Cf(SF)}$
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 $\nu_{\text{bar}}$

First time no strong reduction of $\nu_{\text{bar}}$ at low TKE

\[ \nu(A) = \frac{\int_0^\infty \nu(A,\text{TKE}) Y(A,\text{TKE}) d\text{TKE}}{\int_0^\infty Y(A,\text{TKE}) d\text{TKE}} \]

\[ \nu(\text{TKE}) = \frac{\int_0^\infty \nu(A,\text{TKE}) Y(A,\text{TKE}) dA}{\int_0^\infty Y(A,\text{TKE}) dA} \]

\[ \nu = \int_0^\infty \nu(A) Y(A) dA = 3.763 \]

\[ \nu = \int_0^\infty \nu(TKE) Y(TKE) dTKE = 3.763 \]

$\alpha = -8.5 \text{ MeV/n}$
Average total prompt neutron multiplicity

- $^{252}$Cf(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)
\(^{235}\text{U}(n_{th},f)\) prompt fission neutron spectrum
Requested by subgroup 9 of WPEC

Persisting discrepancies between macroscopic (integral) and microscopic data
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 $^{235}\text{U}$ (97.7%) target 112 $\mu$g/cm$^2$ at centre of ionisation chamber, fission count rate 50,000 /sec

$^{252}\text{Cf}$ target placed simultaneously into the same chamber shifted 5 cm relative to $^{235}\text{U}$ target (20,000 fissions/s)

High Fission Fragment counting efficiency 98%
• Since 3 detectors were used, they can be cross-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
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)
Impact benchmarks $k_{\text{eff}}$ 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

IEU MET FAST, TRIPOLI, Morillon et al.

K_{eff} very sensitive to mean energy and shape of PFNS
Successful modelling needs high quality input data: mass yield and kinetic energy of fission fragments
$^{241}$Am capture and transmission
Resonance parameters for $^{241}\text{Am} + n$

**Transmission**

- Derrien and Lucas *(EXFOR)*
  - Saclay, LINAC (17 m and 53 m)
  - $\text{AmO}_2$ powder
  - $^{10}\text{B}(n,\alpha_1)$, 478 keV with NaI

- Kalebin et al. *At. Energ. 40* (1976) 303
  - Chopper
  - $\text{AmO}_2$ powder
  - $\text{BF}_3$ 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)
  - $\text{AmO}_2 + \text{S}$ powder
  - Total energy detection + WF (C6F6)
  - Normalization: $\sigma(n_{\text{th}},\gamma) = 582$ b

  - LANSCE (20 m)
  - $^{241}\text{Am}$ electroplated on Ti
  - Total absorption (4π)
    - $M_\gamma = 4$ and $3.75 < E_{\gamma\text{tot}} < 5.4$ MeV
    - $\varepsilon_{n,\gamma} = 12.5 \pm 1.0$ %
  - Normalization at 4.9 eV of $^{197}\text{Au}(n,\gamma)$
    - RP for 4.9 eV (not specified)
    - no limits on $M_\gamma$ and $E_{\gamma\text{tot}}$
      - $\Rightarrow \sigma(n_{\text{th}},\gamma) = 655 \pm 33$ b
Impact of target properties $^{242}$PuO$_2$ powder diluted in carbon powder at 300 K

For inhomogeneous target
For $\Gamma_n < \Gamma_\gamma$
$\Gamma_n$ underestimated
$\Gamma_\gamma$ overestimated

Homogeneous sample

REFIT: accounting for the powder grain size
TOF - experiments at GELINA

**Sample (JRC-ITU Karlsruhe)**
- AmO$_2$ homogeneously diluted in a Y$_2$O$_3$ matrix (solgel method)
  - Ø = 22.1 mm
  - Homogeneity verified by X-ray radiography
  - Impurities: mass spectrometry
  - ~ 325 mg $^{241}$Am (40 GBq) by $\gamma$-spectroscopy (calorimetry planned)

**Transmission at 25 m**
- $^6$Li-glass scintillators

**Capture at 12.5 m**
- Total energy detection
- C$_6$D$_6$ detectors + WF (validated by exp.)
- Flux: $^{10}$B(n,$\alpha$) IC
- Normalization
  - Internal: $\Gamma_n$ from transmission
  - External: 4.9 eV of $^{197}$Au+n (saturated)
Comparison with LANSCE (Jandel et al.)

<table>
<thead>
<tr>
<th>$E_r$ / eV</th>
<th>$\Gamma_\gamma$ / meV</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>GELINA</strong></td>
<td><strong>LANSCE</strong></td>
</tr>
<tr>
<td>0.30605 ± 0.000014</td>
<td>0.3051 ± 0.0002</td>
</tr>
<tr>
<td>0.57387 ± 0.00026</td>
<td>0.5724 ± 0.0003</td>
</tr>
<tr>
<td>1.27106 ± 0.00058</td>
<td>1.2718 ± 0.0004</td>
</tr>
</tbody>
</table>

**GELINA**:
- Flight path length traceable to $E_r = 6.6735 \pm 0.0030$ 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
• Impact of target properties and importance of target characterization
• Transmission and capture yield (counts) are fully consistent
  ⇒ in a simultaneous analysis of $T_{\text{exp}}$ and $Y_{\text{exp}}$ 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 $^{197}\text{Au} + n$
• Determination of $^{241}\text{Am}$ quantity by calorimetry ($\delta n/n < 1.0 \%$)
  ⇒ $\sigma(n_{\text{th}}, \gamma)$ and $(E_{r}, g\Gamma_{n}, <\Gamma>)$ up to ~ 300 eV
Thank you for your attention 😊