Cross section measurements of minor actinides at the n_TOF-Ph2 experiment at CERN

Daniel Cano Ott on behalf of the n_TOF collaboration

daniel.cano@ciemat.es
Nuclear data for the transmutation of nuclear waste

Transmutation of the Minor Actinides by \((n,f)\) and \((n,\gamma)\) in new nuclear systems, thus reducing:

- radiotoxicity inventory \(1/100\)
- cooling time \(1/1000\)

Computational design tools

Need of accurate & reliable NUCLEAR DATA
**Nuclear data:** compilation of experimental and/evaluated data describing nuclear properties.

- **Differential and average reaction cross sections**
- Particle emission probabilities and energies (γ, e-, α…)
- Nuclear structure data (half lives, isomers…)

**Needs from various fields and applications:**

- **Transmutation of Nuclear Waste**
- **Nuclear reactor design and safety assessment**
- Hadron therapy
- Dosimetry (space, aircrafts, linacs)
- Shielding design: accelerators, power plants, hospitals…
Microscopic measurements

Integral experiments

Design of Nuclear Systems

Evaluation
Are the nuclear data accurate enough?

How accurately can we answer to the question:

What is transmutation rate of $^{241,243}$Am by fission inside an Accelerator Driven System or a fast reactor?

Lets have a look at the evaluated cross section databases:

• ENDF
• JENDL
• JEFF
• BROND, CENDL,…
$^{241}\text{Am}(n,f)$ cross section data
$^{241}\text{Am}(n,\gamma)$ cross section data
$^{243}\text{Am}(n,\gamma)$ cross section data

No data available for the evaluation of the capture reaction channel.
The reality about Minor Actinide cross sections

- In many cases there is insufficient accuracy (>10%).
- The systematic uncertainties are sometimes larger than indicated.
- There exist significant differences between evaluated data libraries.
- Data for some isotopes/reaction channels are missing.
- Scarce or non-existent covariance data, sometimes “guessed” in absence of experimental information.

Which isotopes, reaction channels and energy ranges are prioritary in the different scenarios?

Result of the available (and ongoing) sensitivity analyses and list of recommendations.
### Table: ABTR, SFR, EFR, GFR, LFR, ADS: Uncertainty Reduction Requirements Needed to Meet Integral Parameter Target Accuracies

<table>
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<tr>
<th>Isotope</th>
<th>Cross-Section</th>
<th>Energy Range</th>
<th>Uncertainty (%)</th>
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<th>Cross-Section</th>
<th>Energy Range</th>
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*2007 Symposium on Nuclear Data*
*November 29 - 30, 2007*
*RICOTTI Convention Center, Tokai, Ibaraki, Japan*

by Aliberti, Oct. 2007
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<td>Thermal 6.07 MeV</td>
<td>1.50%</td>
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Where can these cross sections be measured?
Available neutron time of flight facilities worldwide

I. Photoproduction neutron sources
(GELINA – Europe, ORELA & RPI – USA) + reactors (KURRI - Japan…):

- Good energy resolution.
- Low intensity per accelerator pulse / high repetition rate -> low duty cycle.
- Need of “massive” samples (several 100 mg) -> STABLE ISOTOPES

II. Spallation neutron sources
(LANSCE-LANL - USA and n_TOF @ CERN - Europe)

- Intrinsically worse energy resolution.
- High intensity at even low repetition rates -> high duty cycle, favorable reaction rate to decay rate ratios, even for high intrinsic activities (1 GBq).
- Samples can have masses as low as 1 mg.
(A Google-view of) The n_TOF facility at CERN

n_TOF
185 m
flight path

Pb Spallation
Target

Proton Beam
20GeV/c
7x10^{12} ppp

Neutron Beam
10° prod. angle

Booster
1.4 GeV

Linac
50 MeV

PS
20GeV
The n_TOF collaboration


Funded by the EC 5th Framework programme, CERN and National Funding Agencies.
**n_TOF beam characteristics**

The n_TOF facility was built and commissioned in a period of 2 years and provides unique features for measuring capture and fission cross sections of unstable (and also stable) isotopes.

- **White neutron beam**: 0.1 eV up to 1 GeV
- **ΔE/E ~ 1·10^{-4} in the RRR**

Capture and fission beams:
- **Φ=4 cm beam**
- **Φ=8 cm beam**

A fully digital DAQ!
Pulse shape fitting, particle discrimination and pileup reconstruction for $C_6D_6$ detectors. C. Guerrero et al, submitted to NIM-A.

Pulse shape analysis and pileup reconstruction for the $\text{BaF}_2$ detectors. E. Berthomiueux et al. To be submitted to NIM-A.
Advanced detectors
n_TOF fission detectors

Position sensitive Parallel Plate Avalanche Chambers. Allow to reconstruct the trajectories of the fission fragments.
Tassan-Got et al. To be submitted to NIM-A

Fast induction chamber (FIC). Large number of samples inside a compact detector.
P. Cennini et al. Submitted to NIM-A
Nuclear structure: TAC as a γ-ray spectrometer

Analysis of the calorimeter data and comparison to realistic Monte Carlo simulations of its response to the EM cascades.
The n_TOF Total Absorption Calorimeter (TAC) for (n,γ) measurements

- 40 BaF₂ crystals covering 95% of 4π.
- 98% detection efficiency for capture γ-ray cascades.
The experimental programme
n_TOF experiments 2002-4

**Capture**

- $^{151}\text{Sm}$
- $^{204,206,207,208}\text{Pb},^{209}\text{Bi}$
- $^{232}\text{Th}$
- $^{24,25,26}\text{Mg}$
- $^{90,91,92,94,96}\text{Zr},^{93}\text{Zr}$
- $^{139}\text{La}$
- $^{186,187,188}\text{Os}$
- $^{233,234}\text{U}$
- $^{237}\text{Np},^{240}\text{Pu},^{243}\text{Am}$

**Fission**

- $^{233,234,235,236,238}\text{U}$
- $^{232}\text{Th}$
- $^{209}\text{Bi}$
- $^{237}\text{Np}$
- $^{241,243}\text{Am},^{245}\text{Cm}$

- **Measurements of neutron cross sections relevant for Nuclear Waste Transmutation and related Nuclear Technologies**
  - Th/U fuel cycle (capture & fission)
  - Transmutation of MA (capture & fission)
  - Transmutation of FP (capture)

- **Cross sections relevant for Nuclear Astrophysics**
  - s-process: branchings
  - s-process: presolar grains

- **Neutrons as probes for fundamental Nuclear Physics**
  - Nuclear level density & n-nucleus interaction
(n,f) cross sections

Measurements with PPACs.
C. Paradela et al. In preparation
Fission measurements with the Fission Ionization Chamber (FIC) – 2/2

238U/235U: both isotopes are fission standards up to 200 MeV.

241Am has 239Pu contaminant which significantly contributes to the total σ.

245Cm: poor previous experimental results

Statistical error bars
Systematic errors ~8%

Ratio with FIC2 fission can be extracted up to 300 MeV.

PRELIMINARY

PRELIMINARY
(n,γ) cross section measurements with C₆D₆ detectors

Resolved Resonance Region

Unresolved Resonance Region

level spacing

reduced neutron width distribution

mass (²³²Th) = 2.8037 g
diameter = 1.5 cm
purity = 99.5%

204, 206, 207\text{Pb}, \ 209\text{Bi} (n,\gamma) \text{ measurement}

C.Domingo-Pardo et al. (n_TOF Collaboration), Phys. Rev. C 74/75, 2006/7
$^{237}$Np (n,γ) measurement with the TAC

43.3 mg, 1.29 MBq

\( ^{240}\text{Pu} \ (n,\gamma) \) measurement

51.2 mg, 458 MBq

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<tr>
<th>( \langle D_0 \rangle \text{ (eV)} )</th>
<th>ENDF/B-VII</th>
<th>( \text{n_TOF} )</th>
<th>Bouland et al.</th>
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<td>12.1 0.1</td>
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<td>( \langle \Gamma_\gamma \rangle \text{ (meV)} )</td>
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<td>( S_0(10^{-4}) )</td>
<td>1.13 0.08</td>
<td>1.07</td>
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$^{243}\text{Am} (n,\gamma)$ measurement with the TAC

**First (n,\gamma) measurement EVER. 10 mg, 75 MBq**

233, 234\(^{\text{U}}\) (n,\(\gamma\)), 233, 234, 235\(^{\text{U}}\)(n,f) measurement

Neutron induced capture

\[ 234\text{U}(n,\gamma) \]

- Cross section (b)
- Neutron energy (eV)

Neutron induced fission

\[ 234\text{U}(n,f) \]

- ENDF/B-VI
- n_TOF (PPACS)

W. Dridi, PhD-Thesis, 2006

C. Paradela, PhD-Thesis, 2005
Future plans
### Capture

**Stable Isotopes:**
- Mo, Bi, Ru: r-process residuals
- Fe, Ni, Zn, Se: s-process and structural materials

**Radioactive Isotopes:**
- $^{234,236}\text{U}$, $^{231,233}\text{Pa}$: Th/U fuel cycle
- $^{239,240,242}\text{Pu}$, $^{241,243}\text{Am}$, $^{245}\text{Cm}$: transmutation of minor actinides (FP-6 project IP-EUROTRANS/NUDATRA)

### Fission

- $^{231}\text{Pa}$, $^{234,235,236,238}\text{U}$
- $^{241}\text{Pu}$, $^{245}\text{Cm}$, $^{241,243}\text{Am}$, $^{244,245}\text{Cm}$
- $^{234}\text{U}$: study of vibrational resonances below the barrier
Angular correlations between the fission fragments

neutrons

C3F8 @ 6 mbar

600 μg/cm² Al backing

θ

300 μg/cm²

220 μg/cm² Mylar
Making better measurements at n_TOF-Ph2

Daniel Cano Ott – Information Exchange Meeting on Partitioning and Transmutation, Mito - Japan, October 2008
Improvements of the future measurements

1) $^{240}\text{Pu}(n,\gamma)$ data + 2) + 3) empty Ti canning + 3) kapton windows + sample frame

Background due to the sample container
Window surrounded by neutron absorber ($^{10}$B or $^{6}$Li doped polyethylene)

Thin Al backing + actinide sample

Neutron absorber

Calorimeter

Improved TAC setup
Agreement with VNIEE – Sarov and the Kurchatov Institute.
The (future) second n_TOF experimental area

New experimental area at 20 m

Experimental area at 185 m

Flight-path length: ~20 m
at 90° respect to p-beam direction
50 times more intense!
Summary and conclusions

• Differential neutron cross sections for Minor Actinides need to be improved.

• n_TOF @ CERN is a unique facility for it:
  • High performance detectors for fission (PPAC and FIC) and capture (TAC) cross section measurements
  • Fully digital Data Acquisition System.
  • Already proven for Np & Pu!
  • 1 mg mass samples of highly radioactive materials.

We are ready to do so: the n_TOF operation was interrupted in 2004 and will restart in 2008 (3rd of November)!
A brief history of the n_TOF facility

- 1997 - Concept by C.Rubbia validated by TARC exp. [CERN/EET/Int. Note 97-19]
- May ’98 - Further development of the initial idea towards a working facility [CERN/LHC/98-02+Add]
- 1999 – Construction started
- Oct 2000 – First proton on the spallation target
- 2005 to 2007 – Shutdown due to the LHC startup + modifications of the spallation target
- March 2008. MoU signed by CERN.
- November 2008 – Start of the n_TOF-Ph2 commissioning.
The n_TOF facility
The n_TOF data acquisition system

n_TOF has been the first neutron beam line world-wide proposing, building and operating a fully digital DAQ. Nowadays, it is becoming a standard at every laboratory.

The n_TOF DAQ consists of ~50 flash ADC channels with 8 bit amplitude resolution and sampling of 500 MSample/s.

The full history of EVERY detector is digitised during a period of 16 ms (0.7 eV < E_n < 20 GeV) and recorded permanently on tape.

The system has nearly zero dead time.

Simple electronics but everything needs to be done by software.
The new spallation target commissioning

Designed by CERN and CIEMAT

protons
Better experimental conditions

Photon time distribution (E>1MeV)

![Graph showing photon time distribution](image)

- **H2O (Neutron Flux)**
- **D2O**
- **H2O+1% B**
- **H2O+0.2% B**

**Time (ns)**: $10^{-1}$ to $10^5$

**Photon Flux at 182m** (dndt/dn/cm² / 7 x 10¹²pr)

**Energy (eV)**: $10^8$ to $10^2$
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Reaction</th>
<th>Energy range</th>
<th>Original Uncertainty (%)</th>
<th>Required accuracy (%)</th>
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Revisit the resonance capture in $^{235}$U, $^{238}$U