Neutron capture and fission reactions on $^{235}$U: cross sections, $\alpha$-ratios and prompt fission $\gamma$-rays

C. Guerrero and E. Berthoumieux  
CERN (Geneva, Switzerland)

D. Cano-Ott, E. Gonzalez and E. Mendoza  
CIEMAT (Madrid, Spain)

M. Sabate  
Universidad de Sevilla (Seville, Spain)

(The n_TOF Collaboration, www.cern.ch/nTOF)
Motivation: capture cross sections and $\alpha$-ratios

The criticality of current and fast future reactors must be known within 0.3-0.5% for operation/safety.

(FCA) Fast Critical Assembly (JAEA)

GOAL: Measure $^{235}\text{U} \sigma(n,\gamma)$ below 2.5 keV

Differences up to 2% in the measured and calculated criticality values for FCA (JAERI, Japan) assemblies with different hardness are due to $^{235}\text{U} \sigma(n,\gamma)$ below 2.5 keV.
Motivation: prompt fission $\gamma$-rays

**IMPACT**
The four fast reactor systems of GenIV feature innovative core characteristics for which gamma-ray heating estimates for non-fuel zones require an uncertainty of 7.5%. A similar requirement appears for the experimental Jules Horowitz Reactor (RJH) at Cadarache. Recent studies show evidence of discrepancies on integral measurement in MASURCA, EOLE and MINERVE, from which it is clear that the expectations for GenIV systems and the RJH thermal reactor are not met. Gamma-ray energy release is dominated by $^{239}$Pu and $^{235}$U.

**ACCURACY**
- **Observed**: Discrepancies for C/E ratios in various benchmarks range from 10 to 28%.
- **Target**: 7.5% on the total gamma energy and multiplicity
- **Target**: Best accuracy achievable for the gamma spectrum shape

**COMMENT FROM REQUESTER**
Forty percent of the total gamma-ray energy release results from prompt decay of fission products. No comprehensive analytic expressions exist and Hauser-Feshbach model calculations are involved and presently lack sufficient knowledge to warrant a solution of the problem. New measurements would be needed to guide new evaluation efforts. Present evaluations are based on measurements from the seventies.
Measuring technique

Thermal-epithermal neutrons induce both \((n,\gamma)\) and \((n,f)\) reactions, both emitting \(\gamma\)-rays

**MEASURING THE NEUTRON CROSS SECTIONS & \(\gamma\)-RAY EMISSION REQUIRES**

- A **facility** providing a neutron beam (The n_TOF facility).
- A highly pure **sample**.
- A detection system for **detecting simultaneously fission fragments and \(\gamma\)-rays**
- The **analysis tools** to determine the measured cross sections with the required accuracy.
A Google view of n_TOF

TECHNICAL PAPERS ON n_TOF’s:

NEUTRON FLUENCE, PROFILE AND RESOLUTION:
- NIM-A 513 (2003) 524-537

DATA ACQUISITION SYSTEM (FULLY BASED IN FLASH-ADC)
- NIM-A 538 (2005) 692-702

DETECTION SYSTEMS
- NIM-A 608 (2009) 424-433

More information at www.cern.ch/nTOF
Experimental set-up: The TAC and MGAS detectors

We need to detect capture and fission reactions simultaneously!

**Total Absorption Calorimeter (TAC) for** $(n,\gamma)$
- 40 BaF$_2$ crystals
- $4\pi$ geometry (95% coverage)
- 16% energy resolution at 662 keV
- Used for $\sigma(n,\gamma)$ of actinides since 2004

**MicroMegas (MGAS) for (n,f)**
- Based on Bulk technology
- Double stage gas detector: conversion +amplification
- ~90% efficiency for FF. FF.
- Used for neutron monitoring since 2009

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**Results:** distributions $E_{\text{sum}}$, $m_{\text{cr}}$ & $E_n$

C. Guerrero et al., NIM-A 608 (2009) 424-433

**Results:** distributions Amp. & $E_n$

S. Andriamonje et al., NIM-A 481 (2002) 120–129

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C. Guerrero et al. @ WONDER-2012 Aix-en-Provence (France)
Experimental set-up (2012): Combination of the TAC and MGAS

- 10 $^{235}$U samples of 300 $\mu$g/cm$^2$ (42 mm diameter)
- MGAS filled with Ar/CF$_4$/isobutane at 1 atm
- TAC and MGAS signals digitized at 250 MS/s and 100 MS/s, respectively.

10 MGAS detectors (5 back-to-back assemblies) each equipped with a 4.15 mg $^{235}$U sample (supplied by JRC-IRMM)
Experimental set-up (2012): Combination of the TAC and MGAS

- MGAS signals
- Neutrons
- BaF$_2$ module
- Neutron absorber
- MGAS with $^{235}$U samples
Validation of simultaneous measurement of capture and fission reactions at n_TOF

Letter Of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Spokespersons: C. Guerrero\textsuperscript{1} and E. Berthoumieux\textsuperscript{2}
Technical coordinator: V. Vlachoudis\textsuperscript{3}

C. Guerrero\textsuperscript{1}, E. Berthoumieux\textsuperscript{2}, S. Andriamonje\textsuperscript{2}, D. Cano-Ott\textsuperscript{2}, E. Gonzalez-Romero\textsuperscript{2}, F. Gusing\textsuperscript{2}, T. Martinez\textsuperscript{2}, E. Mendoza\textsuperscript{1}, M. Calviani\textsuperscript{3} and The n_TOF Collaboration (\url{http://cern.ch/n_TOF/})

\textsuperscript{1}Centro de Investigaciones Energéticas Medioambientales y Tecnológicas – Ciemat, Madrid, Spain
\textsuperscript{2}CEA Saclay, IRFU, F-91191 Gif-sur-Yvette, France
\textsuperscript{3}CERN, Geneva, Switzerland
Measurement (2010): $E_{\text{sum}}$ and $m_{\text{crystal}}$ distributions

Deposited energy ($m_{\text{cr}}>2$) and multiplicity ($E_{\text{sum}}>3$) distributions corresponding to resonances:

\[ S_n^{(236U)} \sim 6.5 \text{ MeV} \]
Measurement (2010): Detection efficiencies

With two different detectors and two different types of reactions to detect, it is important to define clearly the different efficiencies that play a role in the measurement and their interrelations.

\[ \varepsilon_{\text{MGAS}}(n,f), \varepsilon_{\text{TAC}}(n,f) \text{ and } \varepsilon_{\text{TAC}}(n,\gamma) \]

When a fission reaction occurs, it can be detected:

a) in both detectors, \( \rightarrow \varepsilon_{\text{MGAS}}(n,f) \cdot \varepsilon_{\text{TAC}}(n,f) \)

b) in none of them, \( \rightarrow (1 - \varepsilon_{\text{MGAS}}(n,f)) \cdot (1 - \varepsilon_{\text{TAC}}(n,f)) \)

c) only in the MGASs \( \rightarrow \varepsilon_{\text{MGAS}}(n,f) \cdot (1 - \varepsilon_{\text{TAC}}(n,f)) \)

d) only in the TAC. \( \rightarrow (1 - \varepsilon_{\text{MGAS}}(n,f)) \cdot \varepsilon_{\text{TAC}}(n,f) \)

When a neutron capture occurs, it can only be detected in the TAC \( \rightarrow \varepsilon_{\text{TAC}}(n,\gamma) \)

The efficiency for detecting fission reactions in each detector is independent from the other, but the calculation from experimental data requires that these four probabilities are properly taken into account.
Measurement (2010): $\varepsilon_{\text{MGAS}}(n,f)$, $\varepsilon_{\text{TAC}}(n,f)$ and $\varepsilon_{\text{TAC}}(n,\gamma)$

Calculation of $\varepsilon_{\text{MGAS}}(n,f)$

**MC simulations**
Samples are 318 $\mu$g/cm$^2$, nearly identical to those of the $^{235}$U samples (316 mg/cm$^2$) used in FIC, for which simulations with FLIKA give $\varepsilon_{\text{MC}}(n,f)\approx0.94$ (6% losses due to absorption in the sample).

**Experimentally:**
Fission events produce high-energy, high-multiplicity TAC events. Assumption $\rightarrow$ $\varepsilon_{\text{TAC}} \approx 100\%$ for such events. Then, the detection efficiency of the MGASs can be calculated as the ratio of tagged to all events for multiplicities higher than $\sim$10 (no capture events).

![Graph showing the efficiency of detection](image)

$\varepsilon_{\text{MC}}(n,f)\approx0.94 \& \varepsilon_{\text{exp}}(n,f)\approx0.90$

$\varepsilon_{\text{MGAS}}(n,f)\approx0.92$
A coincident event in the TAC is found for 97% of the MGAS events (MGASamp>20 channels). This value represents the TAC efficiency for fission events, $\varepsilon_{\text{TAC}}(n,f)$, and is very similar to the efficiency of $\varepsilon_{\text{TAC}}(n,\gamma)=0.974(4)$ for capture events in $^{197}$Au (from GEANT4 Monte Carlo simulations).

The efficiency $\varepsilon_{\text{TAC}}(n,f)$ depends on the analysis conditions for the deposited energy and multiplicity values.

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Measurement (2010): $\varepsilon_{MGAS}(n,f)$, $\varepsilon_{TAC}(n,f)$ and $\varepsilon_{TAC}(n,\gamma)$

Calculation of $\varepsilon_{TAC}(n,\gamma)$

The detection efficiency $\varepsilon_{TAC}(n,\gamma)$ can be calculated accurately by means of Monte Carlo simulations when both the experimental set-up and the details of the capture cascades are properly considered.

- Already done for $^{237}\text{Np}$, $^{240}\text{Pu}$, $^{241,243}\text{Am}$, and $^{233}\text{U}$.
- $^{235}\text{U}(n,\gamma)$ still to be done

Approximation:

$^{235}\text{U}$ is very similar to $^{237}\text{Np}$
- Odd nuclei
- Similar level spacing ($\sim 0.5$ eV)
- Similar Binding Energies ($S_n \sim 6$ MeV)
- Cut at 2.5 MeV ($0.46*S_n$) is to $^{237}\text{Np}$ like 3 MeV is to $^{235}\text{U}$

$$\varepsilon_{TAC}(n,\gamma)=0.70(3)$$

[$E_{\text{sum}}>3$ MeV and $m_{ct}>2$]
Test of TAC+MGAS with $^{235}\text{U}@ n\_\text{TOF}$

- Discrimination $(n,\gamma)$ vs. $(n,f)$
- Normalization to $\sigma(n,f)$
- Efficiency correction
- Background subtraction
- Identification of impurities

Agreement with evaluations at low $E_n$

Measurement (2010): Results and publication
Data taking ongoing at CERN !!!

**Compared to 2010 test measurement:**
10 samples of 4.15 mg each, instead of 3 samples of 1 mg each → **x10 in mass**
Samples 42 mm in diameter instead of 20 mm → **full beam coverage**
Configuration with neutrons absorber → **x0.2 in neutron scattering background**
More beam time, 9 weeks instead of 1 → **x9 statistics**
Very Preliminary

**σ(n,γ)/(n,f) measurement (2012): deposited energy distributions**

**TAC (Mult>2) (Resonances below 20 eV)**

Counts (norm. to $^{235}$U protons)

- All
- Fission
- Backg. ($\times 0.95$ & shifted -40 keV)
- All-Fission
- All-Fission-Backg.

Deposited Energy (keV)
$\sigma(n, \gamma)/(n,f)$ measurement (2012): neutron energy distributions
σ(n, γ)/(n,f) measurement (2012): neutron energy distributions

C. Guerrero et al. @ WONDER-2012  Aix-en-Provence (France)
σ(n, γ)/(n,f) measurement (2012): neutron energy distributions

Very Preliminary

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σ(n, γ)/(n,f) measurement (2012): neutron energy distributions

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$\sigma(n, \gamma)/(n,f)$ measurement (2012): neutron energy distributions
Prompt $\gamma$-rays from fission in $^{235}$U

The $4\pi$ BaF$_2$ Total Absorption Calorimeter (TAC) provides information on the multiplicity and energy of the prompt $\gamma$-ray emission following fission reactions.
Prompt $\gamma$-rays from fission in $^{235}$U

The $4\pi$ BaF$_2$ Total Absorption Calorimeter (TAC) provides information on the multiplicity and energy of the prompt $\gamma$-ray emission following fission reactions.

**PLAN**

Direct measurements of $\gamma$-ray emission

+ Benchmark models through:
  $\gamma$-rays models + simulation & comparison to TAC data

*C. Guerrero et al., NIM-A 671 (2012) 108-117*
Prompt $\gamma$-rays from fission in U & Pu

Independently of n_TOF, a new campaign will take place in 2013 for measuring prompt fission $\gamma$-rays from $^{233}\text{U}$, $^{235}\text{U}$, $^{239}\text{Pu}$ and $^{241}\text{Pu}$ through the combination of:

- PBF1 cold neutron beam from ILL
- EXOGAM high resolution HPGe detector array
Conclusions and perspectives

Measuring the capture cross sections and prompt $\gamma$-rays of fissile isotopes is of upmost importance for the development of present and future (ADS & Gen-IV) nuclear reactors.

**FOLLOWING A SUCCESSFUL TEST, A NEW EXPERIMENT IS RUNNING FOR MEASURING:**

- Capture cross section in the Resolved Resonance Region (RRR) [below 2.25 keV]
- Resonance parameters in the full RRR
- Alpha ratio in the full RRR
- Prompt fission $\gamma$-rays @thermal and as function of $E_n$

Preliminary results from this very fresh data (now being collected) will be presented @ ND-2013 (New York, March 4-8 2013)

*D. Cano-Ott et al., Measurement of the neutron capture cross section of the fissile isotope $^{235}$U with the CERN n_TOF Total Absorption Calorimeter and a fission tagging based on MicroMegas detectors*