# Delayed neutrons: measurements and usage

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## Short Curriculum Vitae

#### 1992-1995 : PhD Thesis at GANIL

 Formation and de-excitation hot nuclei produced in reactions induced by proton (475MeV and 2 GeV) and <sup>3</sup>He (2 GeV) beams

#### 1996-1998 : Post-doctoral position at CEA/DIF (Bruyères-le-Châtel)

Measurement of neutron emission in spallation reaction between 800 MeV and 1600 MeV

#### 1998-2012 : Permanent position at CEA/DIF (Bruyères-le-Châtel)

- n,xn reaction studies
- Production of isomeric nuclei by neutron capture in the resonances
- Delay neutrons measurements in fission induce by neutrons and photon
- Spokesperson of the Neutrons For Science Facility

#### Since 1992 : Permanent position at GANIL

Responsible of the Neutrons For Science facility at GANIL/SPIRAL-2



X. Ledoux, ICTP school lecture, 19-30 oct 2015

#### **GANIL-Spiral2** facility



#### **Neutrons For Science facility: intense fast neutron time-of-flight facility**

## Delayed neutrons: measurements and usage

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- I. Definition and characteristics
  - I. Yield
  - II. Time distribution
- II. Applications :
  - I. Active interrogation techniques
  - II. Reactor
- III. Measurement
  - I. Neutrons detectors for DN
  - II. Microscopic measurements
    - I. Techniques
    - II. Examples of experiments
  - III. Macroscopic measurements
    - I.  $v_d$  measurement
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#### □Nuclear power control and safety:

Some fission products undergo Beta Delayed Neutron Emission which is essential to control the reaction.

#### □Interrogation techniques :

Some active interrogation techniques are based on the detection of delayed neutrons and/or photons. It is the signature of the presence of actinide.

#### **Q**Rapid neutron-capture process of stellar nucleosynthesis:

Short and very high neutron flux produces very neutron rich nuclei in short time.  $\beta$  decay determines the speed of the process.  $\beta$ -n shapes the abundance of the distribution



#### **Nuclear Structure:**

Additionally the measured half-lives ( $T_{1/2}$ ) and  $\beta$ -delayed neutron-emission probabilities (Pn) can be used as first probes of the structure of the  $\beta$ -decay daughter nuclei in this mass region.

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#### Beta – n decay



For enough neutrons rich nuclei,  $Q_b > B_n$ 

- If the decay proceeds to states above  $B_n$ : neutron emission dominates over  $\gamma$ -ray de-excitation
- Emission of a  $\beta$ , a neutron and sometimes photons
- These nucleus are called precursors (more than 300 exist)
- P<sub>n</sub> probability that a precursor emits a neutrons
- The emission time distribution follows the precursor half-life

#### Neutron rich nuclei



Far from stability,  $\beta$ -delayed neutron emission becomes the dominant process

#### Neutrons emission in fission



- After a fission induced by n or  $\gamma$ , 2 large excited fragment are emitted
- They de-excite by neutrons and photons emission (prompt neutrons and photons)
- Leading to fission products, stable or radioactive
- The radioactive fission products de-excite mainly by Beta decay

Number of prompt neutrons (per fission)  $\upsilon or \upsilon_p$ 2 to 4Number of delayed neutrons $\upsilon_d$ 0.1 to 5% $\beta$  fraction of delayed neutrons $\beta = v_d / v_p$ 

#### Neutron induced fission



Fission fragments are neutron rich nuclei  $\rightarrow \beta$  and  $\beta$ -n decay

#### **Delayed Neutron characteristics**

More than 300 precursors exist after a fission

- $\odot$  Life time between 10 ms and 1 min
- $\circ$  DN yield: from 0,1 to 5 per 100 fissions
- Usually merged in groups depending of the precursor half-live

	v delayed	v prompt	β=vd / vp
239-Pu	0,0061	2,87	0,0021
238-U fast	0,0450	2,84	0,01584
241-Pu	0,0154	3,14	0,0049
235-U thermal	0,0166	2,43	0,00683



Energy spectrum softer than prompt fission neutrons:

Delayed Neutrons E<1,5 MeV

Prompt neutrons <E>≈ 2 MeV

#### Groups of delayed neutrons

Due to the number of precursors, the yield and time distribution can not be treated individually
Groups depending of the time decay are defined.

□ The delayed neutrons are characterized by :

- the yield (per/fission)
- the time distribution depending on the half live of the precursors





- Depending on the library and applications : 5,6 or 8 groups have been defined.
- In case of six groups 12 parameters allow characterizing the yield and time distribution

## DN of <sup>239</sup>Pu and <sup>235</sup>U neutron induced fission



Sometime 1 dominant precursor for the group

G. R. Keepin et al, PR107, vol 107, num 4, 1957

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## Actinide detection

Goal : detection of the presence of actinide in a container, waste,....

#### Safeguard

- detection of nuclear weapon
- nuclear material traffic
- dirty bomb

Monitoring the processes in nuclear fuel reprocessing

#### Uwaste management :

➢ presence of actinide in nuclear industry waste

>verification of "old" container or concrete bloc

1. Passive detection gamma detection -> suspicious object



#### 2. Radiography

The detection of high density material gives the area to investigate

#### 3. Active interrogation

Pulsed radiation fields can investigate time-correlated signatures which passive signature techniques generally cannot



# Active interrogation technique

- 1. The "package" is irradiated by neutrons or photons
  - Emission of prompt neutrons and photons
  - Detection difficult : background induced by the beam
- 2. Neutrons and/or photons time distributions are measured after the irradiation
  - Delayed neutrons detection: very probably emitted by fission fragments
  - Yield and time dependence measurement : identification and mass of the actinide in the package
  - Delayed photons are also used in addition to DN



Simulations of the whole set-up including attenuation of induced and emitted neutrons and photons



Relevant data and especially accurate data of  $a_i\,\lambda_i$  for all actinides

#### **Reactor control**

#### Nuclear reactor run with chain reaction :



K<sub>eff</sub> nb of neutrons inducing a fission from one neutron

 $K_{eff} < 1$  subcritical  $K_{eff} > 1$  supercritical

K<sub>eff</sub>=1 in reactor

<sup>235</sup>U + neutron = 2,43 prompt neutrons + 0,0166 delayed neutrons (β=0,00685) <sup>239</sup>Pu + neutron = 2,91 prompt neutrons + 0,0061 delayed neutrons (β=0,0021)

Reactivity:  $\rho = \frac{k_{eff} - 1}{k}$ 

#### Neutrons generation time

• Average generation time  $\Lambda =$ 

time between the birth of two fission neutrons in successive generations

• Population :

$$N(t+\Lambda) = k_{eff} N(t) \qquad \frac{dN(t)}{dt} = \frac{N(t+\Lambda) - N(t)}{\Lambda} = \frac{k_{eff} - 1}{\Lambda} N(t) \qquad N(t) = N(0) \exp\left(\frac{k_{eff} - 1}{\Lambda}t\right)$$

- Prompt neutrons
  - ➢ Fast reactors Λ<sub>prompt</sub>≈1µs ➤Thermal reactor

<sup>235</sup>U n induced fission  $\Lambda_{delayed} \approx 12,5 \text{ s}$ Delayed neutrons:

$$\Lambda_{average} = \Lambda_{prompt} (1 - \beta) + \Lambda_{delayed} (\beta)$$

Neutrons multiplication in 1 sec in the case of PWR and <sup>235</sup>U with k=1,0001  $\beta$ =0,0065  $\rightarrow \Lambda_{average} = 25.10^{-6} \text{ s x } (1-0,0065) + 12,5 \text{ s x } 0,0065$  $\Lambda_{average} = 0,0813 \text{ s}$ 

- Only prompt neutrons :  $\Lambda_{\text{prompt}} \approx 25 \mu s$ 

$$\frac{N(t=1s)}{N(0)} = \exp\left(\frac{1,0001-1}{25.10^{-6}}\right) = \exp(4) = 55$$

- With delayed neutrons  $\rightarrow \Lambda_{average} = 0.0813 \text{ s}$ 

$$\frac{N(t=1s)}{N(0)} = \exp\left(\frac{1,0001-1}{0,0813}\right) = \exp(0,00123) = 1,00123$$

Delayed neutrons are responsible for the ability to control the rate at which power can rise in a reactor. If only prompt neutrons existed, reactor control would not be possible due to the rapid power changes

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Vd/Vtot

1.89%

0.22%

0.13%

0.13%

When actinide mass increases :

•Yield of important delayed neutrons emitters (Br) decreases

β decreases

Number of prompt fission neutrons increases



□ Fast reactors are more nervous than thermal ones
□ When Pu, Am or Cm ∧ in the fuel β ∨

Valeurs des proportions de neutrons retardés ( $\beta$  effectif) par isotope, pour un spectre neutronique rapide

Isotopes	eta effectif en pcm
U 235	670
U 238	1 680
Pu 239	220
Pu 240	270
Pu 241	490
Pu 242	640
Np 237	440
Am 241	110
Am 243	250
Cm 244	100
Cm 245	130

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# Liquid scintillator

- Cell of liquid scintillator coupled to a phototube
- Detection :
  - diffusion (n,p)
  - scintillation
  - light conversion to electric signal by the phototube
- Sensitive to gamma, neutrons and charged particles
- neutron-  $\boldsymbol{\gamma}$  discrimination by pulse shape analysis
- Fast detector ( $\simeq 1 \text{ ns}$ )
- $\boldsymbol{\cdot}$  The intrinsic efficiency depends on the size of the cell
- Compromise between efficiency and energy resolution







## <sup>3</sup>He gas detector





 $\rightarrow$  The neutron must be slowed down to increase the reaction drop s-section

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#### $4 \pi$ neutrons detector

□ How to increase the detection ?

- The <sup>3</sup>He tubes are placed in a matrix of polyethylene
- Increase the number of tubes
- $\succ$  Use a  $4\pi$  geometry
- Slow detector
  - The slowing down process takes several hundredths of microseconds
  - Energy measurement by TOF technique impossible
  - Fast enough compare to precursors half-live

Designed to have an efficiency constant in the energy range of delayed neutrons



# **BELEN: Beta deLayEd Neutron detector**

Two crowns of (8+12) <sup>3</sup>He detectors embedded in a polyethylene matrix with total dimensions 90x90x80 cm<sup>3</sup> and a r=5cm beam hole







The thickness of CH2 and the position of the tube are optimized for an efficiency quite constant with En in below 1 MeV

# BRIKEN

International collaboration using a large number of 3He tubes of different size

#### BRIKEN stands for Beta delayed neutron measurements at RIKEN.





# 174 <sup>3</sup>He tubes of 6 different types:

Ring	Radius (cm)	# <sup>3</sup> He Tubes	Pressure (atm)	Diameter (inch)	Institute
1	9.4	14	10	1	ORNL
2	13	12+12	5.13	1	RIKEN
3	16.8	10+26	10/8	1	GSI/UPC
4	20	18+18	5/8	1.18/1	JINR/UPC
5	27	26	10	2	ORNL
6	35	38	10	2	ORNL

#### $\bullet$ High average efficiency of > 60 %

• Flat efficiency 6% up to 4 MeV, 12% up to 5 MeV.

C. Domingo-Pardo, BRIKEN Construction Proposal, NP-PAC, RIKEN Nishina Center

#### 13-14/12/2013

# **CEA Deleayed Neutrons detector**

#### Characteristics :

- efficiency  $\varepsilon > 20\%$  (sample in centre)
- constant efficiency between 0.1 and 1 MeV
- not sensitive to gamma
- Cylinder of  $CH_2$  ( $\Phi_{int}$ =6 cm,  $\Phi_{ext}$ =16 cm, L=37cm)
- 12 tubes <sup>3</sup>He
- Also used for photofission studies



#### Efficiency :



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# Microscopic measurement

#### The $\beta$ and the neutron are measured in coincidence

- Beta detection: thin target or implantation on Si detector
- Precursor identification :
  - ≻Spectrometry
  - Mass spectrometer
- Neutron detector : Low energy threshold
  - Large to ensure En measurement
  - > Possibility of  $\beta$ ,2n measurement
- Time stamping





# Example : experiment at GSI

U238 fragmentation on Be target -> production of large number of nuclei Implantation of fragments in Si detector



# Time-of-Flight technique

The measurement of the time between the beta and the neutrons allows to determine En



L : flight path (cm) t : time of flight ΔL : flight path resolution Δt : time resolution

Optimization of the energy resolution :

- short  $\Delta t$ : fast detectors ( $\beta$  and neutrons)  $\approx$  ns
- small  $\Delta L$ : thin detector -> reduced efficiency
- large L: small solid angle -> need to increase number or area of detector
- Drawback: Evaluation of Cross-talk between several detectors
- Advantage: Possibility of efficiency correction ε=f(E)
- Neutron detector not sensitive to photons or (n, γ) discrimination capability

→ Liquid scintillator or plastic scintillator



# Detector with plastic scintillator

- Detectors with a large number of plastic scintillator bars equipped with 2 Phototubes
- Neutron localization by the ratio of the PM signals
- Energy measurement by TOF



A. Buta et al., NIMA455, p412 (2000) 32 BC400 plastic scintillator 50% of  $4\pi$ E=30% at 2MeV,  $\delta$ E/E=10%

VANDLE Versatile Array of Neutron Detectors at Low Energy



Proceeding on 10<sup>th</sup> symposium on Nuclei in cosmos, 2008, Mackinac Island, Michigan, USA, Proceeding of Science

## Spectrometer with multiple cells

#### The DESPEC MOdular Neutron SpectromeTER

- •Cylindrical cell of 20 x 5 cm filled with BC501A/EJ301
- •Reasonable intrinsic efficiency (~50% @ 1MeV)
- •Energy threshold ~ 30 keVee (E<sub>n</sub> ~100 keV)
- •Reasonable energy resolution < 10% up to 5 MeV:
- Good neutron timing ~1ns
- Good β timing: < 4ns</li>
- Reasonable flight path 2-3 m TOF
- Good total efficiency: 150 200 detectors

Design similar to other projects (DESIR @ SPIRAL II)

200 de 10cm	tectors, radius	ΔE/E @ 1 MeV		
TOF distance (m)	Geometric efficiency	1ns	4ns	
2 12.5%		3.5%	6.0%	
3	5.6%	2.5%	4.2%	



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The beta and neutron are not detected in coincidence

□What is needed :

□ The fission rate:

- Calculated from
  - sample characteristics (mass and purity are requested)
  - flux and energy distribution (monitoring)
- Measured (detector based on fission of the same actinide)

A beam time cut-off quite fast (with respect to the shorter half life group)

#### Neutron detector(s)

- High efficiency
- Not sensitive to photons
- Efficiency independent of neutrons energy

 $4\pi$  neutrons detector

#### v<sub>d</sub> measurement

A sample of masse m is irradiated by a flux  $\phi$  of neutrons or photons

The fission rate:



σ(E) fission cross-sectionA atomic number of the actinide,

a<sub>i</sub> nb of DN yield of group i

The delayed neutrons rate of group i

- Emitted during the irradiation:  $Y_{n,i}(t) = R a_i (1 e^{-\lambda_i t})$
- During the decay after an irradiation time  $T_{irr}$ :  $Y_{n,i}(t) = R a_i (1 e^{-\lambda_i T_{irr}}) e^{-\lambda_i (t T_{irr})}$

• For all the groups : 
$$Y_n(t) = \sum_i R a_i (1 - e^{-\lambda_i T_{irr}}) e^{-\lambda_i (t - T_{irr})}$$

# $v_d$ measurement strategy ( $T_{irr} >> T_i$ )

 $\textbf{F}_n$  number of detected neutrons with efficiency  $\boldsymbol{\epsilon}$ 

$$Y_n(t) = \frac{F_n(t)}{\varepsilon}$$

If  $T_{irr} >> T_i$ , all the precursors are at equilibrium (infinite irradiation)  $1 - e^{-\lambda_i T_{irr}} = 1$ 

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# $v_d$ measurement strategy ( $T_{irr} \ll T_i$ )



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# **DN time distribution**



The irradiation – detection times must be adapted to the half life of the group to be measured

- Short irradiation < 1 s for groups 5 and 6
- Medium 10 s for groups 4 and 3
- Long irradiation for groups 1and 2

## Strategy for group parameters determination



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# Study of multi-neutron emission in the $\beta$ -decay of <sup>11</sup>Li

Lynda Achouri LPC Caen

for the IS525 Collaboration

XIXth COLLOQUE GANIL, October 12th-16th, Anglet

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# Experimental set up @ ISOLDE



# Experimental set up @ ISOLDE



# DN of <sup>235</sup>U and <sup>239</sup>Pu in n induced fission

Reactor GODIVA, Los Alamos, Instantaneous pulse 10<sup>16</sup> fissions



- no short time cut
- · detection set-up should be placed far away
- only thermal or fast neutron

nematic diagram of the experimental arrangement for delayed-neutron studies at Los Alamos.

G. Keepin et al., PRC 107, num 4 (1957)

#### Keepin's results



# DN yield measurement on <sup>232</sup>Th(n,f)

Goal : measurement of DN yield in neutrons induced fission on Th 232 between 1 and 16 MeV

- Facility : PIAF (PTB)
- Detector :  $4\pi$  <sup>3</sup>He + CH<sub>2</sub>
- Technique : pulsed beam 7s beam-1 s off
- Measurement of Y(t=0)
- •N<sub>fissions</sub> : flux measurement + MCNPX





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# DN yield measurement on <sup>232</sup>Th(n,f)

Corrections to be performed :

- Open source : some neutrons deflected by the detector can induce fission
- Thick target : fission induced by secondary fission
- Not infinite irradiation time



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## DN measurements in fission induced by photons



#### Repetetive cycles of irradiation and counting



# DN in photofission of <sup>238</sup>U



Differencies have been measured in comparison with data from Nikotin Very important for applications : identification by activation technique

D. Doré et al., ND2007, ACCAPP 2007

## Measurement of DN in spallation reaction

- 1 GeV p + thick lead target
- Detection of neutrons yield and time distribution
- Measurement of cross-section production of <sup>87</sup>Br, <sup>88</sup>Br, <sup>9</sup>Li and <sup>17</sup>N
- Comparison with spallation codes



# DN yield in liquid target (1)

- MegaPie target (PSI)
- Production of neutrons by spallation reaction in thick target
- 1 MW liquid metal target
- Loop of Pb-Bi liquid -> DN are emitted outside of the core of the target : displacement of the neutrons dose in the facility



DN flux at the top of the target is of the same order than prompt neutrons

# DN yield in liquid target (2)



## Summary

#### Delayed neutrons play an important role in several topics

- Control of fission reactors
- Active interrogation
- Astrophysics
- Nuclear structure

#### Microscopic measurements

- Pn of an identified nuclei
- Energy spectrum of the DN

□ Macroscopic measurements

- Delayed neutrons yield
- Time distribution
- Groups parameters