# Measurement of (n,xn) reactions

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### OUTLINE

### Introduction

- 1. Applications
- 2. Facilities
- 3. Cross-section measurements
  - 1. Activation technique
  - 2. n,xnγ gamma
  - 3. Direct measurement of secondary neutrons
- 4. Double differential measurements
- 5. Examples of experiments

### n,xn reactions

$$n + {}^{A}_{Z}X \rightarrow {}^{A-x+1}_{Z}X + x n$$

Production of an isotope

Same element (no chemical separation)

 $_{\odot}$  Can decay on other element ( $\beta$  decay....)

Threshold

 $\circ$  n,n' the energy of the first excited level  $\circ$  n,xn depends on the Q-value

$$Q = {}^{A-x+1}_{Z}M + xM_n - \left({}^{A}_{Z}M + M_n\right)$$

$$Q = \Delta(A-1,Z) + (x-1)\Delta n - \Delta(A,Z)$$

$$E_{th} = -Q \frac{M_n + M_X}{M_X} \cong -Q \frac{A+1}{A}$$

 $\Delta(A,Z)$  The mass excess of nuclei A,Z



	Ethres	Q
<sup>208</sup> Pb(n,2n) <sup>207</sup> Pb	7,403	-7,367
<sup>208</sup> Pb(n,3n) <sup>206</sup> Pb	14,174	-14,105
<sup>208</sup> Pb(n,4n) <sup>208</sup> Pb	22,299	-22,192

### **Reaction process**



(n,xn) reactions represent the main part of the reaction cross-section above MeV



#### Reactions on <sup>238</sup>U

## Role of the pre-equilibrium process in (n,xn) reaction

The pre-equilibrium process is an important process of the n,xn reactions





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## **Applications**

### □ Reactor and accelerator driven systems: n,xn contributes to

- Energy loss mechanism
- Neutron multiplication
- Production of radioactive isotopes
- □ Production of nuclear waste (fusion technology)

□ Nuclear data evaluation (all the channels have to be studied)

- Neutron field characterization
  - Fluence
  - Energy and angular distribution

Reliable evaluated data bases are request  $\rightarrow$  accurate measurements

### Neutron field characterization

Threshold reactions including (n,n') and (n,xn) reactions are used for determining the differential flux from neutron sources by activation techniques.

 $\Box$  Irradiation of a set of material i in a flux  $\Phi$  :

□ Measurement of the activity of each sample i :

$$A_{i}^{mes} = k \int_{E_{s}}^{E_{m}} \frac{d\Phi}{dE}(E_{n})\sigma_{i}(E_{n})dE_{n}$$

 $\Box \text{ Use a simulated neutron spectrum: } \psi(E) \text{ to calculate } A_i^{sim} = k \int_{E_s}^{E_m} \frac{d\Psi}{dE}(E_n) \sigma_i(E_n) dE_n$ 

 $\Box$  Adjust  $\psi(E)$  in order to minimize

$$\chi^2 = \frac{1}{m} \sum_i \frac{\left(A_i^{mes} - A_i^{sim}\right)^2}{\varepsilon_i^2}$$

The choice of the reactions depends on:

- Energy threshold in order to cover the energy range of interest
- Product of reaction measurable (radioactive, period, decay mode)
- Reaction (n,p), (n,α), (n,n'), (n,xn)

#### Accurate knowledge of the cross-section reactions is required



N. Jovančević\* et al., Physics Procedia 59 (2014) 154 – 159 Göran Lövestam et al., Radiation Measurements 44 (2009) 72–79

### Above 20 MeV

#### Experimental determination of neutron spectra produced by bombarding thick targets: (100 MeV/u) D + <sup>9</sup>Be, <sup>238</sup>U and (95 MeV/u) <sup>36</sup>Ar + <sup>12</sup>C

Experimental set-up

Incident beam	Energy (MeV/u)	Intensity (p.p.s)	Target	Irradiation time (min)	Activation detectors	Detector position
<sup>36</sup> Ar	95	6.42×10 <sup>11</sup>	Carbon	88	Al, Ni, Bi	0°, 20°, 45°, 90°
Deuterons	100	6.72×1010	Beryllium	373	Al, Ni, Co, Bi	0°, 11°, 36°, 60°, 84°
Deuterons	100	$6.47 \times 10^{10}$	Uranium	376	Al, Ni, Co, Bi	0°, 11°, 36°, 60°, 84°

Reaction and isotopes characteristics

Detector	Reaction	Radionuclide	Half-life	Threshold energy (MeV)	γ-ray (keV)
Al	$(n, \alpha)$	<sup>24</sup> Na	15 h	6	1368.6
Al	(n, spall)	<sup>22</sup> Na	2.6 yr	30	1274.5
Ni	(n, p)	<sup>58</sup> Co	70.78 d	1	810.75
Co	(n, 2n)	<sup>58</sup> Co	70.78 d	11	810.75
Co	(n, 3n)	<sup>57</sup> Co	270 d	20	122.07
Co	(n, p)	<sup>59</sup> Fe	45.1 d	4	1099.22
Bi	(n, 3n)	<sup>207</sup> Bi	31.55 yr	14.42	1063.6
Bi	(n, 4n)	<sup>206</sup> Bi	6.24 d	22.55	803
Bi	(n, 5n)	<sup>205</sup> Bi	15.31 d	29.62	703.3
Bi	(n, 6n)	<sup>204</sup> Bi	11.3 h	38.13	984
Bi	(n, 7n)	<sup>203</sup> Bi	11.8 h	45.37	820.2
Bi	(n, 8n)	<sup>202</sup> Bi	1.8 h	54.24	960.7
Bi	(n, 9n)	<sup>201</sup> Bi	1.85 h	61.69	629.1
Bi	(n, 10n)	<sup>200</sup> Bi	36 min	70.89	1026.5

X. Ledoux, ICTP-School, Trieste, 19-30 Oct 2015 N. Pauwels et al. / Nucl. Instr. and Meth. in Phys. Res. B 160 (2000) 315–327

# Neutron spectra produced by bombarding thick targets: $(100 \text{ MeV/u}) \text{ D} + {}^{9}\text{Be}$ , ${}^{238}\text{U}$ and $(95 \text{ MeV/u}) {}^{36}\text{Ar} + {}^{12}\text{C}$



## OUTLINE

- 1. Definition
- 2. Application

### 3. Facilities

- 4. Cross-section measurements
  - 1. Activation technique
  - 2. n,xn gamma
  - 3. Direct measurement of secondary neutrons
- 5. Double differential measurements
- 6. Particular cases

## **Facilities**

### Neutron production

- $\circ$  Nuclear reaction  $\rightarrow$  accelerator
- $\circ$  Photofission  $\rightarrow$  e<sup>-</sup> accelerator + converter
- $\circ$  Nuclear fission  $\rightarrow$  Reactor

#### Parameters

- Energy range 5-50 MeV for (n,xn) reactions
- Energy distribution mono-kinetic or white spectrum
- $\circ$  Fluence
- Time structure pulsed beam for tof measurement

### Types of facilities

- $\circ$  Open field
- $\circ~$  Collimated beam

## Reactor

- Neutron fission
- High flux
- No time spectrum
- Energy limited to 10 MeV
- Energy spectrum (fast neutrons for n,xn)







## Photo-production of neutrons with bremsstrahlung

- Electron beam
- Photon production by Bremsstrahlung
- Neutron production by  $(\gamma,xn)$  or  $(\gamma,f)$  reaction



- Continuous neutron energy spectrum
- "Low cost" accelerator
- High power Accelerator



nELBE yield:  $3*10^{11}$  n/s with 30 MeV 15  $\mu$ A (Target:Pb, liquid) 200 kHz GELINA yield:  $3*10^{13}$  n/s with100 MeV 96  $\mu$ A (Target: U(Hg cooled)) 800 Hz

## Intermediate energy 20-200MeV

#### Quasi-mono-energetic spectrum:

- Proton beam on thin <sup>7</sup>Li converter
- $\circ$  <sup>7</sup>Li(p,n)<sup>7</sup>Be reaction Q= -1,64 MeV → at 0° En ≈ Ep 2 MeV
- o Forward peak
- o Limitations :
  - Spectrum not purely mono-energetic -> pulsed beam
  - Low melting point of Lithium (limited intensity) -> liquid target
  - Target highly activated (7Be)

#### Continuous spectrum:

- Proton or deuteron beam on thick converter Be or C
- Continuous spectrum up to beam energy
- o Flux increasing with energy
  - The beam stops in the converter
  - Large power deposition  $\rightarrow$  cooling is challenging

#### Several facilities proposes both types of spectra



### Some Quasi-Monoenergetic facilities





### Mono-kinetic neutron sources

Nuclear reactions (The big four)

- p + <sup>7</sup>Li → n + <sup>7</sup>Be p + <sup>3</sup>H → n + <sup>3</sup>He d + <sup>2</sup>H → n + <sup>3</sup>He d + <sup>3</sup>H → n + <sup>4</sup>He
- Proton and deuteron beams with E< 4 MeV
- Purely mono-energetic neutrons for E<7 MeV and 14<E<17 MeV





### Spallation reaction

#### Proton beam with energy > 800MeV

Very intense neutrons source Proton accelerator 1 GeV x 1 mA = 1 MW  $\Rightarrow$  10<sup>17</sup> n/s

- Neutron production up to the proton energy
- Use of moderator to increase neutrons flux at low energy thermal or cold
- N-tof, WNR, SNS, ESS, JPARC





1000

## Type of beam

### Open field

- $\circ$  Neutron emission in  $4\pi$
- $\circ$  Background in the experimental area
- $\circ \mbox{The sample can be placed very close to the source}$

 $\circ$  VdG, PTB

### Collimated beam

- $\circ~$  The detectors can be placed close to the sample
- o Low flux because of the distance source-sample
- $\circ$  WNR, N-TOF, NFS

### □ "Conical" beam

- $_{\odot}$  Kinematic effect
- $\circ$  Neutrons are emitted in a cone in the forward direction
- LICORNE, FRANZ







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## Activation technique

#### Activation and off-line gamma ray spectrometric technique



$$n + {}^{A}_{Z}X \rightarrow {}^{A-x+1}_{Z}X + x n$$



#### Advantages:

- $\circ$ Target do not need to be isotopic if  $E_n < B_n$  (A+1)
- $\circ$  No need of pulsed beam

#### Drawbacks :

- $\circ$  Incident energy is not measured  $\rightarrow$  need mono-energetic beam
- o One measurement for each energy
- One target for each energy

### Cross-section of the <sup>241</sup>Am(n,2n) reaction



X. Ledoux, ICTP-School, Trieste, 19-30 Oct 2015

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## **Prompt** γ-ray spectroscopy

Detection of the y-rays stemming from the decay of excited states of nucleus created by the (n,xn) reaction.

Measure on line of prompt gamma spectroscopy of the  $\frac{A-x+1}{Z}X$ 



### Inelastic scattering and (n,xn) experimental studies

4 HPGe Planar (110°,150°) Actinides samples ΔEn = 10 keV @ En = 1 MeV



GRAPHEME @ FP16/30 m

Pulsed white neutron beam 10 meV - 20 MeV Multi-users facility 10 m to 400 m

<sup>12</sup>C, <sup>23</sup>Na, <sup>24</sup>Mg, <sup>28</sup>Si, <sup>52</sup>Cr, <sup>56</sup>Fe, <sup>58</sup>Ni, <sup>76</sup>Ge, <sup>nat</sup>Zr, <sup>nat,182,183,184,186</sup>W, <sup>206,207,208</sup>Pb, <sup>209</sup>Bi, <sup>232</sup>Th, <sup>235,238</sup>U

### Neutron Time of flight facility **GELINA@IRMM(Geel)**

<sup>52</sup>Cr : L.C. Mihailescu *et al.* **NPA786(2007)1** <sup>209</sup>Bi : L.C. Mihailescu *et al.* **NPA799(2008)1** <sup>208</sup>Pb : L.C. Mihailescu *et al.* **NPA811(2008)1** <sup>23</sup>Na : C. Rouki *et al.* **NIMA672(2012)82** <sup>235</sup>U : M. Kerveno *et al.* **PRC87(2013)024609**   $0\nu2\beta$  : A. Negret *et al.* **PRC88(2013)027601** <sup>28</sup>Si : A. Negret *et al.* **PRC88(2013)034604** <sup>76</sup>Ge : C.Rouki *et al.* **PRC88(2013)054613** <sup>56</sup>Fe : A. Negret *et al.* **PRC90(2014)034602** <sup>24</sup>Mg : A.Olacel *et al.* **PRC90(2014)034603** <sup>232</sup>Th : M.Kerveno *et al.* **EPJA(2014) accepted** <sup>7Li, <sup>12</sup>C, <sup>58</sup>Ni, <sup>nat,184,186</sup>W,<sup>206,207</sup>Pb, <sup>232</sup>Th, <sup>238</sup>U: conf.</sup>



### Measurement of ${}^{235}U(n,n'\gamma)$ and ${}^{235}U(n,2n\gamma)$ cross-section



X. Ledoux, ICTP-School, Trieste, 19-30 Oct 2015

## Measurement of $\sigma(n,xn\gamma)$



## From $(n, 2n\gamma)$ to (n, 2n) cross-section

Measurements :



#### Theoretical model :



## (n,xnγ) measurement at nELBE



## nELBE – double ToF detector setup



sample: <sup>nat</sup>**Fe** (99.8%) → 91.754% <sup>56</sup>Fe mass: 19.82 g → 18.15 g <sup>56</sup>Fe

R. Beyer, PhD Thesis

## <sup>56</sup>Fe(n,xn $\gamma$ ) measurement at nELBE



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## Direct measurement of secondary neutrons

### Measure the x neutrons emitted in the reaction

### Advantages :

- Direct measurement
- o Applicable to all nuclei

#### Drawbacks:

- Need a mono-isotopic target
- Neutron detector of high efficiency

#### Detector type :

- o Neutron balls
- Neutron spectrometer

## The neutron balls

- Measurement of neutron multiplicity event by event
- Composed of a tank filled by liquid scintillator
- Phototubes detected the prompt and delayed signal





### CARMEN

Measurement of neutron Multiplicity distribution  $4\pi$  , high efficiency High sensitivity to background

Application: n,xn cross-section measurements Nubar measurements Hot nuclei studies



## The neutron balls : working process



## Efficiency



### n,xn cross section measurement

Measurement of Mn(x) :number of reactions with neutron multiplicity x



#### Efficiency : Cf source

simulations to take into account the energy and angular distributions

J. Fréhaut, Nuclear Instruments and Methods 135 (1976) 511-518

V. J. Ashby et al., Physical Review 111, num 2 (1958) p 616

## Some $\sigma(n,2n)$ and $\sigma(n,3n)$ measured with neutron balls

Fréhaut et al.



#### L. R. Veeser et al., PRC 16, num 5 (1977) p 1792





## Neutron spectrometer

#### Elastic and inelastic cross-section measurements



## Spectrometer of CEA/Bruyères-le-Châtel



## Sample and shadow bars



Energy measurement:

Flight path : d 
$$\Rightarrow v = \frac{d}{tof_n} \Rightarrow \beta = \frac{v}{c} \Rightarrow \gamma = \frac{1}{\sqrt{1 - \beta^2}} \Rightarrow E = (\gamma - 1)mc^2$$

Start and stop signals : detector + accelerator or active target

Energy resolution:

$$\frac{\Delta E}{E} = \gamma(\gamma + 1)\sqrt{\left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta d}{d}\right)^2}$$

Δt time resolution detector + accelerator ≈1ns

 $\Delta L$  flight path uncertainty





## **TOF** technique



## <sup>190</sup>Os(n,n')<sup>190</sup>Os



## <sup>238</sup>U(n,n')<sup>238</sup>U



#### Neutron source

 $\circ$ I= 3 to 5  $\mu$ A

oTritrium target

Flux : Φ=3.10<sup>8</sup> n.sr<sup>-1</sup>.s<sup>-1</sup>

Sample

 $\circ$  m=30 g

○ A=190

 $\circ$  Distance source sample d = 7 cm

 $d\sigma/d\Omega = 10 \text{ mb.sr}^{-1}$ 

#### Detector:

- $\circ$  Surface detector =  $\pi * r^2 = 122cm^2$
- $\circ$  Intrinsic efficiency = 10%
- $_{\odot}$  Distance from sample L=800 cm

Number of neutrons detected by second :

$$N_{\rm det} = \frac{\Phi}{d^2} \frac{mN_{avo}}{A} \frac{d\sigma}{d\Omega} \frac{S}{L^2} \varepsilon$$

 $N_{det} \approx 1 \text{ counts/s}$ 

## **Multicells detectors**



X. Ledoux, ICTP-School, Trieste, 19-30 Oct 2015

Nuclear Instruments and Methods in Physics Research A 523 (2004) 102-115

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### Measurement of double differential emission cross-section for incident neutrons of 14,1 MeV



The neutron spectrum is the sum of all the channels where at least one neutron is emitted

### Pb(n,2n)

Nuclear Data for Science and Technology (1988 MITO), 229-232, Copyright © 1988 JAERI.



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## <sup>239</sup>Pu(n,2n) reaction cross-section measurement





 $T_{1/2}$ = 24110 y

T<sub>1/2</sub>= 87,7 y

α emitter 5456 keV 5499 keV

□ Activation technique

Measurement of direct neutrons

 $\Box$  n,xn $\gamma$  measurement

## <sup>239</sup>Pu(n,2n)<sup>238</sup>Pu cross-section by Activation technique

<sup>239</sup>Pu(n,2n) cross-section measurement near E<sub>n</sub>=14MeV, R. W. Lougheed, Radiochim. Acta 90, 833-843 (2002)

#### □Irradiation :

- Neutrons produced by d+T reaction
- o Flux : 5,5.10<sup>11</sup> n.cm<sup>-2</sup>.s<sup>-1</sup>
- $_{\odot}$  Integrated fluence : 8,98.10^{17} n in  $4\pi$

#### Sample :

- o130µg.cm<sup>-2</sup>, Φ=3 mm →10µg
- $\circ$  <sup>238</sup>Pu/<sup>239</sup>Pu ≈ 6.10<sup>-10</sup> before irradiation
- Distance source sample 4mm

#### Monitoring : Au(n,2n) cross-section





Before irradiation: A( $^{238}$ Pu)  $\approx 0,004$  Bq Integrated fluence = 8,98.10<sup>17</sup> n.cm<sup>-2</sup> After irradiation of m= 10µg  $\sigma$ =0,3 barn Nat  $^{238}$ Pu = 10<sup>9</sup> A( $^{238}$ Pu)  $\approx 2$  Bq

## $^{239}$ Pu(n,2n) $^{238}$ Pu cross-section from partial $\gamma$ -ray cross-sections (1)

#### LANSCE (Los Alamos Neutron Science Center)

- $_{\odot}$  Spallation source
- $_{\odot}$  White neutron beam
- $\circ$  Pulsed beam

#### GEANIE

- $_{\odot}$  26 high-resolution Ge detectors
- $_{\odot}$  BGO escape-suppression shields.

#### **GErmanium Array for Neutron Induced Excitations**





### <sup>239</sup>Pu(n,2n)<sup>238</sup>Pu cross-section from partial $\gamma$ -ray cross-sections (2)



L. A. Bernstein at al., PHYSICAL REVIEW C 65 021601(R)

## Direct measurement of secondary neutrons

Difficulty : distinguishing fission neutrons from (n,2n)

 $\Box$  Total neutrons multiplicity measurement (n,fis) + (n,2n) + (n,n')

- Neutron ball
- $\circ$  Thick target
- o Trigger on prompt peak
- □ For Mn≥ 4 only fission contribution
  □ Mn=2

$$N_F = \frac{\sum_{i \ge 4} N(i)}{\sum_{\nu \ge 4} P(\nu)} .$$

P(i) fission multiplicity probability



## Results on the <sup>239</sup>Pu(n,2n)<sup>238</sup>Pu cross-section measurements



## Cross-section measurement of the D(n,2n) reaction (1)

#### $n + D \rightarrow p + n + n$ Eth=3.34 MeV

- Three bodies system in the exit channel
- The emission of the 2 neutrons cannot be treated like evaporated neutrons
- Theoretical model based on the resolution of the Faddeev equations (J. Carbonell et B. Morillon)
  - Measurement of the 2 neutrons emitted -> Neutron ball
  - $\circ$  Active target made of C<sub>6</sub>D<sub>6</sub> :
    - $\circ$  reaction tagged by the recoil of the proton
    - o incident neutron energy measured by time of flight
  - $\circ$  Measurement with C<sub>6</sub>H<sub>6</sub> target to subtract the carbon contribution





## Cross-section measurement of the D(n,2n) reaction (2)



# (n,3n) cross section measurement by two techniques



The (n,xny) cross-section measurements are used to extract (n,xn) cross section-> need of theoretical model At NFS the <sup>90</sup>Zr(n,3n) cross-section can be measured by TALYS 1.4 calculation 1000 prompt y spectroscopy and by activation technique at the same time Cross section (mb) 92Zr(n,3n) 90Zr(n,3n)  $\rightarrow$ validation of the theoretical models 800 94Zr(n,3n)

600

400

200

0





Quasi-mono-energetic neutrons from 26 to 32 MeV

M. Kerveno et al., Letter of Intents for NFS facility

## Study of pre-equilibrium process in (n,xn) reaction

Measurement of (n,xn) double differential cross section in coincidence with neutron multiplicity.



#### Method :

- measurement of energy and angle of one neutron
- count of the (x-1) neutrons emitted simultaneously.

#### Experimental set-up :

- NE213 detectors
- CARMEN detector



#### Beam request:

- Quasi-monkinetic beam
- Pulsed
- Well collimated

X. Ledoux et al., Letter of Intents for NFS facility

## Summary

### □ n,xn reactions play an important role in the 1 MeV – 50 MeV range

- Reactor
- Waste production in high neutron flux
- Accurate library of nuclear data
- Several techniques exist
  - Activation
  - n,xnγ reactions
  - Direct outgoing neutron measurement
- □ All the techniques cannot be used in all cases
  - Specific detection set-up
  - Specific neutron facilities

## Yttrium

Thus the (n, xn) reaction of 89Y is important, when the superconductor is used in the neutron field of higher energy such as in the fast reactor and accelerated driven sub-critical system (ADSs).



Measurement of cross-sections for 89Y(n,xn) reaction at average neutron energies of 15–36 MeV

X. Ledoux, ICTP-School, Trieste, 19-30 Oct 2015

## (n,2n) and (n,3n) measurements



D(d,n)3He produces purely mono-energetic neutrons up to  $\overline{7}$  MeV But The low energy component is below the (n,2n) threshold D(d,n)3He can be used up 10 MeV

### Measurement on Carbon sample

Flux : Φ (n.cm<sup>-2</sup>.s<sup>-1</sup>)

Differential cross section :  $d\sigma/d\Omega$  (barn.sr<sup>-1</sup>)

Intrinsic efficiency : 20%

Sample : A=12,  $\Phi$ =3cm, h=3cm, m=15g

$$N_{\rm det}(s^{-1}) = \Phi\left(\frac{m}{A}N_{avo}\right)\varepsilon\frac{d\sigma}{d\Omega}d\Omega$$

 $\Phi = 10^{6} \text{ n.cm}^{-2}.\text{s}^{-1}$ do/d $\Omega = 0,001 \text{ barn.sr}^{-1}$ Ndet = 0,02 count.s<sup>-1</sup>

