



## TRAINING COURSE ON NEUTRON DOSIMETRY, RADIOBIOLOGY AND INSTRUMENTATION: **Passive detectors:** activation detectors and superheated emulsions

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#### **ACTIVATION DETECTORS: introduction**

The method consists in measuring the induced activity of a target exposed to a neutron field and relating it to the neutron fluence rate.





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#### **ACTIVATION DETECTORS:** basic principles

The reaction rate (s<sup>-1</sup>) is related to the neutron fluence rate by:

$$R = N_{TOT} \int_{0}^{\infty} \sigma_{E} \Phi_{E} dE$$

where:

$$N_{TOT} = \frac{N_{AV}}{AW}W$$

is the total number of nuclei inside the target,  $N_{AV}$  is the Avogadro number, AW the atomic weight and W the target weight.



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#### **ACTIVATION DETECTORS: thermal and epithermal neutrons**

The following simplified method assumes that,

for thermal neutrons

$$\sigma_E = \frac{\sigma_0 V_o}{V}$$

- where v is the neutron velocity,  $v_0$  the neutron velocity @ 0.025 eV (2200 m s<sup>-1</sup>),  $\sigma_0$  is the neutron cross section @ 0.025 eV;
- and, for epithermal neutrons:

$$\Phi_E = \frac{\Phi_{epi}}{E}$$

(1/E slowing-down behaviour)

- where  $\Phi_{epi}$  is the epithermal fluence rate per unit In(E)







#### **ACTIVATION DETECTORS: thermal and epithermal neutrons**

the reaction rate R can be written as:

$$R = N_{TOT} \int_{0}^{E_{max}} \sigma_{E} dE = N_{TOT} \left( \int_{0}^{0.5eV} \frac{\sigma_{0}V_{0}}{v} n_{th,E} v dE + \int_{0.5eV}^{E_{max}} \sigma_{E} \frac{\Phi_{epi}}{E} dE \right) =$$
$$= N_{TOT} \left( \sigma_{0}V_{0} \int_{0}^{0.5eV} n_{th,E} dE + \Phi_{epi} \int_{0.5eV}^{E_{max}} \frac{\sigma_{E}}{E} dE \right) =$$
$$= N_{TOT} \sigma_{0}n_{tot}V_{0} + N_{TOT}RI\Phi_{epi} = N_{TOT} \sigma_{0}\Phi_{0} + N_{TOT}RI\Phi_{epi}$$

• where:

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$$RI = \int_{0.5eV}^{E_{max}} \frac{\sigma_E}{E} dE \qquad \text{is the resonance integral (barn}$$

 $\Phi_0$  is the neutron fluence rate defined as the thermal neutron density times the 2200 m s<sup>-1</sup> neutron velocity.







#### **ACTIVATION DETECTORS:** thermal and epithermal neutrons

- The expression in the previous slide is valid for an infinitely thin target:
- but an activation target shows a given thickness and the reaction rate expression must be corrected for the fluence rate depression factors,  $G_{th}$  and  $G_{epi}$

$$R = G_{th} N_{TOT} \sigma_0 \Phi_0 + G_{epi} N_{TOT} R I \Phi_{epi}$$

 G<sub>th</sub> and G<sub>epi</sub> depend on the target material and thickness

Target thickness (mg cm <sup>-2</sup> )	Gold		Indium				
	G <sub>th</sub>	G <sub>epi</sub>	G <sub>th</sub>	G <sub>epi</sub>	Φ		Φ
5	0.995	0.763	0.987	0.649		\ /	· · · ·
7.5	0.994	0.698	0.981	0.573			
10	0.992	0.645	0.976	0.519			
20	0.985	0.521	0.956	0.400			
40	0.969	0.410	0.924	0.294			







#### **ACTIVATION DETECTORS: thermal and epithermal neutrons**

- The thermal neutron component can be discriminated by the epithermal one with a cadmium cover;
- cadmium cut-off @ 0.5 eV





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### **ACTIVATION DETECTORS: target reaction rate and activity**

By neglecting:

- neutron capture on already activated nuclei;
- the target burn-up;
- The number of activated nuclei during irradiation is:

$$\frac{dN^*}{dt} = R - \lambda N^*$$

At the end of irradiation (@ time  $t_{\mbox{\scriptsize irr}}$ ):

$$N^{\star}(t_{irr}) = \frac{R}{\lambda} \left( 1 - e^{-\lambda t_{irr}} \right)$$

The induced activity at the end of irradiation is:

$$A(t_{irr}) = R(1 - e^{-\lambda t_{irr}})$$





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#### **ACTIVATION DETECTORS: target reaction rate and activity**

After a waiting time t<sub>w</sub> (the time from the end of irradiation up to the beginning of counting):

$$A(t_w) = A(t_{irr}) e^{-\lambda t_w}$$

The total counts acquired from  $t_w$  up to  $t_{meas}$  (i.e. counting time  $t_{meas}$ ) are:

$$C_{tot}(t_{meas}) = R\varepsilon b (1 - e^{-\lambda t_{irr}}) e^{-\lambda t_w} \int_{t_w}^{t_w + t_{meas}} e^{-\lambda t} dt =$$
$$= R\varepsilon b (1 - e^{-\lambda t_{irr}}) e^{-\lambda t_w} \frac{1 - e^{-\lambda t_{meas}}}{\lambda}$$

If  $\lambda t_{meas} << 1$ :

$$\frac{1 - \mathbf{e}^{-\lambda t_{meas}}}{\lambda} = \frac{1}{\lambda} \left( 1 - 1 + \lambda t_{meas} \right) = t_{meas}$$









#### **ACTIVATION DETECTORS: target reaction rate and activity**

The reaction rate is assessed by measuring the saturation activity of the activated material (gamma rays with a NaI(TI) or a Ge detector,  $\beta^{-}$  particles with a GM detector):



- Where b is the branching ratio and  $\epsilon$  is the detector (peak) efficiency.
- If a bare and a cadmium covered target are used to separate the thermal and the epithermal components, the cadmium correction factor  $F_{Cd}$  should be used;
  - since cadmium is not completely transparent to epithermal neutrons.

$$C_{epi} = F_{Cd}C_{Cd}$$

- where  $C_{epi}$  are the counts due to epithermal neutrons to be subtracted from the counts from the bare target and  $C_{Cd}$  are the counts from the Cd-covered target.
- $F_{Cd}$  depends on the thickness of the target material and of the Cd cover.



Fig. 12.2.2. The Cadmium correction factor for Gold foils in an isotropic neutron field as a function of Gold foil thickness. Cd filter thickness 1 mm



an isotropic neutron field as a function of Indium foll thickness. Cd filter thickness 0.5 and 1 mm







### **ACTIVATION DETECTORS: target reaction rate and activity**

The specific saturation activities should be subtracted for obtaining that due to thermal neutrons only:

$$R_{th} = \left(\frac{R_{bare}}{W_{bare}} - \frac{R_{epi}}{W_{Cd}}\right) W_{bare}$$

- where  $W_{\text{bare}}$  and  $W_{\text{Cd}}$  are the weights of the bare and Cd-covered target, respectively.
- Finally, for estimating  $\Phi_0$  and  $\Phi_{epi}$ :

$$\Phi_{0} = \frac{R_{th}}{G_{th}N_{tot}\sigma_{0}}$$
$$\Phi_{epi} = \frac{R_{epi}}{G_{epi}N_{tot}RI}$$

• It should be remembered that in the epithermal region:

$$\Phi = \int_{0.5eV}^{E_{\text{max}}} \frac{\Phi_{epi}}{E} dE = \Phi_{epi} \ln \left( \frac{E_{\text{max}}}{0.5eV} \right)$$







# **ACTIVATION DETECTORS:** target materials for thermal and epithermal neutron detection

Main activation reactions for thermal neutron detection:

- ✓ <sup>197</sup>Au(n, $\gamma$ )<sup>198</sup>Au: T<sub>1/2</sub>=2.69 d,  $\sigma$ (0.025 eV)=98.5 b;
- ✓  $^{115}$ In(n, $\gamma$ )<sup>116m</sup>In: T<sub>1/2</sub>=54.15 min,  $\sigma$ (0.025 eV)=157 b;
- ✓ Other materials: Dy, Co, Cu, Ag.

	Gold	Indium
Half-life	2.695 d ( <sup>198</sup> Au)	54.15 min ( <sup>116m</sup> In)
σ <sub>0</sub> (0.025 eV)	98.8 b	157 b
RI	1560 b	2600 b









#### **ACTIVATION DETECTORS: gold and indium foils**





Au-198 and In-116m decay schemes with branching ratios (in brackets)







#### **ACTIVATION DETECTORS:** fast neutrons

Several threshold reactions can be exploited, e.g.:

- ✓  ${}^{58}Ni(n,p){}^{58}Co$   $E_{th} = 1.9 \text{ MeV}$
- ✓  ${}^{59}Co(n,\alpha){}^{56}Mn$   $E_{th} = 5.2 \text{ MeV}$
- ✓  ${}^{54}$ Fe(n,p) ${}^{54}$ Mn  $E_{th} = 2.2 \text{ MeV}$
- ✓  ${}^{58}$ Ni(n,2n) ${}^{57}$ Ni  $E_{th} = 13.0 \text{ MeV}$
- ✓  $^{115}$ In(n,n')<sup>115m</sup>In  $E_{th} = 0.339$  MeV
- ✓  ${}^{32}S(n,p){}^{32}P$   $E_{th} = 2.0 \text{ MeV}$
- ✓  ${}^{12}C(n,2n){}^{11}C$   $E_{th} = 20 \text{ MeV}$
- ✓  ${}^{27}$ Al(n, α) ${}^{24}$ Na  $E_{th} = 4.9 \text{ MeV}$
- ✓  ${}^{27}$ Al(n, p) ${}^{27}$ Mg E<sub>th</sub> = 3.8 MeV
- The neutron spectrum can be reconstructed from the saturation activities assessed with a set of activation foils;
  - → The reaction cross section against energy (the "detector response") must be known for this purpose.







#### **ACTIVATION DETECTORS:** high-energy hadrons

The hadron fluence above about 20 MeV can be assessed through the activation of <sup>11</sup>C (x-sec $\cong$ 20 mb, slightly dependent on hadron energy), i.e. for neutrons through the reaction:

- ✓  $^{12}C(n,2n)^{11}C$   $E_{th} = 20 \text{ MeV}$   $T_{1/2}=20.5 \text{ min}$
- A plastic scintillator is exposed to the hadron field and
- → the <sup>11</sup>C activity is measured by coupling the scintillator to a PM and by counting the positrons emitted by <sup>11</sup>C decay.









#### SUPERHEATED EMULSIONS

- "Superheated emulsion" is the name adopted by ISO and ICRU for detectors based on a superheated liquid suspended in a gel, also known as bubble detectors or superheated drop detectors.
  - The suspended droplets consist of an overexpanded halocarbon and/or hydrocarbon which vaporizes upon exposure to the high-LET recoils from neutron interactions.
  - The superheated emulsion is contained in a vial and acts as a continuously sensitive, miniature bubble chamber.
  - The total number of bubbles evolved from the radiation-induced nucleation of drops gives an integrated measure of the total neutron exposure.







Courtesy of F. d'Errico, Yale Univ. and DMNP Pisa Univ.







#### SUPERHEATED EMULSIONS

 Bubbles can be counted either optically (by eye) of through an acoustic transducer transforming the micro-explosion following bubble formation into an electronic signal.



P.K. Mondal et al. Nucl.Instrum.Meth. A729 (2013) 182-187





Am-Be

789<sub>100</sub>

Neutron Energy (MeV)

**∃**\*(Э)Ф



### SUPERHEATED EMULSIONS

- Superheated emulsions are currently used either as personal and environmental dosemeters or as neutron spectrometers.
  - Neutron spectrometry is performed by exploiting the different response to neutron energy against temperature or pressure of the superheated liquid.
  - Dosemeters: one of their advantages is the possibility of determining an average ambient dose equivalent rate in a pulsed neutron field.
  - They are completely insensitive to low-LET radiation, X and γ rays as well as muons, which is a clear advantage when measuring the neutron component in mixed fields.



4 5 6 7 89<sub>10<sup>0</sup></sub>

2 3

6789<sub>10</sub>1

All figures in this slide: Courtesy of F. d'Errico, Yale Univ. and DMNP Pisa Univ. 0.

4 5 6 7 89 10<sup>1</sup>

2 3

Neutron Energy (MeV)







#### SUPERHEATED EMULSIONS

The  $H^*(10)$  response is underestimated for epithermal neutrons (up to about 100 keV) and is fairly accurate in the neutron energy interval from 100 keV up to about 10 MeV.



All figures in this slide: Courtesy of F. d'Errico, Yale Univ. and DMNP Pisa Univ.



- Low energy response: MCNP calculations
- Fast neutron response: PTB calibrations







#### SUPERHEATED EMULSIONS

- The response to higher energies was measured by irradiating bubble detectors with quasimonoenergetic neutrons in the energy interval 46-133 MeV. The results showed a significant underestimate of the *H*\*(10) (d'Errico et al. RPD 100 \_ (2002) 529-532).
- Measurements were also performed in the mixed field of high-energy radiation available at CERF. An underestimation of about 40% with respect to the reference ambient dose equivalent was observed in that experiment (Mitaroff et al., RPD 102 (2002) 7-22).
- Measurements in high-energy neutron fields generated by various types of hadron beams performed at CERN showed that bubble detectors underestimate the *H*\*(10) by a factor 0.4-0.7 depending on the neutron spectrum (Agosteo et al. Health Phys. 75 (1998) 619-629).

Detector	Ambient dose equivalent rate (µSv h-1)						
	front NA44	side NA44	dump NA45	NA45			
LINUS <sub>sph,UMi</sub>	21.2±0.2	22±1	227±22	108±10			
bubble detectors	19±4	13±1.5	210±44	78±6			







#### SUPERHEATED EMULSIONS

- The possibility of extending the response of bubble detectors to HE neutrons was investigated by exposing the dosemeters inside lead converters of varying thickness at the CERF facility.
  - MC simulations showed that, as the thickness of the lead converter increases, a growing number of evaporation neutrons are generated by the high-energy component of the neutron field, thus enhancing the detector sensitivity.
  - This behaviour was confirmed experimentally. The comparison with the reference H\*(10) indicates that the required thickness of the lead converter is in the interval 1-1.5 cm.









#### SUPERHEATED EMULSIONS: OPTICAL BUBBLE COUNTING

The application of large volume detector chambers for the three-dimensional dosimetry of brachyterapy implants lead to study novel position-sensitive systems for assessing the bubble spatial distribution.

- Optical tomography was proposed by d'Errico et al, 2008 for this purpose.
- The satisfactory results obtained with this technique lead to apply scattered light for bubble counting of superheated emulsions for individual dosimetry (d'Errico et al, 2008).
- The dosemeter is placed in a light-shielded enclosure and illuminated from the bottom by LEDs (light-emitting diodes). The light scattered by the bubbles is detected by photodiodes positioned along the detector wall.
- A very good linearity of the response (photodiode voltage against number of bubbles) of this system was observed.
- The uniformity in size of the drops suspended in the gel was found to be of primary importance for a smooth behaviour of the system. This feature is guaranteed by the manufacturing technique for the superheated emulsions which is capable of providing drops with size in the range 50-150  $\mu$ m with a dispersion lower than 10%.



Adapted from: d'Errico, F., Di Fulvio, A., Mariañski, M. , Selici, S., Torriginai, M., Radiation Measurements 43 (2008) 432-436. 22