

Neutron spectrometry with Bonner spheres

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Before 1960 a number of neutron spectrometry techniques were in use: nuclear emulsions, proportional counters, cloud chambers, organic scintillators and recoil telescopes, ^3He proportional counters, $^6\text{LiI}(\text{Eu})$ scintillators, TOF methods and activation foils techniques.

A crucial development occurred in 1960 with the introduction of the multi-sphere spectrometer (also called Bonner Sphere Spectrometer, BSS), based on a $^6\text{LiI}(\text{Eu})$ scintillator placed at the centre of polyethylene spheres of different diameters (2" up to 18").

Advantages of measuring fast neutrons with moderator based techniques:

- Availability of high sensitivity thermal neutron detectors with good gamma rays rejection;
- Availability of hydrogen based materials with "high" moderation capability and "low" neutron capture cross section;
- "Direct" detection techniques have considerably lower sensitivity.



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The BSS has been widely applied for almost 50 years:

- Isotropy of the response;
- Large variety of active and passive central detectors with different thermal neutron sensitivity and photon rejection capability, to be selected according to the workplace of interest;
- Wide energy range:
 - Thermal up to 20-30 MeV for pure polyethylene;*
 - Up to 10^2 - 10^3 MeV when using heavy metal inserts;*
- Easy operation (but complex analysis!)

Disadvantages

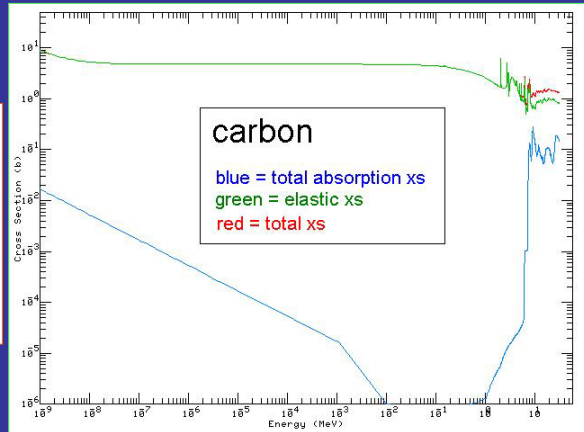
- Weight (especially big or lead loaded spheres in "hard" workplaces);
- Need to sequentially irradiate the spheres (long irradiation sessions);
- Need to uniformly irradiate the spheres (response function definition)
- Poor energy resolution (fine structures ignored);
- Complex unfolding procedures, expert user and pre-information needed;
- Complex analysis to get uncertainties.



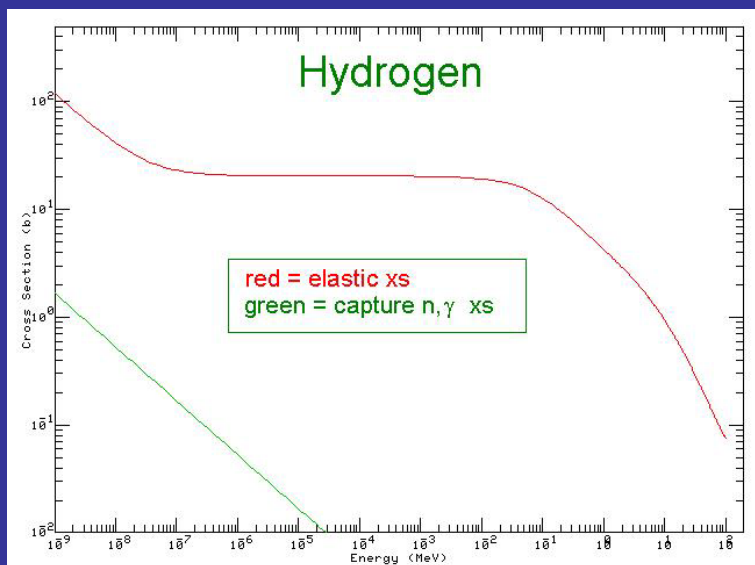
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Energy range	Reaction	Typical xs (barn)
	H(n,n)H	1
	C(n,n)C	1
> 5 MeV	C(n,n' γ)	0.1
> 7 MeV	C(n, α)	0.01 – 0.1
> 8 MeV	C(n,n' 3α)	0.5
> 15 MeV	C(n,p)	0.01
> 15.5 MeV	C(n,d)	0.01 – 0.1
> 18 MeV	C(n,n'p)	0.01



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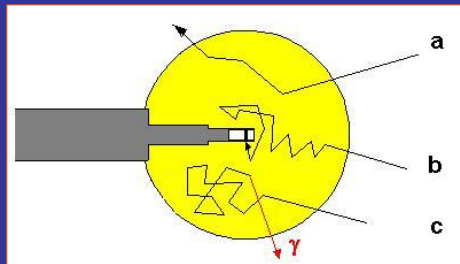


The probability for a neutron to produce a “count” when a given sphere is used, is only function of its energy, E.

For each sphere there is an energy value, E_{max} , at which the probability reaches a maximum. This corresponds to the “best” thermalization condition.

Lower energy neutrons tend to be captured by the polyethylene (giving γ of 2.2 MeV), whilst higher energy neutrons tend to escape from the assembly.

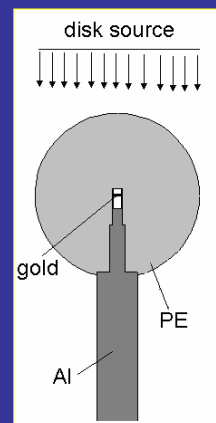
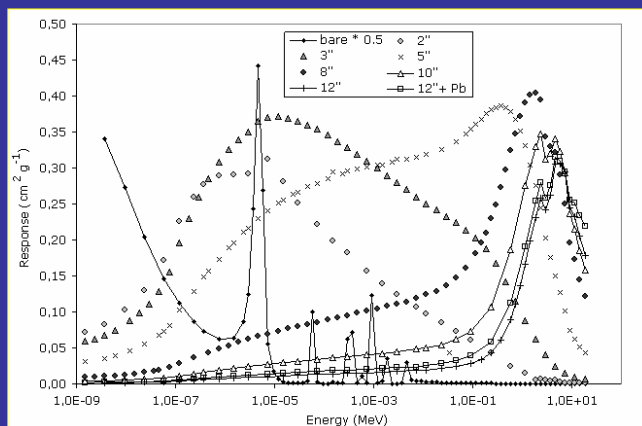
For a fixed polyethylene density, E_{max} only depends on the sphere diameter (better, on the sphere mass).



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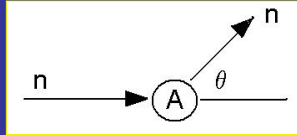
The “response function” of the thermal detector in a given sphere is defined as the detector count per unit fluence, when the sphere is uniformly irradiated, and is only function of the energy.



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Slowing down in infinite, homogeneous, non multiplying media



The scattering is isotropic in the center of mass, but not in the lab. The average cosine of the scattering angle and the logarithmic average energy loss per collision, ξ , only depend on A :

$$\mu = \langle \cos \vartheta \rangle = \frac{2}{3} A$$

$$\xi = 1 + \frac{\alpha}{1 - \alpha} \ln \alpha$$

$$\alpha = \left(\frac{A-1}{A+1} \right)^2$$

The transport mean free path is given by:

$$\lambda_{tr} = \frac{1}{\Sigma_s (1 - \mu)}$$

Note: for thermal scattering (\sim isotropic, $\mu=0$) the mean free path (diffusion theory) is simply $1/\Sigma_s$



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For mixtures:

$$\mu = \frac{2}{3} \cdot \frac{\sum N_i \sigma_s^i A_i}{\sum N_i \sigma_s^i} \quad \xi = \frac{\sum N_i \sigma_s^i \xi_i}{\sum N_i \sigma_s^i}$$

Where N_i is number of atoms per unit volume and σ_s^i the epithermal scattering cross section, for the i -th element. In the case of **polyethylene**

$$\begin{aligned} \mu &= 0.625 & \xi &= 0.95 \\ \Sigma_s &= 3.29 \text{ cm}^{-1} & \Sigma_a &= 0.027 \text{ cm}^{-1} & \lambda_{tr} &= 0.809 \text{ cm} \end{aligned}$$

If, for each sphere, we consider only the neutrons with energy E_{\max} and we calculate the average segmented length to reach the thermal region, L , we realize that there is a roughly constant ratio between L and the radius of the sphere, R ($L/R = 1.5$ with 0.3 st. dev. for a typical commercial system)

Escaping and absorption processes limit the response at higher or lower energies.



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The choice of the thermal neutron detector (or central detector) depends heavily on the following elements which characterize the workplace neutron field :

- Intensity of the field
- Time structure of the field
- Presence of electromagnetic noise
- Photon component

detector	geometry	$R(\text{cm}^2)_{1 \text{ MeV}; 8''}$
4x4 $^6\text{LiI}(\text{Eu})$	cylindrical	0.2
^3He 05NH1	cylindrical	0.4
SP9	spherical	2.7

Active detectors

Cylindrical 4 mm x 4 mm $^6\text{LiI}(\text{Eu})$ scintillator: almost point-like, Q value $^6\text{Li}(n,\alpha)\text{T} = 4.78 \text{ MeV}$. The high Z of iodine implies in principle a not negligible photon sensitivity

SLAC, INFN, Georgia Tech, CIEMAT, UPM Madrid.

^3He filled proportional counter: 8 kPa, cylindrical 10 mm x 9 mm, (typical 05NH1 from Eurisys). Reduced gamma sensitivity. UAB Barcelona, IRSN (France).

Spherical SP9 counter by Centronic Ltd., UK. Very high sensitivity due to the large diameter (3.2 cm). PTB, IPNE, JRC Geel, ITN et al.

12.7 x 12.7 cm² organic scintillator $^{12}\text{C}(n,2n)^{11}\text{C}$. (>18 MeV). High energy fields. SLAC.



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Passive detectors

Active detectors can fail in scenarios characterized by **high dose rate, pulsed fields, high electromagnetic noise and high photon background** (in the vicinity of a reactor core, PET cyclotrons, target of particle accelerators, treatment room of medical LINACs).

TLD pairs. TLD600/TLD700 pairs have been frequently employed. Good n/γ discrimination capability, but limited sensitivity (long irradiations).

Recently, a **INFN / TLD Poland** collaboration allowed obtaining special $^6\text{LiF}(\text{Mg,Cu,P})$ based detectors with 80 mg.cm⁻² sensitive layer:

TLD type	S_{th} / S_{γ} (pGy.cm ⁻²)	$S_{th} / S_{th \text{ MCP-6}}$
TLD-600	220±20	0.15±0.01
MCP-6 (230 mg.cm-2)	192±15	1.00±0.02
MCP-6s (80 mg.cm-2)	337±17	1.07±0.06
TLD-700	0.48 ± 0.03	(1.1±0.1)·10 ⁻⁴

Sensitivity to MCP-6s ~ 10% of $^6\text{LiI}(\text{Eu})$

Gold foils. Photon-insensitive. Mainly used in medical LINACs. Low neutron sensitivity (0.4 Bq_{sat} g⁻¹ cm² s @ 1 MeV in the 8'').

Drawbacks: ^{196}Au (333, 356, 426 keV) when $E_{\gamma} > 8.07 \text{ MeV}$.

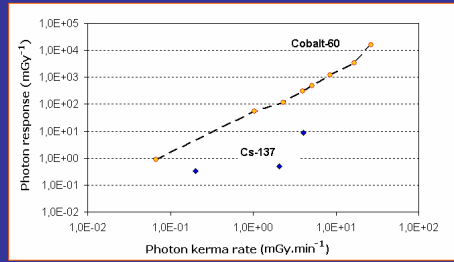
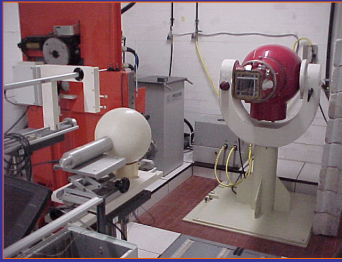
Extra-neutrons from the (γ,n) reactions when $E_{\gamma} > 18 \text{ MeV}$.



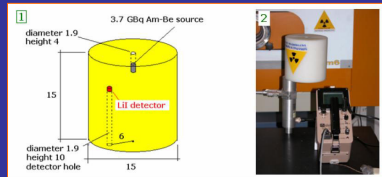
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The sensitivity of BSSs to photons (from the external field or the 2.2 MeV γ from the capture in polyethylene) should be evaluated.



Periodical repeatability tests, usually with a small ^{241}Am -Be source in fixed geometry including a moderator.



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The simulated response matrix needs to be validated in well characterized neutron fields, typically ISO 8529-1 (quasi mono-energetic fields allows checking punctual energies, radionuclide sources cover 2-3 decades).

The comparison between the **measured** fluence (supposing a known spectrum and a given response matrix) and the **delivered** fluence (metrological data) allows estimating the overall accuracy of the matrix (typ. $\pm 3\%$).



Sphere	Thermal	$^{252}\text{Cf}(\text{D}_2\text{O})$	^{252}Cf	$^{241}\text{Am-Be}$
Bare	0.99			
2"		1.03	0.98	1.03
3"		0.99	0.94	1.02
5"		0.97	1.01	0.98
8"		0.96	0.97	0.98
10"		1.01	1.00	1.01
12"		1.03	0.98	1.02
12"+Pb		1.09	1.02	0.97



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The response functions are calculated under “plane parallel” beam uniformly irradiating the spheres.

The validation experiments should be performed under these conditions

- At sufficient distance from a source
- With beam scanning techniques in collimated beam accelerators

The validation experiments require irradiation to a known spectrum:

Techniques to subtract the in-scattered contribution (ISO 8529-2).

Small spheres require small shadow-cones.

Measurements in variable output facilities (accelerators, reactors) require the use of normalization instruments

One instrument if only the rate can vary.

Two instruments with different energy dependence are needed to check the constancy of the energy composition of the beam.



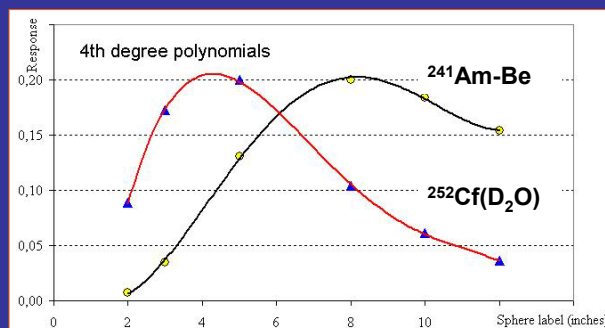
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The neutron spectrometry with Bonner spheres is a under-determined problem (infinite solution are mathematically acceptable)

However, few well chosen spheres (typically 4-5) usually provide nearly the totality of the obtainable information.

This is due to the large overlapping between the response functions and their similar shape, especially for the > 6 ".



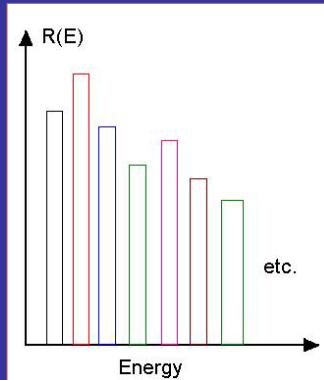
Rank of the
structure matrix
 $TR \cdot R$



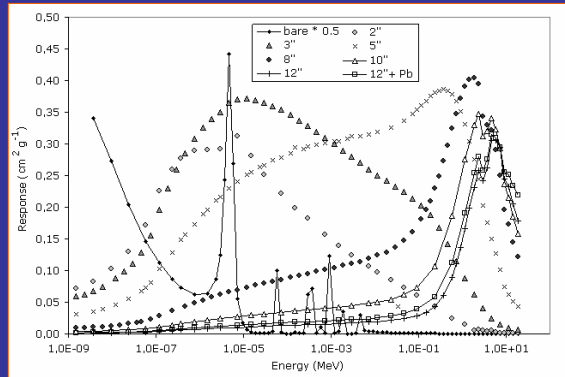
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Why the response matrix of the BSS is redundant?



Ideal response matrix



Realistic response matrix: overlapping and similarity between functions



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The neutron spectrometry with BSS has many sources of uncertainty:

- Statistical counting uncertainty
- Normalization instrument/s uncertainty;
- Response matrix uncertainty;
- Polyethylene density;
- Simulated and realistic irradiation geometry;
- Isotropy of the central counter;
- Stability of the counter and rate dependence;
- Photon contribution;

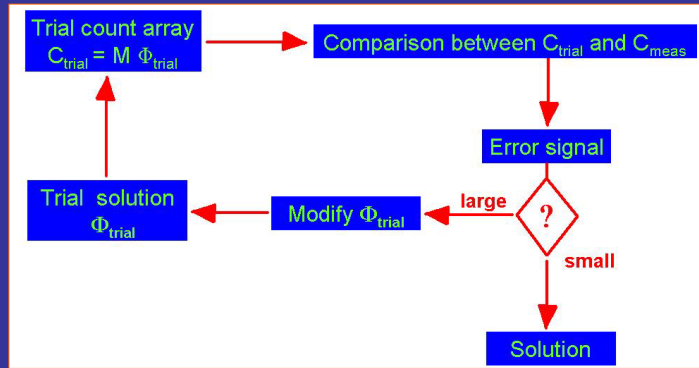
The determination of the uncertainty affecting the results (shape of the spectrum, total fluence, dose equivalent, average conversion coefficient, average energy...) is not a direct propagation problem.

In principle we should study the "probability distribution" of the results when the input quantities are changed according to a given input distribution.



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The **unfolding procedure** is the most complex task in BSS spectrometry

Computerized unfolding procedures:

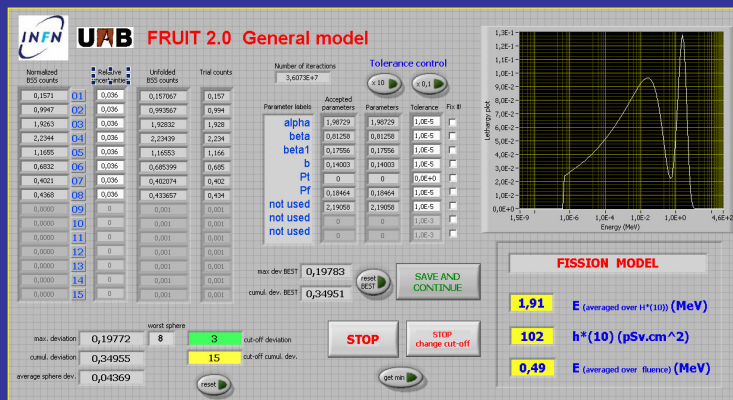
- usually very complex;
- need an expert user;
- need additional suggestions to derive a physically acceptable solution.



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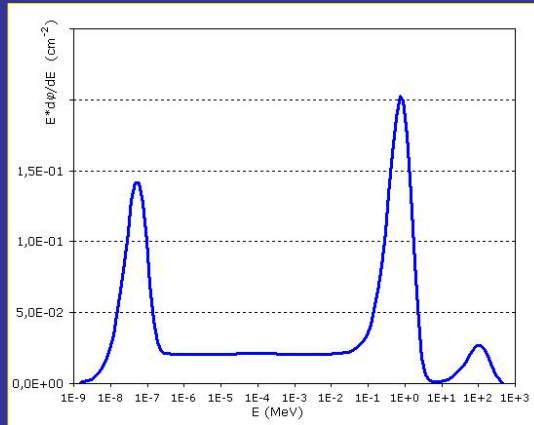
- Parameterization of the spectrum with physical models;
- Limited amount of a priori information. No “default spectrum”;
- The user easily controls the convergence procedure;
- User-friendliness and visual operation. Changes in dosimetric quantities and graphical spectra are continuously displayed. Uncertainties treatment.



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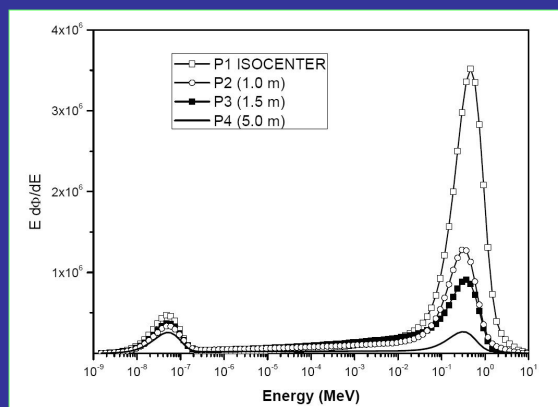
Workplace: DAΦNE e⁺/e⁻ collider; 510 MeV/beam
 Type of detector: 4x4 ⁶Lil(Eu) active scintillator
 Spheres: 2", 3", 5", 8", 10", 12", 12"+ 1 cm Pb
 Irradiation mode: 20 min per sphere + rem counter for normalization



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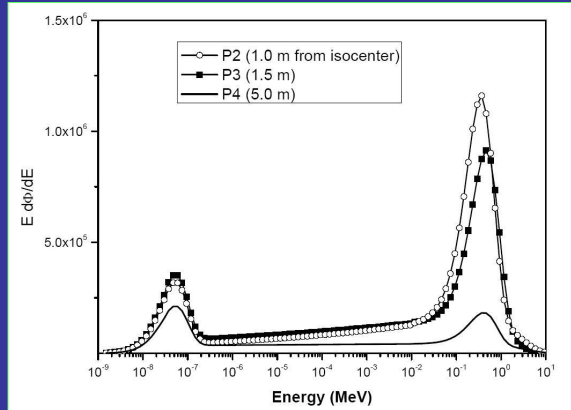
Workplace: 18 MV medical LINAC
 Type of detector: gold foils (10 mm diameter x 0.1 mm)
 Spheres: 2", 3", 5", 8", 10", 12"
 Irradiation mode: 10 Gy at the isocentre per sphere



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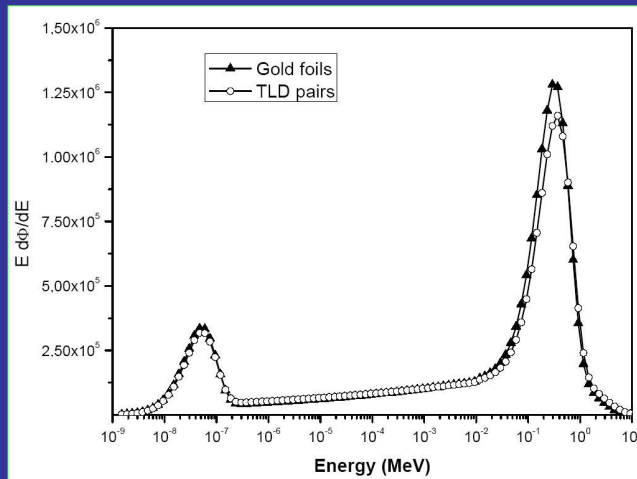
Workplace: 18 MV medical LINAC
 Type of detector: TLD pairs
 Spheres: 2", 3", 5", 8", 10", 12"
 Irradiation mode: 10 Gy at the isocentre per sphere



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Comparison gold foils / TLDs in the point at 1 m from the isocentre



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CONCLUSIONS

- BSSs have been used in a large variety of neutron fields (nuclear plants, medical and research accelerators, radionuclide based facilities, calibration fields, cosmic rays) covering an energy range from thermal up to hundreds MeV.
- Modern unfolding codes, coupled with a minimal knowledge of the radiation environment, allow:
 - limiting the impact of the under-determined nature of the problem;
 - treating the uncertainties;
- **Typical achievable accuracy: <5% for fluence or <10% for dose equivalent.**
- The BSS technique is still regarded as the most adequate for radiation protection applications, where the knowledge of the neutron spectrum is essential to determine the operational quantities for external exposure.



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