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Workplace testing of the new single sphere neutron spectrometer based on Dysprosium activation foils (Dy-SSS)

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ABSTRACT

A photon insensitive passive neutron spectrometer consisting of a single moderating polyethylene sphere with Dysprosium activation foils arranged along three perpendicular axes was designed by CIEMAT and INFN. The device is called Dy-SSS (Dy foil-based Single Sphere Spectrometer). It shows nearly isotropic response in terms of neutron fluence up to 20 MeV. The first prototype, previously calibrated with 14 MeV neutrons, has been recently tested in workplaces having different energy and directional distributions. These are a 2.5 MeV nearly mono-chromatic and mono-directional beam available at the ENEA Frascati Neutron Generator (FNG) and the photo-neutron field produced in a 15 MV Varian CLINAC DHX medical accelerator, located in the Ospedale S. Chiara (Pisa). Both neutron spectra are known through measurements with a Bonner Sphere Spectrometer.

In both cases the experimental response of the Dy-SSS agrees with the reference data. Moreover, it is demonstrated that the spectrometric capability of the new device are independent from the directional distribution of the neutron field. This opens the way to a new generation of moderation-based neutron instruments, presenting all advantages of the Bonner sphere spectrometer without the disadvantage of the repeated exposures. This concept is being developed within the NESCOFI@BTF project of INFN (Commissione Scientifica Nazionale 5).

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1. Introduction

The idea of a multi-detector single-sphere neutron spectrometer with nearly isotropic fluence response was developed within the framework of the INFN-CIEMAT collaboration. This included a moderating polyethylene sphere with thermal neutron detectors arranged along three perpendicular axes. The average of the signals from the detectors located at the same radius proved to be nearly independent from the direction distribution of the incident neutron field up to 20 MeV. On this basis, a prototypal device was produced. The sphere diameter is 30 cm and the measurement positions are placed at fixed radial distances (0, 3, 6, 9, 10.5, 12, 14 cm) along the three axes *X*, *Y* and *Z*. The total number of measurement positions is 37 (12 positions/axis \times 3 axes+one position at the sphere centre). Details on the geometry are given in Ref. [1].

Different types of passive thermal neutron detectors have been considered, namely TLD pairs [2,3] and Dysprosium (Dy) activation foils [1]. Advantages of Dy-foils are: (1) the complete photon

insensitivity, (2) the high activation cross-section of ¹⁶⁴Dy (2700 barn at thermal energy) and (3) the reasonably short half-life of the activation product ¹⁶⁵Dy (2.334 h). The combination of (2) and (3) allows obtaining large values of specific activity in operation conditions [4]. Due to its high energy (end-point 1.287 MeV) and high yield (83%), the beta emission of ¹⁶⁵Dy is suited for in-situ measurements with a portable beta-counter. The prototypal spectrometer equipped with Dy-activation foils is called Dy-SSS (Dy foil-based Single Sphere Spectrometer). The Dy activation foils used in this device have diameter 1.2 cm, thickness 0.01 cm and purity higher than 99.9%. Density of the foils is 8.55 g cm⁻³.

The first calibration factor of the Dy-SSS was derived with a nearly mono-directional 14 MeV neutron beam [1]. This work shows further tests aimed at verifying the spectrometer response at different energy and direction distributions. Two neutron fields were chosen, namely a 2.5 MeV beam available at the ENEA Frascati Neutron Generator (FNG) [5] and the photo-neutron field produced in a 15 MV Varian CLINAC DHX medical accelerator, located in the Ospedale S. Chiara (Pisa). Whilst the 2.5 MeV beam is nearly mono-directional, the angle distribution of the LINAC field is not known a priori. In this second case, the isotropy of the device response is experimentally tested for the first time.

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The energy distribution of the neutron fluence is known in both cases through measurements with a Bonner Sphere Spectrometer equipped with Dy activation foils [4,6,7], also called Dy-BSS.

2. Response of the Dy-SSS

The response matrix of the Dy-SSS is presented in Ref. [1]. This was derived from 1.5 meV to 146 MeV, on the basis of a 59 groups equi-logarithmic energy structure, using the Monte Carlo code MCNPX 2.6 [8] and cross section data from the ENDF/B-VII nuclear library [9]. The scattering in polyethylene was modelled using the S(α , β) treatment. The number of (n, γ) capture events in ¹⁶⁴Dy foils per unit incident neutron fluence was scored as a function of the measurement point, of the neutron energy and of the irradiation geometry.

This is called response matrix,

 $M_i^{geom}(E)$ (units cm²),

where: "*i*", with i=1,...,37, denotes the measurement position within the sphere; "*geom*" denotes the irradiation geometry under which the response matrix was derived. Three irradiation geometries were simulated: uni-directional along one axis, namely (1 0 0), uni-directional along (1 1 1) direction and isotropic (ISO).

Ref. [2] shows that the quantity obtained by averaging the response of the six detectors located at the same radius within the sphere (called "six-points-average") is basically geometry-independent, thus demonstrating that the device has nearly isotropic response. Small deviations from isotropy are observed below 1 keV.

The integral $\int dE \times M_i^{geom}(E) \times \dot{\Phi}(E)$, where $\dot{\Phi}(E)$ (units cm⁻² s⁻¹ MeV⁻¹) is the energy-dependent neutron fluence, is expected to be proportional to the specific activity measured in the foils under saturation irradiation condition (called C_i , where "i" denotes the position). The procedure needed to obtain C_i from the result of the beta counting [4] needs corrections for (1) the decay between the end of the irradiation and the beginning of the measurement, (2) the decay during the measurement and (3) the fraction of the saturation activity reached during the irradiation.

The spectrometer calibration factor, F, is here intended as the proportionality factor between the simulated response of *i*-th measurement position and the corresponding experimental value, C_i . The value of F should not depend from the measurement position, the energy or direction distribution of the field. As described in Ref. [1], F was derived in a nearly mono-energetic (14 MeV) and mono-directional field. Eq. (1) was applied

$$F_{i} = \frac{C_{i}}{\int dEM_{i}^{(1\ 0\ 0)}(E)\dot{\Phi}(E)} = \frac{C_{i}}{\dot{\phi}\int dEM_{i}^{(1\ 0\ 0)}(E)\varphi(E)} = \frac{C_{i}}{\dot{\phi}f_{i}}$$
(1)

where $\varphi(E)$ is called unit spectrum (units MeV⁻¹) and represents the neutron spectrum normalised to the unit fluence; $\varphi(E) = \dot{\Phi}(E)/\dot{\phi}$, where $\dot{\phi}$ is the total fluence rate (units cm⁻² s⁻¹) $f_i = \int dEM_i^{(1 \ 0 \ 0)}(E)\varphi(E)$ (units cm²).

A set of eleven F_i values was derived for different measurement positions and its average is regarded as the best estimation of the spectrometer calibration factor. This value is $F=0.141 \pm 0.003$.

3. Irradiation test at 2.5 MeV neutrons

The objective of this test was to confirm that the device calibration does not depend on the energy distribution of the neutron field. Thus, a nearly mono-energetic 2.5 MeV field was used to verify the Dy-SSS calibration factor. Because the present irradiation took place in the low-scattering room of the ENEA Frascati Neutron Generator, using a target-to-detector distance large enough to neglect the field divergence, the direction distribution of the neutron field can be assumed to be monodirectional. The irradiation scenario is sketched in Fig. 1. Seven measurements positions aligned with the radiation field have been equipped with Dy activation foils. Their *x* coordinates, with respect to the sphere centre, were: -10.5, -9.0, -6.0, -3.0, 0.0(centre), 3.0 and 6.0.

Because the neutron fluence varied with time, a NE213 scintillation counter was used for normalisation purposes and the step-by-step correction described in Ref. [6] was adopted to determine the fraction of saturation activity reached in the Dy foils during the irradiation.

The neutron spectrum and the reference fluence (fluence normalised to NE213 reading) for the 2.5 MeV beam were previously determined with a Bonner Sphere Spectrometer equipped with Dy-foils [4,6,7], called Dy-BSS. The average fluence rate at the position of the sphere centre was approx. 1.6×10^3 cm⁻² s⁻¹. The Dy-SSS was exposed for approximately 1 h and the Dy-foils were immediately counted. Count rates spanning from 2 to 16 s⁻¹ were registered. The corresponding saturation count rates, C_i , were derived as described in Section 2. For this irradiation test the irradiation geometry is uni-directional along one axis, namely (1 0 0). Therefore the response matrix M_i^{100} was used for calculations. Referring to Eq. (1), the best estimation of the calibration factor derived at 2.5 MeV, $F_{2.5 \text{ MeV}}$, is given by

$$F_{2.5 \text{ MeV}} = \frac{1}{\dot{\phi}} \left\langle \frac{C_i}{f_i} \right\rangle_{i = 1, \dots, 7}$$
(2)

where the ratio C_i/f_i is averaged over the seven measurement positions used in this test. The result is $F_{2.5 \text{ MeV}} = 0.138 \pm 0.004$, which agrees and confirms the calibration factor derived at 14 MeV ($F = 0.141 \pm 0.003$).

The values of C_i/f_i obtained for every measurement positions are given in Table 1.



Fig. 1. Sketch of the irradiation test in 2.5 MeV nearly mono-chromatic and monodirectional neutrons. A cut view of the sphere is shown. The measurement positions are the black points.

Table 1

Calibration table of the Dy-SSS at 2.5 MeV. The X-axis corresponds to the beam direction. The points with minus sign are located along the radius opposing the beam. Uncertainties are the quadratic combination of the counting uncertainties with the overall uncertainty of the Dy-SSS response matrix (estimated as \pm 3%, see Ref. [1]).

$\begin{array}{cccc} -10.5 & 222 \pm 10 \\ -9.0 & 219 \pm 10 \\ -6.0 & 213 \pm 9 \\ -3.0 & 207 \pm 9 \\ 0.0 \ (centre) & 213 \pm 10 \\ 3.0 & 213 \pm 10 \\ 6.0 & 230 + 12 \end{array}$	X coordinate (cm)	$C_i/f_i (cm^{-2} s^{-1})$
	- 10.5 - 9.0 - 6.0 - 3.0 0.0 (centre) 3.0 6.0	$\begin{array}{c} 222\pm 10\\ 219\pm 10\\ 213\pm 9\\ 207\pm 9\\ 213\pm 10\\ 213\pm 10\\ 230\pm 12\\ \end{array}$

4. Irradiation test in the photo-neutron field produced in a medical LINAC

This section describes a further test, where not only the neutron spectrum but also the irradiation geometry changed with respect to the first calibration conditions. The treatment room of the 15 MV Varian CLINAC DHX medical accelerator and the location of the point of test is shown in Fig. 2. The spectrometer was positioned 150 cm laterally with respect to the isocenter, at the height of the isocenter. This test is especially challenging for the Dy-SSS because

- (1) the neutron spectrum covers all neutron energies from thermal up to several MeV;
- (2) the angular distribution of the neutron field is unknown. Whilst the fast neutron component comes mainly from the LINAC head, the floor, ceiling and the walls significantly contribute to the thermal and epithermal components.

The neutron spectrum in the point of test, called reference spectrum, was determined using the Bonner Sphere Spectrometer equipped with Dy-foils (Dy-BSS) and based on the following spheres: 2 in., 2.5 in., 3 in., 3.5 in., 4 in., 4.5 in., 5 in., 7 in., 8 in., 10 in. and 12 in. The FRUIT code [10] was used to unfold the experimental data. Details of these measurements are given in Ref. [7]. The reference spectrum, normalised to the unit absorbed dose to water at the isocenter and in equi-lethargy representation, is given in Fig. 3. The total neutron fluence rate associated to the reference spectrum is $(3.18 \pm 0.10) \times 10^5$ cm⁻² s⁻¹ for an accelerator output of 500 ± 10 MU min⁻¹.

The Dy-SSS was exposed for 25 min at the point of test. Dy activation foils were positioned in the following measurement positions within the sphere: r=0 (centre sphere), r=6 cm (six foils), r=9 cm (6 foils), r=10.5 cm (6 foils), and r=12.5 cm (6 foils). The total number of detectors used was 25. The LINAC conditions were the same as in the reference spectrum measurement: the accelerator output was 500 ± 10 MU min⁻¹ and the treatment field was 10 cm × 10 cm at the isocenter. After irradiation the foils were counted, each for 2 min., obtaining in all cases a counting uncertainty of $\pm 1\%$ (one s.d.) or better. These data were elaborated as indicated in Section 2 to obtain the count rate



Fig. 2. The 15 MV Varian CLINAC DHX medical accelerator of the Ospedale S. Chiara (Pisa), the treatment room and the location of the point of test. The spectrometer is located 150 cm laterally with respect to the isocenter. Dimensions of the room are approx. 8 m \times 6.7 m \times 3 m (height).Upper part: view from top; lower part: lateral view.



Fig. 3. Reference neutron spectrum (Dy-BSS) and Dy-SSS unfolded spectrum in the point of test at 150 cm from the isocenter. The spectra are normalised to the unit absorbed dose to water at the isocenter and are reported in equi-lethargy representation.



Fig. 4. Comparing the experimental C_r values with the "folded" C_{rf} values obtained by folding the spectrometer response matrix $(M^{ISO}(r, E))$ with the unfolded spectrum.

in saturation condition, C_i . The twenty-five C_i values were reduced to five C_r values (r=0, 6.0 cm, 9.0 cm, 10.5 cm, 12.0 cm) using the six-points-average procedure. The response matrix derived for isotropic irradiation and based on the six-points-average, $M^{ISO}(r, E)$ [1], was used for unfolding purposes.

The C_r values were unfolded using the FRUIT code in nonparametric mode (or FRUIT/SGM) [11]. As a suitable guess spectrum, the reference spectrum derived with the Dy-BSS was used. As far as the input uncertainties (needed to determine the uncertainties on the unfolded data) are concerned, the following values were considered: $\pm 3\%$ for the C_r values (quadratic combination of the counting uncertainty and that associated to the beta counter reproducibility) and $\pm 3\%$ as overall uncertainty of the response matrix. The unfolded spectrum is reported in Fig. 3.

The coherence between the Dy-SSS unfolded spectrum and the experimental data (C_r values) may be checked by comparing the C_r values with the "folded" C_{rf} values, namely those obtained by folding the spectrometer response matrix ($M^{ISO}(r, E)$) with the unfolded spectrum. This is done in Fig. 4 and shows a good agreement for all radial positions. This figure also serves to show that the reading as a function of the position is a smooth curve, as

Table 2

Comparing the Dy-BSS (regarded as reference instrument) and the Dy-SSS in terms of spectrum-integrated quantities in the photo-neutron field of the 15 MV medical Linac. The fluence rate is referred to an accelerator output of 500 MU min⁻¹.

Quantity	Dy-BSS	Dy-SSS
$\dot{\phi}$ (cm ⁻² s ⁻¹) ϕ/D (cm ⁻² MU ⁻¹) $H^*(10)/D$ (mSv Gy ⁻¹) Fluence fractions	$\begin{array}{c} (3.18\pm0.10)\times10^5 \\ (3.82\pm0.14)\times10^4 \\ 0.383\pm0.018 \end{array}$	$\begin{array}{c} (2.9\pm0.2)\times10^5 \\ (3.5\pm0.3)\times10^4 \\ 0.36\pm0.04 \end{array}$
E < 0.4 eV 0.4 eV < E < 10 keV E > 10 keV	22.0% 21.8% 56.2%	21.3% 22.2% 56.5%

required [12] to moderation-based instruments providing spectrum-integrated readings, as the Bonner spheres as well as the Dy-SSS.

Relevant spectrum-integrated quantities such as the total fluence, the ambient dose equivalent $H^*(10)$ and the fractions of fluence under given energy intervals (E < 0.4 eV, corresponding to the thermal domain, 0.4 eV < E < 10 keV, corresponding to the intermediate region, and E > 10 keV, corresponding to the evaporation peak) are given in Table 2. From either Fig. 3 or Table 2 it can be noted that the Dy-SSS provides accurate results in terms of neutron spectrum or integral quantities.

Final uncertainties for the Dy-SSS are almost double than for the Dy-BSS. This is due to two main reasons

- (1) The shape of the response functions of the Dy-BSS and the Dy-SSS are similar. However, the Dy-BSS relies on eleven responses (equal to the number of spheres used) whilst the Dy-SSS only relies on five responses. Therefore, the first system has higher resolving power than the second;
- (2) The Dy-BSS has intrinsically isotropic response, whilst for the Dy-SSS the isotropic response is approximated using the sixpoints-average procedure. In Ref. [2] is shown that this approximation is affected by systematic errors, especially for the shallowest positions (r=12.0 cm in the case of this test) and the energy region below 1 keV.

Nevertheless, it should be kept in mind that the possibility of determining the neutron spectrum in a single exposure is a clear advantage of the Dy-SSS. Moreover, the achievable uncertainties for integral quantities (lower than 10%) and for the single bins of the spectrum (<30% in the thermal domain, 20%-25% in the intermediate region, 10%-20% in the evaporation peak) are fully acceptable for the purposes of neutron monitoring in workplaces.

5. Conclusions

A single moderating sphere neutron spectrometer called Dy-SSS (Dy foil-based Single Sphere Spectrometer), which conceptual design was previously established, was experimentally tested in irradiation scenarios different from the field used for the first calibration. The first calibration took place in a nearly monochromatic and mono-directional 14 MeV beam. Because this device was designed to determine the neutron spectrum independently on its energy or direction distribution, the following tests were performed:

1. Exposure in a nearly mono-chromatic and mono-directional 2.5 MeV beam. With respect to the calibration condition, the

energy changed but the direction distribution remained unchanged.The calibration factor obtained at 2.5 MeV agrees within 3% with that obtained in the first calibration at 14 MeV.

2. Exposure in the photo-neutron field produced in a 15 MV medical LINAC. The neutron spectrum spanned over a broad energy interval (thermal energies up to approx. 10 MeV) and the directional distribution of the field was unknown. The Dy-SSS experimental readings were combined using the six-points-average procedure and unfolded using the FRUIT code and the response matrix for isotropic irradiation, $M^{ISO}(r, E)$. The agreement between the Dy-SSS unfolded spectrum and the reference spectrum, obtained with the Dy-BSS, is satisfactory. Uncertainties on the spectrum and the integral quantities are higher for the Dy-SSS than for the Dy-BSS, because of the lower number of responses used and because of the limitations included in the use of an isotropic response matrix and of the six-points-average procedure.

Both tests gave satisfactory results. A remarkable result is that the Dy-SSS is able to determine the neutron spectrum over a broad energy interval in a single exposure, independently on the direction distribution of the field. This opens the way to a new generation of moderation-based neutron instruments that will present the advantages of the Bonner sphere spectrometer without the disadvantage of the repeated exposures. This study is being continued in the NESCOFI@BTF project (INFN-CSN 5) with the adaptation of the Dy-SSS structure to measure high-energy neutrons (E > 20 MeV) [13] and the replacement of Dy foils with direct reading detectors. This will result in a real-time, singleexposure, broad-energy range neutron spectrometer well suited for the spectrometric monitoring of a large variety of neutron producing facilities.

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