# REVISION OF THE <sup>252</sup>Cf AND D<sub>2</sub>O-MODERATED <sup>252</sup>Cf REFERENCE NEUTRON FIELDS FOR USE IN RADIATION PROTECTION DOSIMETRY

R. Méndez, J.M. Gómez-Ros, D. J. Thomas, A. K. Thompson, R. Bedogni

# ABSTRACT

ISO Standard 8529-1:2001 on Reference neutron radiations is under revision. The spectral data for the D<sub>2</sub>O-moderated <sup>252</sup>Cf field in the 2001 version of the standard refer to calculations performed in the early 80s. There are changes in the revised standard, and these arise from updated calculations, described in this paper, based on a new <sup>252</sup>Cf spectrum (from ENDF/B-VIII.0) and the MCNP6.2 transport code with ENDF/B-VIII.0 cross sections. In addition, the impact of different possible construction details of the D<sub>2</sub>O-moderated <sup>252</sup>Cf assembly is provided. Lastly, in view of the need to perform irradiations of dosemeters on-phantom, the variation of spectrum and fluence across a 30 cm × 30 cm surface placed 100 cm from the source has been evaluated.

# INTRODUCTION

The first part of ISO International Standard 8529 "Reference neutron radiations - Characteristics and methods of production" is being revised by the working group ISO/TC85 (Nuclear Energy) /SC2 (Radiological Protection)/WG2 (Reference Radiation Fields)/SG3(Neutrons). The historical recommended calibration fields of <sup>241</sup>Am-Be, <sup>241</sup>Am-B, <sup>252</sup>Cf and D<sub>2</sub>O moderated <sup>252</sup>Cf have therefore been reassessed in light of new cross sections, new transport codes, and experimental evidence [1]. Whilst the discussion about <sup>241</sup>Am-Be and <sup>241</sup>Am-B is reported in Refs. [1, 2], this work describes proposed changes for the <sup>252</sup>Cf and D<sub>2</sub>O-moderated <sup>252</sup>Cf fields.

According to ISO 8529-1:2001 [3], the <sup>252</sup>Cf spectrum tabulated and plotted there was obtained by integrating a Maxwellian distribution with nuclear temperature 1.42 MeV. This was discussed in recent papers [4, 5], where it was suggested that better data are available from ENDF/B data files, the most recent edition being ENDF/B-VIII.0 [6]. This suggestion was adopted in the revised standard. As far as the D<sub>2</sub>O-moderated <sup>252</sup>Cf field is concerned, 8529-1:2001 data refer to calculations performed in the early 80s [7] using the "05R" Monte Carlo code [8] with ENDF/B-V cross sections [9]. Here a Maxwellian distribution with nuclear temperature 1.42 MeV was used for the <sup>252</sup>Cf spectrum. In this work the simplified geometry from Ing and Cross [7] was kept but MCNP6.2 [10] with ENDF/B-VIII.0 <sup>252</sup>Cf spectrum was chosen as the source term. Calculations were also made to account for different construction details of the moderating assembly such as the sphere diameter, D<sub>2</sub>O purity, and source positioning system. Lastly, to provide information relevant to the irradiation of personal dosemeters on-phantom, the variation of spectrum and fluence across a 30 cm × 30 cm surface placed 100 cm from the source was evaluated.

# MATERIALS AND METHODS

# <sup>252</sup>Cf field

As mentioned in the Introduction, recent work [5, 4] underlined some issues with the <sup>252</sup>Cf spectrum in ISO 8529-1:2001. Although the text there says it was derived from a Maxwellian function with nuclear temperature of 1.42 MeV, this procedure does not actually reproduce the tabulated data. The present authors believe that ENDF/B point data, originated by Mannhart's evaluation of experimental measurements [11], and presented as both group and point values, would constitute a more robust basis for the new recommendations. The newly proposed spectrum coincides with the ENDF/B-VIII.0 group data above 35 keV (69 groups). Below this energy the two groups of the ENDF/B-VIII.0 group

data, which are very broad when plotted on a logarithmic scale, were replaced by 20 groups assembled by re-binning the point data with 3 bins per decade, down to 10 meV. The total number of energy groups is 89. The complete spectrum is shown in Fig. 1, which compares the proposed new spectrum from Table A of the Annex with the ISO 8529-1 (2001) spectrum. The spectrum-averaged fluence-to-dose equivalent conversion coefficients for this new <sup>252</sup>Cf spectrum, based on ICRP 74 [12], coincide with those reported in ISO 8529-1:2001.



Figure 1. Comparison of proposed and ISO-8529-1:2001 spectra for  ${}^{252}$ Cf.  $B_E$  is the spectral neutron emission rate from the source.

### D<sub>2</sub>O-moderated <sup>252</sup>Cf field

Over the years, calibration laboratories have adopted different construction solutions for the D<sub>2</sub>O-moderated  $^{252}$ Cf field, producing slightly different neutron spectra [13, 14, 15]. Differences are found in the guide used to locate the source in the sphere centre, the material used to contain the heavy water, and the structure used to suspend or hold the moderating sphere. In addition, every moderating assembly has a specific D<sub>2</sub>O purity and  $^{252}$ Cf source capsule. The revised Standard will again rely on an idealized model, very similar to the original one of Ing and Cross [7], but a recommendation will be given to derive the spectrum and the spectrum-averaged quantities by characterising each specific assembly as it is constructed. With the aim of supporting this work, this paper presents plots and Tables showing the effect of small variations in the D<sub>2</sub>O purity and construction details on the spectrum and integral quantities.



Figure 2. Schematic model for the D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron source.

The idealized model of the D<sub>2</sub>O-moderated <sup>252</sup>Cf source is shown schematically in Fig. 2. It comprises a spherical <sup>252</sup>Cf volume source (radius 6.4 mm) located in the centre of a D<sub>2</sub>O sphere with a diameter of 300 mm. The <sup>252</sup>Cf source has the energy distribution specified in Fig. 1 and Table A. The guide used to introduce the source to the centre of the moderating sphere was modelled as an iron tube of 6.4 mm internal radius with 0.8 mm thick walls. The wall of the D<sub>2</sub>O sphere is 0.8 mm of iron and is surrounded by a 1 mm thick shell of cadmium. The assembly is located in an idealized infinite-sized room in vacuum, meaning that no walls have been modelled. These construction parameters are hereafter called "proposed assembly". The spectrum calculated at 100 cm from the source centre, in the equatorial plane of the assembly, using a ring detector (F5 in MCNP) is called the "proposed spectrum". This was derived using MCNP6.2 [10] with ENDF/B-VIII.0 cross sections. Statistical uncertainties associated with Monte Carlo calculations are around 0.1% for every component of the fluence distribution, except in a few energy intervals with values around 1.5%, as a consequence of the number of simulated particles.

Material composition was obtained from Refs [16, 17] with densities for heavy water and air 1.105 g.cm<sup>-3</sup> and  $1.205 \cdot 10^{-3}$  g.cm<sup>-3</sup> (dry air at sea level). The thermal treatment S( $\alpha$ , $\beta$ ) has been considered for D and O in heavy water, as well as for <sup>56</sup>Fe metal.

Different sets of simulations were made by varying the parameters indicated below:

- distance from source centre: from 25 to 200 cm;
- moderating sphere submersed in vacuum (proposed assembly) or air;
- Cd shell thickness: 0 (absent) and 1 mm (proposed assembly);
- D<sub>2</sub>O purity: from 95 to 100 % (proposed assembly);
- D<sub>2</sub>O sphere diameter: from 29.7 cm to 30.3 cm;
- Internal diameter of the source positioning tube: from 12.8 mm (proposed assembly) to 25 mm.

The spectra are here reported using ISO 8529-1:2001 formalism, Fig. 1, 3 and 6. For the generic energy group between  $E_i$  and  $E_{i+1}$ , the group emission rate  $B_i$  is given as

$$B_i = \int_{E_i}^{E_{i+1}} B_E \, dE$$

where  $B_E = dB/dE$  is the spectral emission rate. Data are normalized to the unit total source emission rate,  $B = 1 \text{ s}^{-1}$ . Following the conventions of ISO 8529-1:2001, the graphical representations are equilethargic, i.e., plotted in such a way that equal areas under curves represent equal group emission rate proportions, being  $B_E E = dB/d(\ln E/E_0) = EdB/dE$ , defined as fluence per unit lethargy. The spectra are represented as histograms of  $B_i/\ln(E_{i+1}/E_i)$  (linear scale) versus the neutron energy,  $E_n$  (logarithmic scale).

Ambient and personal dose equivalents were calculated using the fluence-to-dose equivalent conversion coefficients,  $h^*_{\Phi}(E)$  and  $h_{p\Phi}(E, \alpha)$ , provided by ICRP 74 [12]. The spectrum averaged operational quantities are calculated as:

$$H = \int_0^\infty \Phi_E \cdot h(E) dE$$

where *H* is the ambient or personal dose equivalent depending on whether h(E) represents  $h_{\phi}^{*}(E)$  or  $h_{p\phi}(E, \alpha)$ .

Spectrum-averaged conversion coefficients,  $\bar{h}$  , have been calculated as

$$\bar{h} = \frac{H}{\Phi}$$

where  $\Phi$  is the total fluence.

The fluence-average energy,  $\overline{E}_{\Phi}$ , and ambient dose-equivalent average  $\overline{E}_{H^*}$  were determined as follows:

$$\bar{E}_{\Phi} = \frac{1}{\Phi} \int_{0}^{\infty} E \cdot \Phi_{E}(E) dE$$
$$\bar{E}_{H^{*}} = \frac{1}{H^{*}} \int_{0}^{\infty} E \cdot \Phi_{E}(E) \cdot h_{\Phi}^{*}(E) dE$$

where  $\Phi_E(E)$  is the spectral fluence at energy *E*.

In order to determine the fraction of neutrons that are absorbed in the moderating assembly, i.e., absorbed fraction,  $f_{abs}$ , neutron current over a spherical surface surrounding the D<sub>2</sub>O assembly was simulated, that corresponds to tally F1 in MCNP.



Figure 3. Comparison of D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron spectra with and without Cd (both in vacuum) and ISO 8529-1:2001.

#### RESULTS

#### Effects of distance, air submersion and cadmium shell

The proposed spectrum at 100 cm from the centre of the D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron source, Table B of the Annex, is compared with the ISO 8529-1:2001 data in Fig. 3. This distance explains the fact that the fluence is higher in air.

Integral quantities are compared in Table 1. The spectrum shape from 0.1 to 1 MeV results from the anisotropic scattering in deuterium. The small dips at 0.44 and 1.0 MeV come from scattering resonances of <sup>16</sup>O [18]. Compared to the current ISO spectrum, the proposed one is slightly harder, exhibiting more neutrons from 0.4 to 3 MeV. This causes the fluence-average energy to increase from 0.55 to 0.57 MeV. By contrast, due to the energy dependence of the ICRP 74 fluence-to-dose equivalent conversion coefficient, the dose equivalent-average energy decreases from 2.1 to 1.98 MeV.

Dosimetric Magnitudes	Ф	<b>f</b> abs	$\overline{E}_{\Phi}$	$\overline{E}_{\mathrm{H}^*}$	$\overline{h}^*(10)$	$\overline{h}_p(10,0)$
	[cm <sup>-2</sup> ]	[ %]	[MeV]	[MeV]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]
ISO 8529-1:2000	-	11.50	0.55	2.1	105	110
Proposed spectrum	7.00E-06	11.40	0.57	1.98	115	116
Proposed in air	7.11E-06	11.22	0.56	1.98	114	117
Proposed without Cd	7.91E-06	0.38	0.51	1.96	104	116

Table 1. Parameters of interest for current and proposed models of the D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron standard. Variation of the parameters for the proposed standard are shown when either including air or removing Cd.

The absorbed fraction obtained considering the whole space around the assembly is  $f_{abs,4\pi}$  = 11.4 % (cf. 11.5 % in 8529-1:2001). If only the directions on the equatorial plane are considered (90° to the vertical axis of the assembly),  $f_{abs,90^\circ}$ =12 %. From these values, anisotropy factor at 90° for D<sub>2</sub>O-moderated <sup>252</sup>Cf can be derived,  $F_{I}(90^\circ)$  = 0.993, and can be compared with experimental values for <sup>252</sup>Cf and Am-Be neutron sources [20]:

$$F_{l}(90^{\circ}) = \frac{1 - f_{abs,90^{\circ}}}{1 - f_{abs,4\pi}}$$

The spectrum-averaged fluence to ambient dose equivalent conversion coefficient is  $\overline{h^*}(10)$ = 115 pSv·cm<sup>2</sup>, 10 % larger than that in 8529-1:2001 (105 pSv·cm<sup>2</sup>). The spectrum-averaged fluence to personal dose equivalent at 0° conversion coefficient is  $\overline{h_p}(10,0^\circ)$  = 116 pSv·cm<sup>2</sup> (to be compared with 110 pSv·cm<sup>2</sup> from ISO 8529-3:1998 [19]). The calculation for other recommended angles has also been performed and results are shown in Table 2. An increase was found for all angles, ranging from +5 % to +9 %.

d	Ф	$\overline{E}_{\Phi}$	$\overline{E}_{\mathrm{H}^*}$	<i>h</i> <sup>*</sup> ( <b>10</b> )	π <sub>p</sub> (10,0°)	<del>П<sub>р</sub></del> (10,15°)	<del>П<sub>р</sub></del> (10,30°)	<del>П<sub>р</sub></del> (10,45°)	<u> </u>	<u>ћ<sub>р</sub></u> (10,75°)
[cm]	[cm <sup>-2</sup> ]	[MeV]	[MeV]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]
25	1.20E-04	0.55	1.95	111.98	112.79	114.87	115.66	107.56	92.05	58.37
50	2.83E-05	0.57	1.98	114.29	115.13	117.23	118.12	109.96	94.25	59.96
75	1.25E-05	0.57	1.98	114.69	115.54	117.64	118.55	110.37	94.63	60.22
100	7.00E-06	0.57	1.98	114.88	115.75	117.82	118.74	110.55	94.79	60.33
125	4.48E-06	0.57	1.98	114.96	115.81	117.92	118.83	110.64	94.86	60.33
150	3.11E-06	0.57	1.98	114.98	115.84	117.95	118.86	110.67	94.89	60.40
175	2.28E-06	0.57	1.98	115.04	115.89	118.01	118.92	110.72	94.94	60.43
200	1.75E-06	0.57	1.98	115.04	115.89	118.00	118.92	110.72	94.93	60.43
ISO 8529-1*		0.55	2.1	105	110	109	109	102	87.4	56.1

Table 2. Magnitudes for quantities of interest, including fluence-to-personal dose conversion coefficients as a function of angle, for current and proposed models of the D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron standard as a function of distance from the centre of the source to the point of calculation. \*ISO 8529-1 does not provide the magnitude of the total fluence.

The dependence of spectrum-averaged quantities on the distance is shown in Table 2. This study is needed because the  $D_2O$ -moderated <sup>252</sup>Cf assembly is a volume source. To study the changes in the neutron spectrum with the distance and the departure from the inverse-square law expected in the vicinity of the assembly, Fig. 4 shows the spectra variation for a few distances defined as the quotient:

$$q_r = \frac{\Phi_r(E) \cdot r^2}{\Phi_{ref}(E) \cdot r_{ref}^2}$$

where

*r* distance from the source centre for the generic spectrum

 $\Phi_r(E)$  the energy distribution of the neutron fluence in the equatorial plane at radial distance *r* reference distance = 100 cm  $\Phi_{ref}(E)$  reference spectrum



Figure 4. Spectra variation for D<sub>2</sub>O-moderated <sup>252</sup>Cf as a function of distance and relative to reference spectrum at 100 cm. The upper plot focuses on neutron energies above 1 MeV.

This figure indicates at what energies the spectrum changes with distance. Since the biggest changes are in the low and intermediate energy range this will be relevant for instruments with a high response in the low and intermediate energy range, i.e. they would read higher closer to the source than further away.

Fig. 5 shows, in turn, the distance dependence of the spectrum-averaged fluence-to-dose equivalent conversion coefficients. As expected, at closer distances the spectrum is softer [21]. As clearly shown by Fig. 5, the spectrum can be regarded as "stabilized" for distance larger than 50 cm and the conversion coefficients do not change for distances greater than 100 cm. A compromise value of 75 cm is the recommended distance for calibrating personal dosemeters. These aspects should be carefully considered for measurements near the assembly.

When submersing the moderating assembly in air, very small effects can be observed in the spectrumintegrated quantities (See Table 1). By contrast, no visible differences are found between "vacuum" and "air" in the plotted spectrum. For this reason, the "air" spectrum was not represented. The quotient of the total neutron fluence in air / vacuum provides the net air inscatter minus outscatter increase in fluence at 100 cm, and its value for proposed spectrum is 1.016 compared with 1.044 reported in Annex D of ISO 8529-2:2000 [22].

Removing the cadmium shell has large impact on the spectrum as well as on the integral quantities, as shown in Fig. 2 and Table 1. This is clearly due to the presence of thermalized neutrons.



Figure 5. Variation of fluence-to-ambient dose equivalent and fluence-to-personal dose equivalent conversion coefficients with distance for D<sub>2</sub>O-moderated <sup>252</sup>Cf.

# Effect of D<sub>2</sub>O purity

As reported in literature [23, 24], existing moderating assemblies show slightly different D<sub>2</sub>O purity. To help the users of the revised Standard to understand which field properties may be expected from a specific purity value, the result of a sensitivity study on D<sub>2</sub>O purity is here reported. Compared to all other assembly parameters studied, D<sub>2</sub>O purity has the greatest impact.

The effects of variations in the  $D_2O$  purity, from 100 % (proposed assembly) to 95 % (i.e., with 5 % light water) are shown in Fig. 6 (a) and (b) and Table 3. All other parameters are those of the "proposed assembly".



Figure 6. (a) The variation of D<sub>2</sub>O purity from 95 % to 100 % for D<sub>2</sub>O-moderated  $^{252}$ Cf neutron spectra, and (b) expressed as ratios to the 100 % D<sub>2</sub>O-moderated  $^{252}$ Cf spectrum. Only a few purity variations are shown: 100%, 99%, 97.5% and 95%.

D2O [ %]	Φ	<b>f</b> abs	$\overline{E}_{\Phi}$	$\overline{E}_{\mathrm{H}^*}$	<b>h</b> <sup>*</sup> (10)	π <sub>ρ</sub> (10,0°)
	[cm- <sup>2</sup> s- <sup>1</sup> ]	[%]	[MeV]	[MeV]	[uSv/h]	[pSv cm <sup>2</sup> ]
100.0	7.00E-06	11.40	0.57	1.98	114.88	115.75
99.5	6.95E-06	12.18	0.57	1.99	115.33	116.17
99.0	6.88E-06	12.96	0.58	1.99	115.92	116.89
98.5	6.82E-06	13.74	0.58	2.00	116.53	117.51
98.0	6.76E-06	14.53	0.59	2.00	117.16	118.63
97.5	6.69E-06	15.32	0.59	2.01	117.82	119.29
97.0	6.63E-06	16.11	0.60	2.01	118.49	119.29
95.0	6.38E-06	19.24	0.62	2.03	121.34	122.11

Table 3. Magnitudes for quantities of interest for the  $D_2O$ -moderated  $^{252}Cf$  neutron source as a function of  $D_2O$  purity. Magnitudes for the proposed assembly are highlighted.

From Figure 6 (b), it is clear that decreasing D<sub>2</sub>O purity decreases the fluence between 0.1 keV and 0.3 MeV, and increases the fluence between the Cd cut-off and 0.1 keV, and above 0.3 MeV. As the net effect of reducing D<sub>2</sub>O purity is to introduce neutron absorption at low energies, the fluence-average energy is higher at lower purities (from 0.57 at 100 % to 0.62 MeV at 95 %, as is shown in Table 3). The same applies to the ambient-dose-equivalent-average energy, changing from 1.98 MeV at 100 % to 2.03 MeV at 95 %. The spectrum-averaged conversion coefficients  $\overline{h^*}(10)$  and  $\overline{h_p}(10,0^\circ)$  follow the same behaviour,  $\overline{h^*}(10)$  changing from 115 pSv·cm<sup>2</sup> at 100 % to 121 pSv·cm<sup>2</sup> at 95 %, and  $\overline{h_p}(10,0^\circ)$  from 116 pSv·cm<sup>2</sup> at 100 % to 122 pSv·cm<sup>2</sup> at 95 %.

As summarized in Table 6, a -1 % change in D<sub>2</sub>O purity in the analysed range from 100 to 95%, produces +14 % in absorbed fraction, -1.8 % in fluence, and -0.8 % in  $H^*(10)$ . For this reason, it is very important to take into account the precise value for D<sub>2</sub>O concentration.

#### **Effects of construction parameters**

Sphere diameter	Ф	<b>f</b> <sub>abs</sub>	$\overline{E}_{\mathbf{\Phi}}$	$\overline{E}_{\mathbf{H}^*}$	<b>h</b> <sup>*</sup> (10)	h <sub>ρ</sub> (10,0°)
[cm]	[cm <sup>-2</sup> ]	[ %]	[MeV]	[MeV]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]
30.3	6.97E-06	11.84	0.56	1.98	113.36	113.82
30.2	6.99E-06	11.70	0.57	1.98	114.35	114.46
30.1	7.00E-06	11.36	0.57	1.98	113.64	115.10
30	7.00E-06	11.40	0.57	1.98	114.88	115.75
29.9	7.02E-06	11.25	0.57	1.98	115.23	116.39
29.8	7.03E-06	11.11	0.57	1.98	115.70	117.04
29.7	7.04E-06	10.97	0.58	1.98	116.18	117.70

Table 4. Magnitudes of interest for the D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron source as a function of sphere diameter. Magnitudes for the proposed assembly are highlighted.

Small construction differences between different moderating assemblies are frequent in practice [23,13]. To represent realistic deviations of practical assemblies from the proposed one, the following ranges were considered in this study:

- D<sub>2</sub>O sphere diameter: from 29.7 cm to 30.3 cm;
- Internal diameter of the source positioning tube: from 12.8 mm to 25 mm. The proposed assembly corresponds to 12.8 mm.

As shown in Tables 6 and 7, the assumed variability of the construction parameters is so small that integral quantities vary linearly with them. As expected, an increase in the D<sub>2</sub>O sphere diameter, Table 4, causes an increase in the absorbed fraction. From this it follows a decrease in total fluence and in the fluence-average energy and in the conversion coefficients values. The dose-equivalent average energy increases slightly with sphere diameter because thermal absorptions produce a shift in balance increasing the relative proportion of those neutrons with higher contribution to dose. On the contrary, an increase in tube diameter implies a decrease in effective D<sub>2</sub>O diameter and effects are the opposite, Table 5. See a summary of these results in Table 6.

Tube diameter	ф	<b>f</b> abs	$\overline{E}_{\mathbf{\Phi}}$	$\overline{E}_{\mathrm{H}^*}$	<i>॑H</i> <sup>∗</sup> (10)	<b>h</b> <sup>*</sup> ( <b>10</b> )	π <sub>ρ</sub> (10,0°)
[mm]	[cm <sup>-2</sup> s <sup>-1</sup> ]	[ %]	[MeV]	[MeV]	[uSv/h]	[pSv cm <sup>2</sup> ]	[pSv cm <sup>2</sup> ]
12.8	7.00E-06	11.40	0.57	1.98	2.89E-06	114.88	115.75
15	6.99E-06	11.36	0.57	1.98	2.90E-06	117.59	116.19
20	6.97E-06	10.90	0.59	1.99	2.95E-06	117.87	118.69
25	6.96E-06	10.41	0.60	1.99	3.01E-06	120.52	121.31

Table 5. Magnitudes of interest for the D<sub>2</sub>O-moderated <sup>252</sup>Cf neutron source as a function of source-positioning tube diameter. Magnitudes for the proposed assembly are highlighted.

Quantity and variation	$\Delta f_{abs} / f_{abs}$	$\Delta \Phi / \Phi$	$\Delta H^{*}(10) / H^{*}(10)$
-1 % in D <sub>2</sub> O purity	+14 %	-1.8 %	-0.8 %
+1 mm in D <sub>2</sub> O sphere diameter	+1.3 %	-0.16 %	-0.6 %
+1 mm in the internal tube diameter	-0.8 %	-0.04 %	+0.4 %

Table 6. Variations in the  $D_2O$ -moderated <sup>252</sup>Cf field as a result of small variations in the  $D_2O$  purity and construction details.

From Table 6 it is possible to evaluate the effect of small variations in  $D_2O$  composition or sphere diameter and internal tube diameter. The most important effect is related with variation in  $D_2O$  purity. A 1% decrease produces an increase of 14% in relative absorption. Variations of 1 mm in sphere diameter or internal tube diameter have a limited influence in absorption, fluence or ambient dose equivalent.

# Homogeneity across a 30 cm x 30 cm surface at 100 cm

In the original work by Ing and Cross [7] the effect of central tube on the spherical symmetry was studied as a function of the latitude from the equatorial plane. They determined average energy values with 15° angular latitude intervals from 15° to 75°. Negligible variations were found.

In this work, the spectral homogeneity has been evaluated on a 30 cm x 30 cm surface at 100 cm from the source centre. Integral quantities were calculated in a grid with 5 cm steps. For symmetry reasons, only one quadrant was simulated. Results for normalized fluence to central value and conversion coefficients,  $\overline{h^*}(10)$  and  $\overline{h_p}(10,0^\circ)$ , are shown in Table 7. No corrections for non-zero angle of

incidence have been considered. Conversion coefficients are basically constant over the surface, establishing robust basis for studying dosemeter irradiation on the ISO water phantom [19].

Φ	[normalized]			
Z				
15	0.98	0.98	0.97	0.96
10	0.99	0.99	0.98	0.97
5	1	1	0.99	0.98
0	1	1	0.99	0.98
	0	5	10	15

$\overline{h^*}(10)$	[pSvcm <sup>2</sup> ]				<u>h_</u> p(10,0°)	[pSvcm <sup>2</sup> ]			
Z					Z				
15	114.93	114.93	114.93	114.94	15	115.80	115.80	115.80	115.81
10	114.91	114.90	114.90	114.91	10	115.77	115.77	115.77	115.78
5	114.89	114.89	114.89	114.89	5	115.76	115.76	115.76	115.76
0	114.88	114.88	114.88	114.88	0	115.75	115.75	115.75	115.76
	0	5	10	15		0	5	10	15

Table 7. Dosimetric parameters in the first quadrant of a 30 cm x 30 cm grid. Fluence is normalized to the value at the centre of the phantom face.

# CONCLUSIONS

In support of the revision of ISO 8529-1, numerical studies have been performed covering the  $^{252}$ Cf and D<sub>2</sub>O-moderated  $^{252}$ Cf fields. Concerning the  $^{252}$ Cf, some inconsistencies in the current tabulation were resolved by adopting the ENDF/B-VIII.0  $^{252}$ Cf spectrum (with partial re-binning).

As the current D<sub>2</sub>O-moderated <sup>252</sup>Cf spectrum comes from calculations performed in the early 80s, new data were generated using MCNP6.2 with ENDF/B-VIII.0 cross sections.  $S(\alpha,\beta)$  thermal treatment was included for D<sub>2</sub>O and <sup>56</sup>Fe. A simplified model of geometry and materials, very similar to that in the original work from Ing and Cross, was adopted for these calculations. Calculations were also made to account for different construction details of the moderating assembly such as the sphere diameter, D<sub>2</sub>O purity, and source positioning system. Lastly, in order to study the irradiation of dosemeters on phantom, the variation of spectrum and fluence across a 30 cm x 30 cm surface placed at 1 m was evaluated.

The new spectrum-averaged fluence to ambient dose equivalent conversion coefficient is  $\overline{h^*}(10)$  = 115 pSv.cm<sup>2</sup>, 10 % larger than that in 8529-1:2001 (105 pSv·cm<sup>2</sup>). The spectrum-averaged fluence to personal dose equivalent at 0° conversion coefficient is  $\overline{h_p}(10,0^\circ)$  = 116 pSv·cm<sup>2</sup> (to be compared with 110 pSv·cm<sup>2</sup> from ISO 8529-3:1998). These new values will have a direct influence in the determination of the calibration factors of neutron measurement devices.

In addition, the fraction of neutrons that are absorbed in the moderating assembly, or absorbed fraction, was revised. The new value is  $f_{abs,4\pi}$  =11.4 % very close to 11.5 % in 8529-1:2001, which confirms the validity of the simulated model with the most recent versions of MCNP and ENDF/B cross section libraries.

As it was expected, this work proves that when construction parameters are assumed to vary over reasonable intervals, they cause linear variations in the spectrum-integrated quantities. Among the

construction parameters, D<sub>2</sub>O purity proved to be the most important in determining the spectrum shape and integral quantities above thermal energies. A -1 % in D<sub>2</sub>O purity causes -1.8 % in fluence, +14 % in absorbed fraction, and -0.8 % in  $H^*(10)$ .

The spectrum and integral quantities were found to be essentially uniform across a 30 cm  $\times$  30 cm surface 100 cm from the source centre. Compliance with this condition is especially important for the simultaneous irradiation of several personal dosemeters on a slab phantom, positioned inside a circle of 15 cm diameter, as is described in ISO-8529-3.

Although, D<sub>2</sub>O moderated <sup>252</sup>Cf source is an internationally recognised calibration source the practical implementation of this standard may differ from one to other laboratory, with specific constructive details and heavy water composition, especially for old installations. In this sense, it is necessary to establish a reference, as simple as possible, and to know the effect of these constructive differences to provide identical calibrations for the same device, independently of the laboratory.

This work constitutes a validation of the original model performed with the last MCNP version and an improvement with new available data. Some changes found are significant, e.g. the new spectrum-averaged conversion coefficients for D<sub>2</sub>O moderated <sup>252</sup>Cf have changed by more than the uncertainty of 4% recommended in the current version of ISO 8529-3. These changes should be taken into account by the neutron standards laboratories.

#### REFERENCES

- 1. Thomas. D., Bedogni. R., Méndez. R., Thompson. A. and Zimbal. A. *Revision of ISO 8529 Reference neutron radiations*. Radiat. Prot. Dosim. **180**. 21-24 (2018).
- 2. Bedogni. R., Domingo. C., Roberts. N., Thomas. D.J., Chiti. M., Esposito. A., Garcia. M.J., Gentile. A., Liu. Z.Z., De-San-Pedro. M. *Investigation of the neutron spectrum of americium-beryllium sources by Bonner sphere spectrometry*. Nucl. Instr. Meth. **763** (2014) 547-552
- 3. International Organization for Standardization, *Reference neutron radiations Part 1: Characteristics and methods of production*, ISO 8529-1:2001, (Geneva, Switzerland).
- 4. Croft, S. and Miller, K. A. *Group representation of the prompt fission neutron spectrum of* <sup>252</sup>*Cf.* ESARDA Bull. **46**, 112–114 (2011).
- 5. Thomas, D. J. *The* <sup>252</sup>*Cf neutron spectrum in ISO Standard 8529*. ESARDA Bull. **51**, 45–50 (2014).
- 6. Brown D.A. et al., ENDF/B-VIII.0: *The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data.* Nucl. Data Sheets (N.Y. N.Y.). 2018, 148 pp. 1–142
- 7. Ing H., Cross W.G., Spectral and Dosimetric Characteristics of a D<sub>2</sub>O-Moderated <sup>252</sup>Cf Calibration Facility. Health Phys. 1984, **46** (1) pp. 97–106
- Coveyou R. R., Sullivan J. G., Carter H. P., Irving D. C., Freestone, Jr. R. M. and Kam F. B. K., 1965, "05R, a General-purpose Monte Carlo Neutron Transport Code", Oak Ridge National Laboratory, Oak Ridge, TN, ORNL-3622.
- 9. Kinsey, R. (1979). *ENDF/B Summary Doc.*". BNL-NCS-17541 (ENDF 201), 3rd. ENDF/B-V, Brookhaven National Laboratory, Upton
- 10. Werner, C.J. (editor), "MCNP User's Manual Code Version 6.2", LA-UR-17-29981 (2017).
- 11. Mannhart, W. (1989). Status of the Cf-252 fission neutron spectrum evaluation with regard to recent experiments (INDC(NDS)--220). Lemmel, H.D. (Ed.). International Atomic Energy Agency (IAEA).
- 12. ICRP Publication 74, Conversion Coefficients for use in Radiological Protection against *External Radiation*, Annals of the ICRP, Vol. **26**, No.3/4 (1996).
- 13. Kowatari, M., Fujii, K., Tsutsumi M., Bong-Hwan, K., et al. (2008) *An Inter-comparison of the Neutron Calibration Fields by D<sub>2</sub>O Moderated 252Cf Source at JAEA and KAERI*, Journal of Nuclear Science and Technology, **45** : sup5, 217-220.
- Kowatari, M., Fujii, K., Takahashi, M., et al., *Evaluation of the characteristics of the neutron* reference field using D2O-moderated <sup>252</sup>Cf source, Radiat. Prot. Dosim., Volume **126**, Issue 1-4, (2007) 138–144.
- 15. Kluge, H., Rasp, H., Hunt, J.B., An intercomparison of the D<sub>2</sub>O-Moderated <sup>252</sup>Cf reference neutron sources at PTB and NPL. Radiat. Prot. Dosim. 1985, **11**, 61-63.
- 16. McConn Jr., R.J. et al. Compendium of Material Composition Data for Radiation Transport Modelling Report PNNL-15870 Rev. 1 (PNNL, USA, March 4, 2011).
- 17. Coursey, J.S., Schwab, D.J., Tsai, J.J., et al. Atomic Weights and Isotopic Compositions with Relative Atomic Masses NIST Standard Reference Database 144 "https://www.nist.gov/pml/atomic-weights-and-isotopic-compositions-relative-atomicmasses" (Last update: January 2015).
- 18. Ing H., Cross W.G., *Calculated spectra for the dosimetry of D*<sub>2</sub>O-*Moderated neutrons*. Health Phys. 1977, **32** (May) pp. 351-357.
- 19. International Organization for Standardization, Reference neutron radiations *Part 3: Calibration of area and personal dosimeters and determination of response as a function of energy and angle of incidence*, ISO 8529-3:1998, (Geneva, Switzerland).
- 20. Hawkes, N.P., Freedman, R., Tagziria, H., Thomas, D.J., Measurements and calculation of the emission anisotropy of an X1 <sup>252</sup>Cf neutron source. *Radiat. Prot. Dosim.* 2007, **126** pp. 78-82.

- 21. Kluge, H., Weise. K., Hunt, J.B., Calibration of Neutron Sensitive Spherical Devices with Bare and D<sub>2</sub>O-Moderated <sup>252</sup>Cf Sources in Rooms of Different Sizes. Radiat. Prot. Dosim. 1990, **32**, 233-244.
- 22. International Organization for Standardization, Reference neutron radiations *Part 2: Calibration fundamentals of radiation protection devices related to the basic quantities characterizing the radiation field*, ISO 8529-2:2000, (Geneva, Switzerland).
- 23. Kluge, H., Rasp, W. and Hunt, B. *An intercomparison of the D*<sub>2</sub>O *moderated* <sup>252</sup>Cf *reference neutron sources at PTB and NPL* Radiat. Prot. Dosim. **11**, No 1, (1985) 61-63.
- 24. Masuda, A., Harano, H., Matsumoto, T., et al. "Development of a neutron standard field using a heavy-water moderated 252Cfsource at NMI1-AIST", *Prog. Nucl. Sci. Technol.*, vol. **4**, pp. 400-403, 2014.

ANNEX. Tables

E <sub>i</sub> (MeV)	<i>B</i> <sup><i>i</i></sup> (s <sup>-1</sup> )	E <sub>i</sub> (MeV)	<i>B</i> <sub>i</sub> (s <sup>-1</sup> )	Ei (MeV)	<i>B</i> <sub>i</sub> (s <sup>-1</sup> )
1.00E-08	9.19E-13	3.05E-01	1.48E-02	3.55E+00	2.90E-02
2.15E-08	2.87E-12	3.55E-01	1.53E-02	3.85E+00	2.42E-02
4.64E-08	9.18E-12	4.05E-01	1.58E-02	4.15E+00	2.02E-02
1.00E-07	2.90E-11	4.55E-01	1.61E-02	4.45E+00	1.68E-02
2.15E-07	9.09E-11	5.05E-01	1.64E-02	4.75E+00	1.39E-02
4.64E-07	2.90E-10	5.55E-01	1.66E-02	5.05E+00	1.81E-02
1.00E-06	9.19E-10	6.05E-01	1.67E-02	5.55E+00	1.31E-02
2.15E-06	2.87E-09	6.55E-01	1.68E-02	6.05E+00	9.49E-03
4.64E-06	9.18E-09	7.05E-01	1.68E-02	6.55E+00	6.83E-03
1.00E-05	2.90E-08	7.55E-01	1.68E-02	7.05E+00	4.91E-03
2.15E-05	9.09E-08	8.05E-01	1.68E-02	7.55E+00	3.52E-03
4.64E-05	2.90E-07	8.55E-01	1.67E-02	8.05E+00	2.52E-03
1.00E-04	9.19E-07	9.05E-01	1.66E-02	8.55E+00	1.81E-03
2.15E-04	2.87E-06	9.55E-01	3.12E-02	9.05E+00	1.29E-03
4.64E-04	9.18E-06	1.05E+00	3.22E-02	9.55E+00	9.27E-04
1.00E-03	2.90E-05	1.15E+00	3.15E-02	1.01E+01	6.62E-04
2.15E-03	9.06E-05	1.25E+00	3.06E-02	1.06E+01	4.74E-04
4.64E-03	2.89E-04	1.35E+00	2.97E-02	1.11E+01	3.39E-04
1.00E-02	8.97E-04	1.45E+00	2.87E-02	1.16E+01	2.42E-04
2.15E-02	1.42E-03	1.55E+00	2.77E-02	1.21E+01	1.73E-04
3.50E-02	2.64E-03	1.65E+00	2.67E-02	1.26E+01	1.23E-04
5.50E-02	3.13E-03	1.75E+00	2.56E-02	1.31E+01	8.75E-05
7.50E-02	3.53E-03	1.85E+00	2.46E-02	1.36E+01	6.22E-05
9.50E-02	3.87E-03	1.95E+00	4.60E-02	1.41E+01	4.78E-05
1.15E-01	4.17E-03	2.15E+00	4.20E-02	1.46E+01	6.27E-05
1.35E-01	6.74E-03	2.35E+00	3.81E-02	1.59E+01	2.15E-05
1.65E-01	7.23E-03	2.55E+00	3.45E-02	1.69E+01	1.09E-05
1.95E-01	7.66E-03	2.75E+00	3.11E-02	1.79E+01	6.12E-06
2.25E-01	8.02E-03	2.95E+00	4.08E-02	1.91E+01	2.18E-06
2.55E-01	1.41E-02	3.25E+00	3.44E-02	2.00E+01	

TT 1 1 A	<b>T</b> 71 C		• •		1 / 1	25200
Lable A	values of g	group source	emission	rate for	unmoderated	232( .Ť
rubic 11.	values of a	Broup source	cimobion	fute for	unnoucluted	01

E <sub>i</sub> (MeV)	<i>B</i> <sub>i</sub> (s <sup>-1</sup> )	<i>E</i> i (MeV)	<i>B<sub>i</sub></i> (s <sup>-1</sup> )	<i>E</i> i (MeV)	<i>B<sub>i</sub></i> (s <sup>-1</sup> )
1.00E-08	4.50E-09	3.05E-01	8.75E-03	3.55E+00	6.14E-03
2.15E-08	7.99E-08	3.55E-01	5.84E-03	3.85E+00	6.63E-03
4.64E-08	5.38E-08	4.05E-01	3.19E-03	4.15E+00	5.27E-03
1.00E-07	1.17E-17	4.55E-01	5.02E-03	4.45E+00	4.96E-03
2.15E-07	1.17E-03	5.05E-01	5.63E-03	4.75E+00	4.30E-03
4.64E-07	1.33E-02	5.55E-01	5.33E-03	5.05E+00	5.45E-03
1.00E-06	2.06E-02	6.05E-01	5.05E-03	5.55E+00	3.86E-03
2.15E-06	2.36E-02	6.55E-01	4.73E-03	6.05E+00	3.10E-03
4.64E-06	2.63E-02	7.05E-01	4.54E-03	6.55E+00	2.28E-03
1.00E-05	2.93E-02	7.55E-01	4.40E-03	7.05E+00	1.53E-03
2.15E-05	3.27E-02	8.05E-01	3.98E-03	7.55E+00	1.13E-03
4.64E-05	3.57E-02	8.55E-01	3.28E-03	8.05E+00	7.99E-04
1.00E-04	4.08E-02	9.05E-01	2.65E-03	8.55E+00	5.90E-04
2.15E-04	4.53E-02	9.55E-01	3.50E-03	9.05E+00	4.42E-04
4.64E-04	5.07E-02	1.05E+00	5.46E-03	9.55E+00	3.09E-04
1.00E-03	5.55E-02	1.15E+00	6.41E-03	1.01E+01	2.21E-04
2.15E-03	6.03E-02	1.25E+00	5.05E-03	1.06E+01	1.71E-04
4.64E-03	6.30E-02	1.35E+00	6.00E-03	1.11E+01	1.11E-04
1.00E-02	6.68E-02	1.45E+00	6.02E-03	1.16E+01	8.00E-05
2.15E-02	4.05E-02	1.55E+00	5.70E-03	1.21E+01	6.20E-05
3.50E-02	3.80E-02	1.65E+00	5.63E-03	1.26E+01	4.49E-05
5.50E-02	2.53E-02	1.75E+00	5.66E-03	1.31E+01	3.20E-05
7.50E-02	1.83E-02	1.85E+00	4.94E-03	1.36E+01	2.32E-05
9.50E-02	1.44E-02	1.95E+00	1.06E-02	1.41E+01	1.82E-05
1.15E-01	1.17E-02	2.15E+00	1.16E-02	1.46E+01	2.42E-05
1.35E-01	1.39E-02	2.35E+00	1.12E-02	1.59E+01	8.81E-06
1.65E-01	1.11E-02	2.55E+00	9.41E-03	1.69E+01	4.68E-06
1.95E-01	9.18E-03	2.75E+00	8.33E-03	1.79E+01	2.65E-06
2.25E-01	7.84E-03	2.95E+00	1.10E-02	1.91E+01	8.13E-07
2.55E-01	1.09E-02	3.25E+00	7.23E-03	2.00E+01	

 Table B. Modelled values of group source emission rate at 100 cm from the centre of the D2O-moderated <sup>252</sup>Cf