Neutron Shielding for a ^{252}Cf Source

Héctor René Vega-Carrillo*, Eduardo Manzanares-Acuña, Víctor M. Hernández-Dávila
Unidades Académicas de Estudios Nucleares e Ingeniería Eléctrica
Universidad Autónoma de Zacatecas
C. Ciprés 10, Fracc. La Peñuela, 98068 Zacatecas, Zac. México
fermineutron@yahoo.com; emanz@uaz.edu.mx; victorh@uaz.edu.mx

Eduardo Gallego, Alfredo Lorente
Depto. de Ingeniería Nuclear, ETS Ingenieros Industriales
Universidad Politécnica de Madrid
C/José Gutiérrez Abascal 2, 28006 Madrid, Spain
eduardo.gallego@upm.es; alfredo.lorente@upm.es

Abstract

To determine the neutron shielding features of water-extended polyester a Monte Carlo study was carried out. Materials with low atomic number are predominantly used for neutron shielding because these materials effectively attenuate neutrons, mainly through inelastic collisions and absorption reactions. During the selection of materials to design a neutron shield, prompt gamma production as well as radionuclide production induced by neutron activation must be considered. In this investigation the Monte Carlo method was used to evaluate the performance of a water-extended polyester shield designed for the transportation, storage, and use of a ^{252}Cf isotopic neutron source. During calculations a detailed model for the ^{252}Cf and the shield was utilized. To compare the shielding features of water extended polyester, the calculations were also made for the bare ^{252}Cf in vacuum, air and the shield filled with water. For all cases the calculated neutron spectra was utilized to determine the ambient equivalent neutron dose at four sites around the shielding. In the case of water extended polyester and water shielding the calculations were extended to include the prompt gamma rays produced during neutron interactions, with this information the Kerma in air was calculated at the same locations where the ambient equivalent neutron dose was determined.

1. INTRODUCTION

The isotopic neutron sources are relatively small, compact, portable and easy to handle. These sources offer a simple and economical way to use Neutron Activation Analysis in industries for process control; are widely utilized to calibrate radiation protection devices[1] and to carried out experiments in biology and medicine.

* Corresponding author.

Many transuranium elements which undergo α-decay also disintegrate by spontaneous fission releasing neutrons in the process. In this group is ^{252}Cf that in 3.2% decays by spontaneous fission releasing 3.7 neutrons per fission, each disintegration yields an average of 0.12 neutrons through, $^{252}\text{Cf} \rightarrow {}^{140}\text{Xe} + {}^{108}\text{Ru} + 4\text{ n} + \text{Q}$ and $^{252}\text{Cf} \rightarrow {}^{140}\text{Cs} + {}^{109}\text{Tc} + 3\text{ n} + \text{Q}$. The main features of ^{252}Cf are shown in Table I.

Table I. Nuclear and radiological characteristics of ^{252}Cf

Feature		Value
Mode of decay	α-emission Spontaneous fission	96.9% 3.1%
Half life	α-decay Spontaneous fission Effective	2.731 ± 0.007 y 85.5 ± 0.5 y 2.646 ± 0.004 y
Neutron emission rate		2.4 x 10^{12} neutrons-s^{-1}-g^{-1}
Neutrons emitted per fission		3.76
Average neutron energy		2.348 MeV
Average α-particle energy		6.117 MeV
Gamma emission rate		1.3E(13) γ- s^{-1}-g^{-1}
Dose equivalent average energy		2.4 MeV
Specific neutron dose equivalent rate		6.5 x 10^{-3} Sv-s^{-1}-g^{-1} at 1 m
Specific photon dose equivalent rate		3.1x10^{-4} Sv-s^{-1}-g^{-1} at 1 m
Ambient dose equivalent, H*(10)		380 pSv-cm^2
Personal dose equivalent H_p(10)		400 pSv-cm^2
Decay heat	α-decay Spontaneous fission	18.8 W-g^{-1} 19.7 W-g^{-1}
Source volume (excluding void space for He)		< 1 cm^3 – g^{-1}

The Nuclear Engineering Teaching Laboratory (NETL) at the University of Texas at Austin has a ^{252}Cf isotopic neutron source from Oak Ridge National Laboratory (ORNL) through the U.S. Department of Energy's Californium University Loan Program. In December 31st, 1994, the mass of ^{252}Cf source was 16.42 μg, with a neutron emission rate of 2.31 x 10^6 s^{-1}-μg^{-1}. The Cf isotopic composition is shown in Table II.

Table II. Isotopic composition of ^{252}Cf neutron source

Nuclide	Isotopic composition [o/a]
Cf – 249	11.04
Cf – 250	12.10
Cf – 251	4.05
Cf – 252	72.81
Cf – 253	< 0.001
Cf – 254	< 0.00013

Californium is in the form of oxysulfate, $Cf_2O_2SO_4$; the source has a double encapsulation made of standard stainless steel 304L [2].

The neutron source was stored in a water-extended polyester shield structure, originally designed and constructed to transport and store a 1 milligram ^{252}Cf source [3]. The shield was built in 1971, and the personnel involved in the construction are no longer The University of Texas at Austin, hence information about the rationale of using WEP for the construction is not available. The shield had been stored outdoors for some years, and was beginning to show water/rust damage until its resurrection/restoration in 1994.

WEP resin is a desirable material to be used as neutron shielding because can be easily prepared, has high water content, it is resistant to fire and their mechanical strength is between wood and concrete.[4] Unsaturated polyester resins readily emulsify with water. When properly catalyzed, the emulsion cures by an exothermic chemical reaction into a hard material, similar in appearance to a fine-grained plaster. WEP consists of equal parts of styrene monomer and polyester resin combined with up to 65-70% by weight of water. The final cured emulsion has an average atomic number of 3.50 grams/gram-mol and an average mass density of 1.1 grams/cm^3. [2, 3] To compare this material with those normally utilized in neutron shield Table III shows the elemental concentration, in weight percent, for hydrogen, carbon, and oxygen for WEP, water and polymethyl methacrylate (Lucite). In terms of hydrogen content, WEP compares favorably to Lucite and is close to water

Table III. Percent in weight of WEP, Water and Lucite

Element	WEP	Water	Lucite
H	9.7	11.2	8.0
C	25.3	0.0	60.0
O	65.0	88.8	32.0

The Laboratory of the Nuclear Engineering Department of the Polytechnic University of Madrid (DIN-UPM) owns a facility for neutron dosimetry research with two isotopic sources of ^{241}AmBe of 74 and 111 GBq. Substantial efforts has been realized to have this facility functional. Nowadays this is the first and unique facility in Spain with the capability to perform neutron instrumentation calibration and to made controlled irradiation with neutron fields.[4] Having only ^{241}AmBe sources the facility has several limitations; therefore a ^{252}Cf source is going to be purchased. The nuclear properties of ^{252}Cf require a specific investigation about the handling, shielding and storage conditions.

The aim of this investigation is to study the shielding feature for neutrons [5], and prompt gamma-rays produced during the transport of neutrons in WEP shielding and to compare with a similar shield made of water. The study was carried doing Monte Carlo calculation using MCNP 4C [6] code, also the ambient dose equivalent (H*(10)) was calculated at four points around the shield structure and are compared with the H*(10) produced by the bare source in vacuum, in air and the water shielding.

2. MATERIALS AND METHODS

The NETL WEP shielding is shown in Figure 1. This has two radial ports and one axial port; during this investigation ports were filled with WEP enclosures therefore it was studied as a single piece. The WEP shield is a right cylinder, 121.9 cm diameter by 132.0 cm height. It was constructed from a 0.152 cm thick stainless steel pipe which had been welded to a 1.27 cm-thick steel base plate. A 0.635 cm thick top plate was welded to the top. The geometry and composition of the WEP shield structure and details of the source encapsulation, like dimensions and elemental concentration, were modeled using the 3-D modeling features of MCNP code.

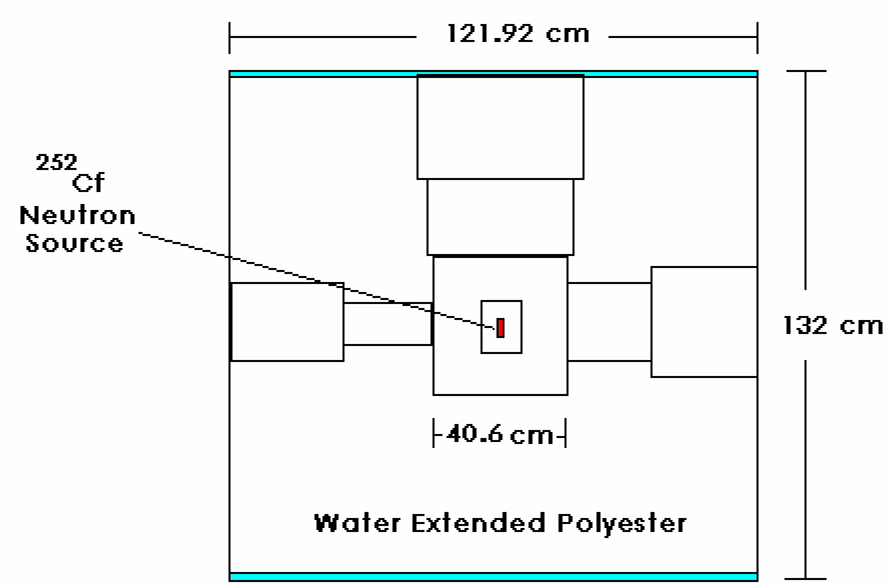

Figure 1. NETL WEP neutron shielding

The ^{252}Cf source is doubly encapsulated in 304L stainless steel. Cf oxysulfate was modeled as a point located in the cell of the encapsulation that actually contained the ^{252}Cf source. The Cf compound is pressed with an aluminum pellet, and there is a vacuum gap on the top. This gap was modeled as air-filled in the Monte Carlo model. As source term the reference spectra recommended by ISO [7] was utilized.

The Monte Carlo model is shown in Figure 2. Here, points A, B, C and D are these sites where neutron spectra and H*(10) were calculated. A large spherical cavity filled with air was included to simulate the room, to take into account the neutron skyshine. Also a concrete base was used below the WEP shielding to include the contribution of neutron groundshine.

To compare the WEP shielding performance, three extra calculations were also carried out: With the bare source capsule in vacuum, with the source capsule in the air and with the source capsule in the same shielding but assuming it is made of water instead of WEP. For water and WEP materials the prompt gamma rays induced during neutron transport was included and the Kerma in air due to photons, at sites A, B, C, and D, were also calculated. All calculations were performed with MCNP 4C and with the ENDF/B6 [6] neutron cross sections, the number of neutron histories was different for each case, but large enough to have an uncertainty less than 5%.

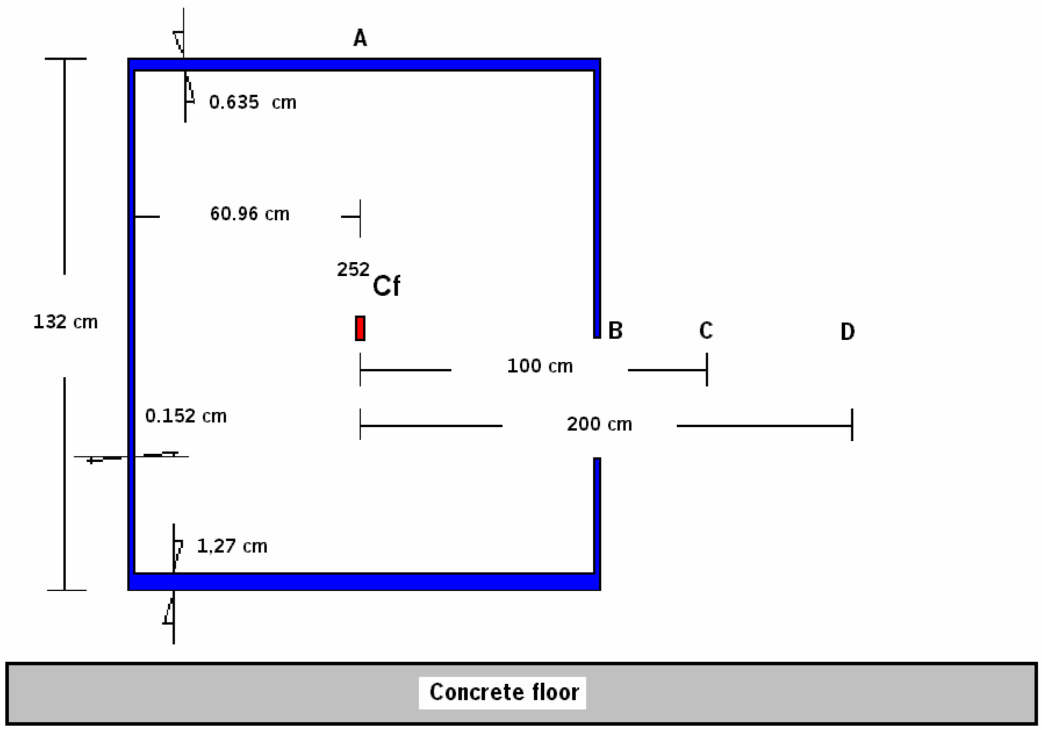

Figure 2. Monte Carlo model of WEP shield structure.

3. RESULTS AND DISCUSSION

In Table IV the calculated H*(10) produced by a ^{252}Cf source with a strength of 1 neutron/s, in vacuum, air, water and WEP is shown. Comparing the H*(10) at all sites in vacuum and air it can be noticed that the presence of air increases the dose, the relative increment being proportional to the distance. The reason for that effect is the skyshine of neutrons in air. In vacuum, the ^{252}Cf source has an ambient dose equivalent coefficient of 380 pSv-cm^2 at 1 m (point C); this is the ratio between H*(10) and the neutron fluence rate at this site (8.0672×10^{-6} cm^{-2}), this value is in agreement with the coefficient 385 pSv-cm^2, that is reported by ISO for a ^{252}Cf point-like source in vacuum [7].

Table IV. Monte Carlo calculated H*(10) due to ^{252}Cf with a 1 n/s source strength

Point	Vacuum [pSv]	Air [pSv]	WEP shielding [pSv]	Water shielding [pSv]
A	$4.7965 \times 10^{-3} \pm 0.01\%$	$6.1550 \times 10^{-3} \pm 0.03\%$	$9.9755 \times 10^{-7} \pm 4.9\%$	$1.3381 \times 10^{-6} \pm 4.9\%$
B	$7.9747 \times 10^{-3} \pm 0.01\%$	$9.1300 \times 10^{-3} \pm 0.05\%$	$3.0420 \times 10^{-6} \pm 4.9\%$	$3.9536 \times 10^{-6} \pm 4.8\%$
C	$3.0656 \times 10^{-3} \pm 0.01\%$	$3.7571 \times 10^{-3} \pm 0.09\%$	$8.3155 \times 10^{-7} \pm 4.9\%$	$9.7042 \times 10^{-7} \pm 2.6\%$
D	$7.6644 \times 10^{-4} \pm 0.01\%$	$1.0511 \times 10^{-3} \pm 0.03\%$	$1.8784 \times 10^{-7} \pm 4.4\%$	$2.1583 \times 10^{-7} \pm 2.8\%$

At point B the source in air produces an ambient dose equivalent 3001 times larger that the source inside the WEP shielding and 2309 times larger that the shield made with water. At A, B, C and D locations the doses are larger than 10% with the water in comparison with WEP shielding, therefore WEP has a better performance. According with NCRP [8], one gram of ^{252}Cf produces 2.4 $x \cdot 10^{12}$ neutrons per second, using this parameter the ambient dose equivalent rate produced by 1 μg of ^{252}Cf is shown in Table V

Table V. H*(10) due to 1 μg of ^{252}Cf

Point	Vacuum [nSv/s]	Air [nSv/s]	WEP shielding [nSv/s]	Water shielding [nSv/s]
A	11.51	14.77	$2.39 \text{ x}\cdot 10^{-3}$	$3.21 \text{ x}\cdot 10^{-3}$
B	19.13	21.91	$7.30 \text{ x}\cdot 10^{-3}$	$9.49 \text{ x}\cdot 10^{-3}$
C	7.35	9.01	$1.99 \text{ x}\cdot 10^{-3}$	$2.33 \text{ x}\cdot 10^{-3}$
D	1.84	2.52	$4.51 \text{ x}\cdot 10^{-4}$	$5.18 \text{ x}\cdot 10^{-4}$

Assuming that the maximum permissible ambient dose equivalent rate for occupational workers is 2.78 nSv/s [9], it can be noticed that with the bare source in air, calculated doses in sites A, B, and C are beyond to be safe for a continuous work; the safe distance, measured from the center of the ^{252}Cf neutron source would be at 200 cm (location D). With the source inside the WEP shielding, the worst scenario is at point B (in the lateral surface of the shielding) where the ambient dose equivalent rate is 7.30 $x \cdot 10^{-3}$ nSv/s. Under this assumption, this shielding could hold a ^{252}Cf up to 381 μg to allow full working time near the container.

With proper radiation safety rules the proximity of personnel can be easily limited to 100 cm from the center of WEP shielding; if here (point C) the maximum dose rate is set as 2.78 nSv/s, then the shield could host a ^{252}Cf neutron source as large as 1.397 mg, which can produce up to 3.35 $x \cdot 10^{9}$ neutrons per second. If the safety working distance would be limited to 200 cm (point D), then the source could be as large as 6.164 mg, thus producing 1.48 $x \cdot 10^{10}$ neutrons per second. This source strength would be suitable to perform thermal and prompt gamma-ray neutron activation analysis.

In Figure 5 the lethargy neutron fluence spectra of the ^{252}Cf source in vacuum, air, water and WEP, calculated at point C (100 cm from the source axis), are shown as they result from the MCNP calculation; the spectra in vacuum and air show similar features in the peak from 0.1 to 15 MeV; for energies less than 0.1 MeV the spectrum in air shows the presence of epithermal an thermal neutrons, this is probably due to the concrete ground layer that produces groundshine neutrons as well due to the skyshine neutrons in air.

In WEP and in water shielding, two peaks, in the thermal energies as well as around 5 MeV, can be noticed. The thermal peak and epithermal neutrons come from moderation effect produced by the hydrogen content in both shielding materials and the C content in WEP shielding. Both spectra have similar features, therefore they have alike moderating features, although in terms of dose, the WEP shielding shows a better performance.

In the table VI is shown the gamma ray fluence and Kerma in air due to gamma-rays produced during neutron transport in the WEP and water shielding per each neutron emitted by the ^{252}Cf. Comparing the kerma in air in each location can be noticed that the kerma in air in water shield is approximately 15%, 21%, 27% and 28% larger than WEP shielding in locations A, B, C and D respectively. Therefore, WEP shield is better to shield capture photons than water shielding.

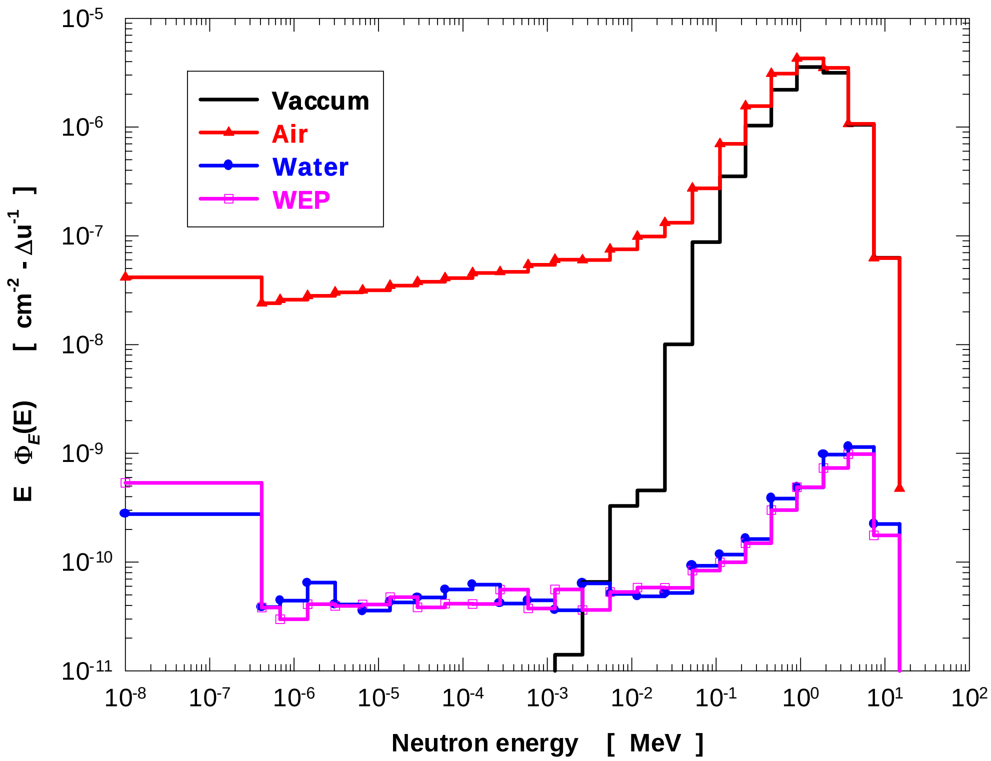

Figure 5. ^{252}Cf neutron spectra at site C.

Table VI. WEP and water gamma-ray features produced during neutron transport

Point	WEP $\Phi\gamma$ [cm^{-2}]	WEP K_{Air} [pSv]	Water $\Phi\gamma$ [cm^{-2}]	Water K_{Air} [pSv]
A	$5.1225 \times 10^{-6} \pm 4.13\%$	$1.9767 \times 10^{-5} \pm 4.21\%$	$6.4146 \times 10^{-6} \pm 3.52\%$	$2.4098 \times 10^{-5} \pm 3.16\%$
B	$7.5460 \times 10^{-6} \pm 1.22\%$	$2.8435 \times 10^{-5} \pm 1.43\%$	$8.7839 \times 10^{-6} \pm 1.18\%$	$3.3210 \times 10^{-5} \pm 1.22\%$
C	$2.6476 \times 10^{-6} \pm 0.21\%$	$1.0099 \times 10^{-5} \pm 0.20\%$	$3.1024 \times 10^{-6} \pm 0.24\%$	$1.1855 \times 10^{-5} \pm 0.19\%$
D	$6.6301 \times 10^{-7} \pm 0.23\%$	$2.4703 \times 10^{-6} \pm 0.15\%$	$7.8378 \times 10^{-7} \pm 0.21\%$	$2.9173 \times 10^{-6} \pm 0.19\%$

4. CONCLUSIONES

Neutrons emitted by ^{252}Cf suffer the in-scattering effect in air (skyshine) and in-scattering in concrete floor (groundshine), these effects tends to increase the number of neutrons, beyond the r^{-2} rule, at any location surrounding the shielding. The scattering tends to modify the neutron energy shifting the neutron spectrum from larger energies to epithermal and thermal energies.

For the studied shield configuration with WEP material, the largest ambient dose equivalent is observed at point B (at 62 cm from the source axial axis) near to the shielding vessel surface, which is the closest site to the neutron source. At this point, the WEP shield can reduce the dose by a factor of 3001 in comparison with the source in air, while a water-made shielding would reduce the dose by a factor of 2309. At point C (at 100 cm from the source axial axis), the ^{252}Cf source in air produces an ambient dose equivalent 4528 times larger that the source inside the WEP shielding, and 3867 times larger that the shield made with water. Consequently, the WEP material results in better shielding than water.

During transport in moderating materials neutrons loose energy reaching thermal energies where they can be captured and produce prompt gamma-rays. These photons also contribute to the dose at any point around the shielding. With the shield with water, at sites A, B, C and D the kerma in air is approximately 22, 17, 17 and 18% larger than the WEP shielding.

For neutrons produced by ^{252}Cf source WEP has a better performance to shield neutrons and capture gamma-rays.

Considering that neutrons are the unique shielding problem and assuming that the closest distance for working conditions is fixed at 200 cm, the WEP shielding can hold a ^{252}Cf source of up to 6.164 mg. Such a source would produce 1.48×10^{10} s^{-1}. If prompt gamma-rays are also considered, and no special shield is added to reduce them, the maximum ^{252}Cf mass is 2.12 mg that produces 5.09×10^9 s^{-1}.

Due to spectrum features of ^{252}Cf, the shielding can be modified to perform activities related with thermal neutron activation analysis, prompt gamma-ray neutron activation analysis, and to calibrate neutron-measuring instruments with different spectra.

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