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The comparison between simple and advanced shielding materials for the shield of portable neutron sources

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ABSTRACT

▶ ***Original article***

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Background: Monte Carlo simulations play a vital role in the calculation of the necessary shielding both for neutrons and photons. Advanced and simple shielding materials against neutron and gamma rays were compared by simulation using the MCNB4B Monte Carlo code. The simulations were carried out for the three common neutron sources, namely the ^{252}Cf, the ^{241}Am/Be and the DD neutron generator which are suitable for transportable facilities. ***Materials and Methods***: The source has been simulated as sphere with 3 cm diameter while the necessary shielding is designed in the form of a sphere around the neutron source. The materials considered were chosen according to the EU Directive 2002/95/EC, hence excluding lead and cadmium. ***Results:*** In the case of DD neutron generator the thickness, the weight and the volume of the shield can decrease up to 41.3, 44, and 78.4% correspondingly. With regard to the ^{252}Cf neutron source the use of advanced shielding materials can reduce the corresponding parameters up to 32.7, 40.7, and 68.4% respectively. As regards the ^{241}Am/Be neutron source, based on advanced shielding materials the thickness, the mass and the volume of the shield can decrease by 33.8, 49.5, and 70% respectively. ***Conclusion:*** The obtained results showed that the use of advanced shielding materials has led to reduce greatly the weight and the volume of the necessary shield.

Keywords: *Shielding materials, MCNP, RoHS directive.*

INTRODUCTION

Today neutron sources are used in a numerous applications. The Prompt Gamma Neutron Activation Analysis technique has been widely used in composition studies related to environmental and industrial applications (1-5). Neutron radiography is used for the non-destructive testing of objects in security applications, engineering studies and industry in order to determine structural defects, geology, medicine and biological research (6-10). Neutron Capture Therapy is a promising approach to cancer therapy for the cases where conventional radiation therapies fail (11-15). Neutron sources are also used in many explosives and land mine detection systems (16-20).

In these applications very often there is a need for compact and portable units (21-25). Neutron source (dimensions, spectrum and intensity) and the necessary shielding for radiation protection purposes, determines the possibility if a unit can be portable. Today nuclear reactors, accelerators and isotopic sources provide the necessary neutron beams. However, only isotopic neutron sources and portable neutron generators can be easily incorporated in transportable units.

Advanced neutron shielding materials are used today mainly for really special applications such as the shielding of compact nuclear reactions. According to the previous published work the hydrogen-rich hydrides demonstrate higher neutron shielding capability compared to the conventional materials. This means that the presence of advanced materials can reduce the thickness and the weight of the necessary shielding [26-27]. Advanced gamma ray shielding materials which based usually in Tungsten (or Tungsten alloy) are widely used in applications such as isotope containers, collimators, nuclear shielding, beam stops in medical, industrial radiation shielding and in nuclear engineering and research. These materials not only are more effective shielding materials but also can eradicate the environmental and toxic hazards which are related with the use of the lead [28-30].

The aim of this work is to evaluate many simple and advanced shielding materials in order to shield three different neutron sources, namely ^{252}Cf, ^{241}Am/Be and a Deuterium–Deuterium (DD) portable neutron generator. The simulations were carried out using the Monte Carlo code MCNP4B [31] in order to compare the performance of each material. The materials considered in the present study have been chosen according to article 4 of the RoHS Directive 2002/95/EC. Hence, lead, mercury, cadmium, hexavalent chromium, poly-brominated biphenyls (PBB) and poly-brominated diphenyl ethers (PBDE) have been excluded [32-33].

Isotopic neutron sources such as ^{241}Am/Be and ^{252}Cf have found application where maximal portability is required. A ^{252}Cf neutron source is considered, with an isotropic emission of 2.31×10^6 neutrons s^{-1} per µg of ^{252}Cf. The spectrum of the emitted neutrons extends up to 10 MeV with a mean energy at 2.3 MeV and modeled as a Watt fission spectrum using the coefficients provided by the MCNP 4B code. Additional to the neutron emission, ^{252}Cf emits 1.3×10^7 photons s^{-1} per µg with a mean energy of 0.8 MeV [34]. ^{241}Am/Be source has a long half-life period (432.7 yr) and is therefore used in many applications. The spectrum of the emitted neutrons expands up to 12 MeV with average neutron energy about of 4 MeV and was derived from Ref 37. ^{241}Am/Be is not only a common neutron source but also a gamma source. For gamma ray yield calculations, the energy spectrum of ^{241}Am/Be source, was taken from Ref. 38 [35-38]. DD neutron generator is based on D–D reaction producing neutrons with average energy 2.5 MeV. DD neutron generator emits only neutrons and offer an on/off switching of the emitted neutrons, for this reason would not require adequate shielding like isotopic neutron sources such as ^{241}Am/Be and ^{252}Cf.

It is known that the hydrogen-rich material has the potential to effectively shield neutrons because the contained hydrogen acts as a moderator of fast neutrons, reducing the fast neutron flux. High density polyethylene (HD-Poly) can be machined very easily and frequently used as a moderator in order to slow fast neutrons down to thermal energies [39]. The hydrogen concentration of HD-Poly is more than 7.8×10^{28} H-atoms/m^3. Borated polyethylene (Poly-B) is polyethylene with 5% boron and is widely used in neutron shielding applications because of its good nuclear and physical characteristics [40]. The boron and hydrogen concentrations of Poly-B are 2.6×10^{27} atoms/m^3 and 6.6×10^{28} atoms/m^3 respectively. 7.5% Lithium-Polyethylene (Poly-Li) is polyethylene with 7.5% Lithium and it is very effective in reducing gammas because it produces no capture gammas. The lithium and hydrogen concentrations of Poly-Li are 6.7×10^{27} atoms/m^3 and 5.44×10^{28} atoms/m^3 correspondingly.

Except from this, common shielding materials there are and advanced neutron shielding materials with higher hydrogen densities than those of conventional materials. Zirconium borohydride ($Zr(BH_4)_4$) is a candidate neutron shielding material [27]. The anticipated hydrogen concentration of ($Zr(BH_4)_4$) is 7.5×10^{28} H-atoms/m^3 while the boron concentration is 1.9×10^{28} B-atoms/m^3. Titanium hydride (TiH_2), has hydrogen concentration as high as 9.1×10^{28} H-atoms/m^3 surpassing this of HD-Poly. Finally, magnesium borohydride ($Mg(BH_4)_2$), is one of the most promising materials to store more hydrogen with the highest anticipated concentration equal to 1.32×10^{29} H-atoms/m^3 [26].

MATERIALS AND METHODS

Sources and materials

^{252}Cf and ^{241}Am/Be are also gamma sources and what's more there are gamma rays from the interaction of the neutrons with shielding materials, for example the 2.223 MeV gamma from the $^{1}H(n, \gamma)\ ^{2}H$ reaction; there is necessity not only for neutron shielding materials but also for gamma ray shielding materials. Materials containing lead were excluded from the design according to the EU Directive 2002/95/EC and lead was used only as indicator of the effectiveness of other gamma ray shielding materials.

Bismuth is a very good material for gamma-ray filtering. Compared to lead has nearly identical gamma ray attenuation and lower neutron-attenuation coefficient. The Stainless Steel is a good shielding material for gamma rays with logical cost. Tungsten is one of the best solutions as radiation shielding material because of its excellent radiation shielding effect realized by its high density with ability to withstand mechanical stress. However, pure tungsten is quite expensive and an extremely difficult material to machine. Tungsten Carbide is also used as radiation shielding material because of its high density originated from main component tungsten and its superior machinability to pure tungsten (41). Kennertium is machinable tungsten, which is much more amenable to fabrication but retains most of the characteristics of pure tungsten (42).

A range of simple and advanced materials were considered (tables 1 and 2), which would provide effective shielding while still rendering the unit transportable. The chemical composition of the shielding materials and the weight percentages of elements, which were examined, are provided in tables 1 and 2 for neutrons and gammas respectively.

The top view of the simulated facility is shown in figure 1. In all circumstances, the neutron source was simulated as sphere with 3 cm diameter which is symmetrically placed at the centre of the unit. In detail, the proposed facility comprises (the numbering refers to figure 1): of 1 or 2 or 3 spheres, from which the bigger incorporates always the smaller sphere.

In this study 200 μg of ^{252}Cf neutron source was considered. In the case of the ^{241}Am/Be neutron source the total activity was equal to 200 Ci which emits 4.4×10^{8} n s^{-1}. Based on previous job from Croft (35) and from Mowlavi and Koohi-Fayegh (36) gamma ray flux was estimated equal to 2.62×10^{8} gamma s^{-1}. The DD neutron generator is considered to provide a neutron yield 5×10^{8} n s^{-1}.

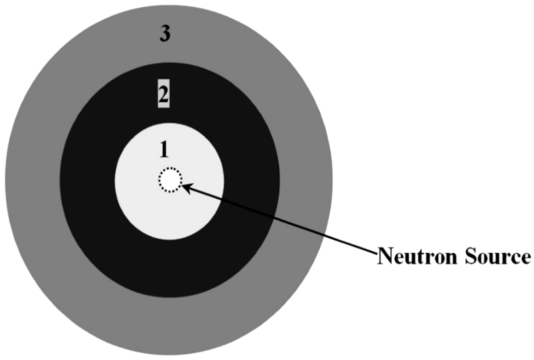

Figure 1. Side view of the geometric configuration of the irradiating system (not in scale).

Table 1. Neutron shielding compositions in mass fraction.

	HD-Poly	Poly-B	Poly-Li	$Zr(BH_4)_4$	TiH_2	$Mg(BH_4)_2$
H	0.143	0.116	0.076	0.107	0.0404	0.1492
O		0.222				
C	0.857	0.612	0.459			
B		0.05		0.3772		0.4006
Li			0.125			
Zr				0.6058		
Ti					0.9596	
Mg						0.4502
F			0.34			
Density (g/cm^3)	0.98	0.94	1.2	1.18	3.77	1.48

Table 2. Gamma ray shielding compositions in mass

	Lead	Bismuth	Steel	Tungsten	Tungsten Carbide	Kennertium
H						0.028
C						0.165
Pb	1					
Bi		1				0.807
W			1		0.76	
Ni					0.09	
Cu					0.15	
Density (g/cm^3)	11.35	9.8	8.92	19.3	15.6	16.8

RESULTS AND DISCUSSION

The shielding was designed as sphere incorporating the neutron source, comprise different materials with variable thicknesses bearing in mind the RoHS directive, the weight and dimensions which would render the unit transportable and the occupational dose limit of 25 μSv h^{-1} by ICRP-26 [43]. The total Dose Equivalent Rate (DER) for ^{252}Cf and ^{241}Am/Be comprises the dose due to the neutrons (DER1) and photons (DER2) from neutron source and the dose from the interaction of the neutrons and the shielding materials (DER3). In the case of DD neutron generator the DER contain only two components the DER1 and the DER3.

DD neutron generator

For DD neutron generator with a neutron yield 5 × 10^8 n s^{-1} figure 2 shows the minimum thickness for the six neutron shielding materials in order to the DER1 would remain below the annual occupational dose limit at the external surface of the shielding. Hd-Poly, Poly-B, and Poly-Li require 37, 44.05 and 43.3 cm respectively. Advanced materials show superior shielding capability than the conventional materials, as shown in figure 2, $Zr(BH_4)_4$, TiH_2,and $Mg(BH_4)_2$ require 41.1, 31.5 and 25.7 cm respectively. The mass, the volume and the DER3 from this thickness of each investigated materials are shown in table 3. Hd-Poly is the best solutions from simple materials and the Mg $(BH_4)_2$ from the advanced materials. The Mg $(BH_4)_2$ reduce the thickness and the weight of the shield by 30.5 and 46.6% compared to Hd-Poly, respectively, while simultaneously the DER3 is 3.8 times lower.

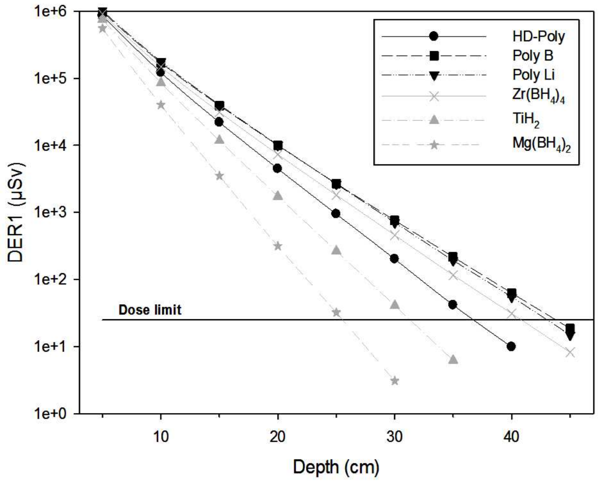

Figure 2. Comparison of DER1 for 6 neutron shielding materials for DD neutron generator.

The total DER estimates, for combinations of different layers of the materials shown in tables 1 and 2, are given in table 4. Using only conventional materials 46 cm of Poly-Li offer the lighter shield and has the same shielding thickness and volume with 38 cm Hd-Poly and 8 cm Bismuth. Based on advanced shielding materials 35 cm of $Mg(BH_4)_2$ reduce the total weight more than 44% and the combination of 26 cm of $Mg(BH_4)_2$ and 1 cm tungsten can decrease the total thickness and volume about 41.3 and 78.4%. It is obvious that the advanced shielding materials have higher shielding capabilities compared to the conventional materials.

Table 3. Volume, mass, DER3, and the minimum thickness for 6 neutron shielding materials in order DER1 does not overcome the 25 μSv h^{-1} (DD neutron generator).

	HD-Poly	Poly-B	Poly-Li	$Zr(BH_4)_4$	TiH_2	$Mg(BH_4)_2$
Depth (cm) for DER1=25μSv	37.00	44.05	43.30	41.10	31.50	25.70
Kg	234	372	452	382	567	125
V(m^3)	0.239	0.396	0.376	0.324	0.15	0.084
DER3 (μSv)	384.2	34.8	10.8	13.3	261.8	101.3

Table 4. Estimates of the dose rate, weight and volume for different shielding configurations using simple and advanced shielding materials (DD neutron generator).

	Shielding materials thickness (cm)					Weight (kg)	Volume (m^3)	Dose rate (μSv h^{-1})		
	HD Poly	Poly-Li	$Mg(BH_4)_2$	Bismuth	Tungsten			DER1	DER2	Total
Layer1		46				538	0.449	12.76	8.44	21.20
Layer1	38					2121	0.449	10.90	10.14	21.04
Layer2				8						
Layer1			35			301	0.204	0.27	23.83	24.10
Layer1			26			319	0.097	17.97	6.94	24.91
Layer2					1					

^{252}Cf neutron source

Figures 3 and 4 show the calculated DER1 and DER2 for 200 μg ^{252}Cf neutron source. Calculations with the MCNP4B code have shown that thickness of at least 46, 53.4, and 51 cm are required for Hd-Poly, Poly-B, and Poly-Li respectively in order to keep DER1 within the recommended limit. The corresponding thickness for $Zr(BH_4)_4$, TiH_2, and $Mg(BH_4)_2$ was 51.2, 36.6, and 33.1 cm respectively. Table 5 lists the results of the mass, the volume, the DER2, and the DER3 from the six neutron shielding materials. According to the results from the table 5, the advanced materials show superior shielding capability than typical neutron shielding materials.

In terms of gamma rays generated by the ^{252}Cf source pure tungsten and tungsten alloys seem as better solution as replacement for lead, which has been usually used for this application. Simulation with MCNP4B Monte Carlo code (figure 5) have shown that thickness of at least 13.65, 14.95, and 20.75 cm are required for lead, bismuth and steal correspondingly with intention the DER2 does not overcome the 25 μSv h^{-1}. 9.45 cm from pure tungsten or 10.55 cm from Kennertium or 11.2 cm from tungsten carbide are enough to prevent the dose rate from exceed over the limit. Table 6 shows the mass, the volume and the DER1 when the DER2 are equal to 25 μSv h^{-1} for each of the six candidate materials as gamma shielding material.

The total DER for different combinations between of simple neutron and gamma ray shielding materials and between the advanced neutron and gamma ray shielding materials are shown in table 7. Choosing only usual shielding materials, two layers of bismuth with 2 cm thickness, with 56 cm layer of Poly-Li between of them is the combination that ensures the minimum weight of the shield. The smallest volume and thickness of the shielding realized with 5 cm of bismuth, 49 cm HD-Poly and 4 cm bismuth (from inward to outward). Using advanced shielding materials a 35 cm layer of $Mg(BH_4)_2$ sandwiched between of the two layers of the tungsten with 2 cm thickness can reduce the thickness of the shield by 32.7%, the mass and the volume by 40.7 and 68.4% respectively compared to the common shielding materials.

^{241}Am/Be neutron source

In order to keep the DER1 and DER2 within the recommended limit based on a 200 Ci ^{241}Am/Be neutron source, the necessary shielding surrounding the source, requires different thickness accordingly to the selected material. Figure 5 illustrates the effectiveness of each neutron shielding material and figure 6 shows the efficiency of each gamma ray shielding materials. Hd-Poly, Poly-B, Poly-Li, Zr $(BH_4)_4$, TiH_2,and $Mg(BH_4)_2$ require at least 55.5, 64.4, 60.6, 61.25, 43.25, and 39.35 cm thickness

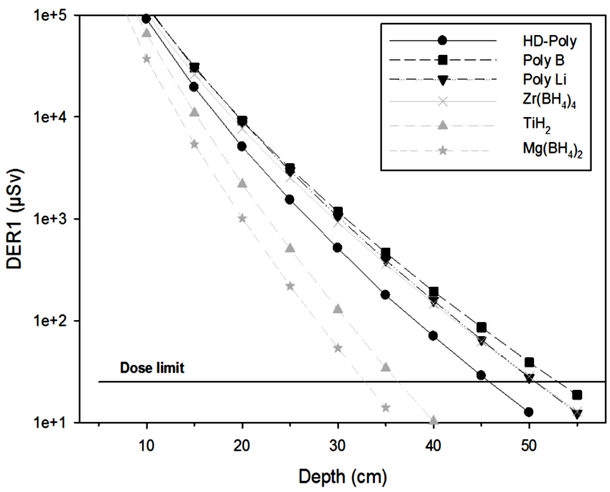

Figure 3. Comparison of DER1 for 6 neutron shielding materials for ^{252}Cf neutron source.

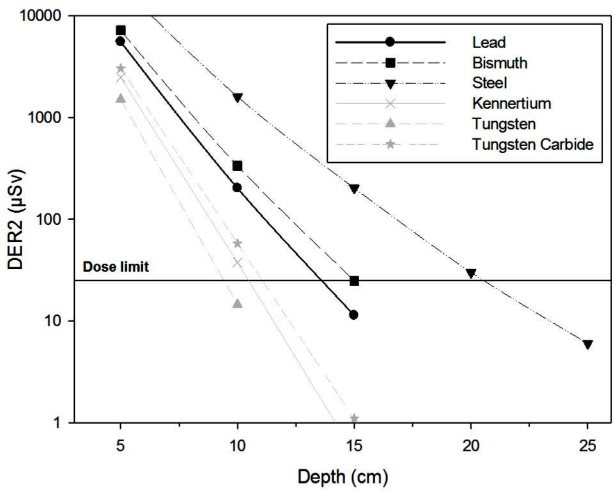

Figure 4. Comparison of DER2 for 6 gamma ray shielding materials for ^{252}Cf neutron source.

for DER1 below of the annual occupational dose limit. Table 8 illustrates the mass, the volume, DER2 and the DER3 when the DER1 are equal to 25 μSv h-1 for each of the six neutron shielding material. In terms of gamma ray shielding materials the minimum thickness of lead, bismuth, steal, Kennertium, pure tungsten, and tungsten carbide for DER2 below than 25 μSv h-1 are 10.05, 11, 15.05, 8.05, 7.2, and 8.45 cm correspondingly (figure 6). The comparison of the six gamma ray shielding materials is listed on table 9.

Table 10 shows the best results using simple and advanced shielding materials. Again, the supremacy of the advanced material is unquestioned fact. 1 cm pure tungsten combined with 48 cm $Mg(BH_4)_2$ can reduce the weight of the shield more than 49.5% compared to 70 cm Poly-B. Simultaneously 41 cm $Mg(BH_4)_2$ sandwiched between of the two layers of the tungsten with 1 cm thickness can decrease the thickness and the volume of the shield by 33.8 and 70% respectively.

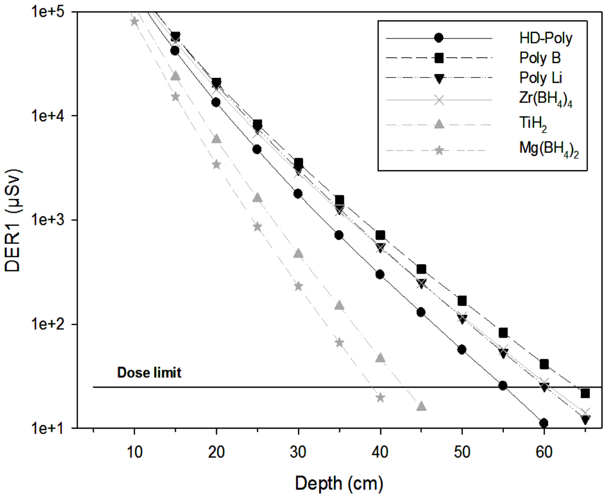

Figure 5. Comparison of DER1 for 6 neutron shielding materials for ^{241}Am/Be neutron source.

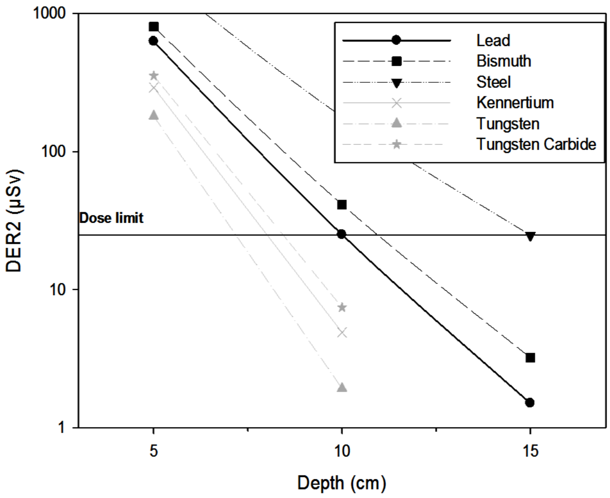

Figure 6. Comparison of DER2 for 6 gamma ray shielding materials for ^{241}Am/Be neutron source.

Table 5. Volume, mass, DER2, DER3 and the minimum thickness for 6 neutron shielding materials in order DER1 does not overcome the 25 μSv h^{-1} (^{252}Cf neutron source).

	HD-Poly	Poly-B	Poly-Li	$Zr(BH_4)_4$	TiH_2	$Mg(BH_4)_2$
Depth (cm) for DER1=25μSv	46.0	53.4	51.0	51.2	36.6	33.1
Kg	440	651	727	735	873	257
V(m^3)	0.449	0.693	0.606	0.613	0.231	0.173
DER2 (μSv)	296.3	184.1	146.5	118.6	19.5	450.6
DER3 (μSv)	172.2	14.6	6.3	4.1	107.9	32.6
DER2+DER3 (μSv)	468.5	198.7	152.8	122.7	127.4	483.2

Table 6. Volume, mass, DER1, and the minimum thickness for 6 gamma ray shielding materials in order DER2 does not overcome the 25 μSv h^{-1} (^{252}Cf neutron source).

	Lead	Bismuth	Steel	Kennertium	Tungsten	Tungsten Carbide
Depth(cm) for DER2=25μSv	13.65	14.95	20.75	10.55	9.45	11.2
Kg	165	182	360	123	106	134
V(m^3)	0.015	0.019	0.046	0.007	0.005	0.009
DER1 (μSv)	2.35E+5	2.00E+5	7.14E+4	2.14E+5	2.57E+5	1.41E+5

Table 7. Estimates of the dose rate, weight and volume for different shielding configurations using simple and advanced shielding materials (^{252}Cf neutron source).

	Shielding materials thickness (cm)					Weight (kg)	Volume (m^3)	Dose rate ($\mu Sv\ h^{-1}$)			
	HD Poly	Poly-Li	$Mg(BH_4)_2$	Bismuth	Tungsten			DER1	DER2	DER3	Total
Layer1				2							
Layer2		56				1878	0.927	9.18	13.63	1.68	24.49
Layer3				2							
Layer1				5							
Layer2	49					2339	0.882	5.35	3.25	15.58	24.18
Layer3				4							
Layer1					2						
Layer2			35			1113	0.278	6.88	12.37	3.15	22.40
Layer3					2						

Table 8. Volume, mass, DER2, DER3 and the minimum thickness for 6 neutron shielding materials in order DER1 does not overcome the 25 $\mu Sv\ h^{-1}$ (^{241}Am/Be neutron source).

	HD-Poly	Poly-B	Poly-Li	$Zr(BH_4)_4$	TiH_2	$Mg(BH_4)_2$
Depth (cm) for DER1=25μSv	55.5	64.40	60.60	61.25	43.25	39.35
Kg	760	1126	1183	1220	1414	422
V(m^3)	0.775	1.198	1.002	1.034	0.375	0.285
DER2 (μSv)	1.5	0.9	0.8	0.6	0.1	2.5
DER3 (μSv)	91.8	9.8	7.0	3.1	46.4	24.7
DER2+DER3 (μSv)	93.3	10.7	7.8	3.7	46.5	27.2

Table 9. Volume, mass, DER1, and the minimum thickness for 6 gamma ray shielding materials in order DER2 does not overcome the 25 $\mu Sv\ h^{-1}$ (^{241}Am/Be neutron source).

	Lead	Bismuth	Steel	Kennertium	Tungsten	Tungsten Carbide
Depth (cm) for DER2=25μSv	10.05	11.00	15.05	8.05	7.20	8.45
Kg	73	80	150	61	53	64
V(m^3)	0.006	0.008	0.019	0.004	0.003	0.004
DER1 (μSv)	4.00E+5	3.40E+5	1.50E+5	4.09E+5	4.95E+5	3.33E+5

Table 10. Estimates of the dose rate, weight and volume for different shielding configurations using simple and advanced shielding materials (^{241}Am/Be neutron source).

	Shielding materials thickness (cm)				Weight (kg)	Volume (m^3)	Dose rate ($\mu Sv\ h^{-1}$)			
	Poly-B	Poly-Li	$Mg(BH_4)_2$	Tungsten			DER1	DER2	DER3	Total
Layer1	70				1438	1.53	11.9	6.46	6.61	24.97
Layer1		65			1477	1.231	13.13	5.63	5.18	23.94
Layer1				1	725	0.508	3.64	5.50	13.37	22.51
Layer2			48							
Layer1				1						
Layer2			41		980	0.369	11.35	8.08	3.97	23.41
Layer3				1						

CONCLUSION

Three neutron sources ^{252}Cf, ^{241}Am/Be, and a compact DD neutron generator has been simulated, for dosimetric purposes, using the MCNP4B Monte Carlo code. The materials considered were compatible with the European Union Directive on 'Restriction of Hazardous Substances' (RoHS) 2002/95/EC, hence excluding the use of cadmium and lead. Hd-Poly, Poly-B, Poly-Li compared with advanced candidate neutron shielding materials such as the $Zr(BH_4)_4$, the TiH_2, and the $Mg(BH_4)_2$. In all circumstances the $Mg(BH_4)_2$ show superior neutron shielding capabilities.

Three simple gamma radiation shielding materials namely bismuth, steal and lead compared with pure tungsten, tungsten carbide and Kennertium. Pure tungsten primary and the other tungsten alloys secondary show excellent gamma ray shielding capabilities. If the cost of advanced materials is not obstacle, these materials can reduce tremendously the weight of the shield. In case of DD neutron generator the mass of the shield can reduce more than 44% using advanced shielding materials. The corresponding decrements on the weight for ^{252}Cf and ^{241}Am/Be were 40.7 and 49.5%.

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Conflicts of interest: *none to declare.*

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