AMERICIUM-BERYLLIUM SHIELD CONTAINER OPTIMIZATION WITH NEW HIGH ALLOYED STAINLESS STEEL AND COMPOSITE MATERIAL USING MCNPX CODE

by

Ali HOOSHMAND FINI, Mohsen KHERADMAND SAADI*, and Mitra ATHARI ALLAF

Department of Nuclear Engineering, Science and Research Branch, Islamic Azad University, Tehran, Iran

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In this paper the goal is to optimize a shield for the americium-beryllium radioactive source, which generates a mixed neutron and gamma-ray radiation field, using two different materials designed with simultaneous moderation and absorption of neutrons and gamma-ray shielding in mind: a new type of stainless steel and epoxy resin ($C_{21}H_{25}ClO_5$) composite with a 40 % NiO additive. For this purpose, MCNPX was used and the equivalent dose rate for both types of radiation was calculated. After the optimization process, the shield volume experienced a 54 % reduction while its weight, due to the use of a steel alloy, was increased by 3.56 %. At a distance of 1 m from the center of the source, the equivalent dose rate was reduced by 10.98 % in comparison to that of the original system.

Key words: americium-beryllium source, shield optimization, high alloyed stainless steel, epoxy resin, MCNPX

INTRODUCTION

Nuclear reactors are considered the most prolific and well-known neutron sources, however, their most important and obvious drawbacks are high cost, safety, and non-transportability. Isotopic neutron sources, on the other hand, because of their low cost, flexibility, and portability can be a suitable alternative [1]. Americium-beryllium (Am-Be) is one of the most widely used isotopic neutron sources with different applications in science and industry such as neutron imaging, irradiation of samples, and neutron calibration of detectors and monitoring instruments [2].

Neutrons released by this source have a spectrum with a mean energy of about 4.5 MeV and maximum energy of about 11 MeV [3]. The neutron spectrum of this source is shown in fig. 1. The emitted neutrons are then followed by 4.8 MeV gamma-rays. These photons are responsible for 37.5 % of the total activity of the source [1]. Considering their high penetration power and lack of electric charge, necessary protective measures and shielding must be provided to ensure the safety of personnel from the harmful effects of the radiation fields generated by these sources. Shielding against neutrons is more complex than other types of radiation since it involves attenuation of not only primary neutrons but also the production and attenuation of secondary particles. These associated problems include the production of photons from neutron inelastic scattering, neutron slowing down and capture of thermal neutrons which is leading to the capture of gamma rays [4]. Shielding against neutron sources is accomplished through three logical steps [5]:

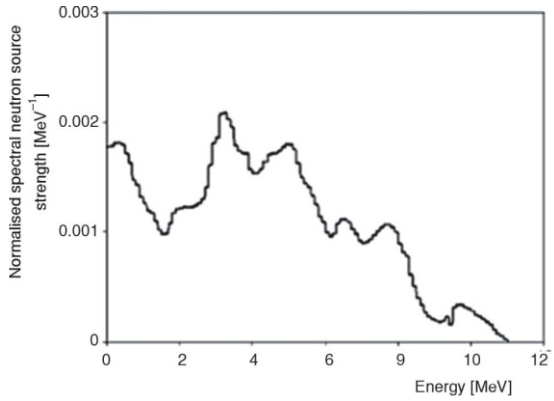


Figure 1. Am-Be neutron spectrum [1]

(1) Neutrons must be artificially produced, and when initially formed, regardless of their origins, they are all categorized as fast neutrons. Fast neutrons have a very low absorption cross-section, but their scattering cross-section is very high in many materials, especially in materials with low Z such as hydrogen as they may lose a large fraction of their energy in individual interactions with them. The

* Corresponding author, e-mail: mohsen.kheradmand@gmail.com

first step in shielding against neutrons is to place a hydrogen-rich material near the source to force the neutrons to slow down to thermal energies by inducing scattering reactions.

(2) Thermal neutrons have a much higher absorption cross-section, particularly in materials such as boron and its compounds. So the second step in neutron shielding is the absorption of the moderated neutrons by materials with a high neutron absorption cross-section.

(3) After their moderation and capture, gamma rays are emitted, so the third step is using a certain amount of suitable material for shielding against gamma photons.

The drawback of this approach is that it can often lead to an increase in the volume and weight of the shield to the point that in some cases, because of its weight and volume, the source is considered a fixed one rather than a portable one. Plus, this approach complicates the design since, for each step of the process, a different suitable material must be chosen and provided in sufficient quantities.

Recent years have seen a growth in the development and application of materials with simultaneous shielding capabilities in both neutron moderation and absorption and gamma-ray shielding. Polymers, thanks to their flexible nature, are of particular interest in this field since some of their properties can be enriched with the use of the right additives. The right choice of polymer and additive can result in a composite material suitable for both neutron and gamma attenuation. For example, the shielding performance of epoxy resin with a 40 % tantalum additive was tested against Am-Be 4.5 MeV neutrons and 1.25 MeV ^{60}Co photons. The reported neutron effective removal cross-section and gamma linear attenuation coefficient, 0.677 cm^{-1} and 0.43 cm^{-1} respectively, of this composite, were rather high in comparison to other similar materials [6]. As an example, the neutron effective removal cross-section reported for epoxy resin with 5 % of B_4C was 0.345 cm^{-1} against 4.5 MeV neutrons [7], and the reported gamma linear attenuation coefficient for tungsten powder filled epoxy resin was 0.27 cm^{-1} against ^{60}Co photons [8]. Advances in the science of metalworking and alloy development have resulted in the development of steel alloys that can act as both neutron and gamma shields with properties superior to that of 316LN stainless steel, an alloy with widespread use in nuclear applications. In fact, 316LN has low radiation absorption abilities against both fast neutrons and gamma radiations [9].

Some research and studies have already shown the potential of polymer compounds in gamma-ray shielding. Chaitali *et al*., mixed epoxy resin polymer specimens with four different metal chlorides ($BiCl_3$, $CdCl_2$, CsCl and $PbCl_2$) at three different weight fractions and demonstrated improved shielding properties against gamma-ray photons ranging from 0.05 ev up to 3 MeV [10]. Abbas *et al*., loaded PVC samples with micro and nano PbO/CuO particles at different weight fractions between 10 % up to 40 % [11]. The mass and linear attenuation coefficients were measured as a function of gamma-ray energies ranging from 59.53 keV to 1408.01 keV employing different sources. They concluded, to minimize the use of lead as an absorber one can replace the pure lead blocks with composites loaded with a mixture of CuO and PbO nano particles [11]. In another research, two batches of concrete sample shields filled by synthesized PbO and PbO-H_3BO_3 nano particles were developed for gamma and mixed neutron-gamma fields, respectively [12, 13]. Results showed that for the ordinary concrete reinforced with 5 wt. % nano PbO particles, the HVL parameter was reduced by 64 % at 511 keV and 48 % at 1332 keV [12]. For both batches, the employment of a low concentration of fillers up to 5 wt. % was recommended [12] [13].

The main objective of current research is the improvement of an Am-Be beryllium shield using epoxy resin with 40 % NiO and a new type of high alloyed stainless steel. Both materials were developed with simultaneous shielding against neutrons and gamma-ray photons in mind and both showed exceptional performance in comparison to similar materials used for radiation shielding. The main objectives are reduction in volume, equivalent dose rate, and simplification of the shield structural configuration. Model validation and dose calculations were accomplished using the MCNPX Monte Carlo code.

MATERIALS AND METHODS

Reference system

The reference system was taken from the work of Moadab and Kheradmand Saadi [14] and consists of a cylinder with a 23 cm height and 21.75 cm radius. At the center of the shield is an Am-Be beryllium neutron-gamma source with a 3 cm height, 2.5 cm diameter, total activity of 5 Ci, and neutron and gamma intensities of 1.23 10^7 and 7.31 10^6 particles per second, respectively. A hollow incomplete cylinder of polyethylene (C_2H_4) with a thickness of 5.5 cm surrounds the source and acts as its primary moderator. This cylinder is completed by two truncated graphite cones in the upper and lower regions which act as neutron reflectors and have a height of 5 cm and a radius of 6.75 cm. The secondary moderator which surrounds the first one is another incomplete cylinder with a thickness of 10 cm and consists of C_2H_4-5%Bi. C_2H_4-75 %Bi with a thickness of 1.5 cm and Mg(BH4)2 with a thickness of 2 cm act as a gamma-ray shield and neutron absorber respectively and are repeated in radial and axial directions [14]. Figure 2 shows the layer configuration of the shield and tab. 1 shows the composition of each layer.

Involved materials

The polymer used for this work is epoxy resin +40 % NiO. This composite was prepared by mixing

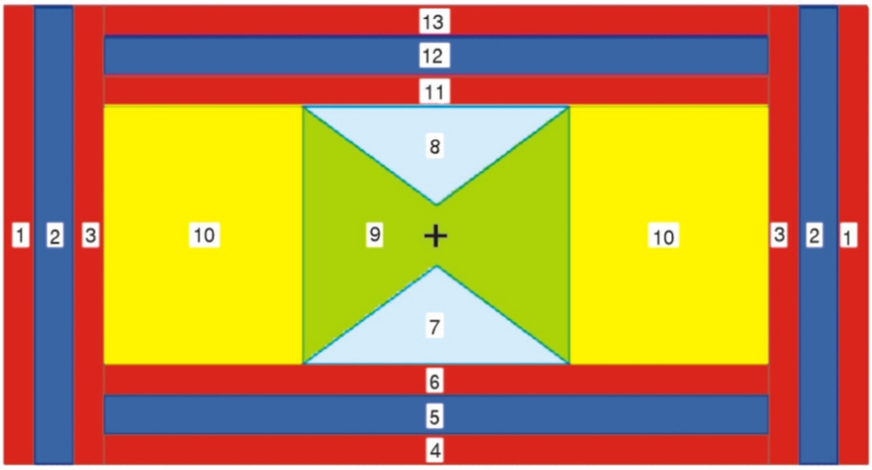


Figure 2. Layer configuration of the shield

Table 1. Layer composition of the shield

Layer number	Material
1	C_2H_4-75 %Bi
2	$Mg(BH_4)_2$
3	C_2H_4-75 %Bi
4	C_2H_4-75 %Bi
5	$Mg(BH_4)_2$
6	C_2H_4-75 %Bi
7	Graphite
8	Graphite
9	C_2H_4
10	C_2H_4-5 %Bi
11	C_2H_4-75 %Bi
12	$Mg(BH_4)_2$
13	C_2H_4-75 %Bi

Table 2. Epoxy resin-40 %NiO chemical composition

Element	wt %
C	38.5
H	3.8
Cl	5.5
O	20.8
Ni	31.4

the additive, hardener ($C_9H_{10}O_3$), and commercially available nickel oxide powder at a constant speed for 10 minutes at 500 rounds per minute until they were homogeneous. The resulting mixture was then poured into a cylindrical silicone rubber mold and dried at room temperature. The sample was then coated with a soudal calofer sodium silicate sealant, a sodium silicate-based high-quality asbestos-free sealant used in high-temperature environments up to 1500 °C to increase its temperature resistance against high temperature. This sample has a density of 3.40 gcm^{-3} and its chemical composition is shown in tab. 2 [15]. This composite will replace C_2H_4, C_2H_4-5 % Bi, and $Mg(BH_4)_2$ during the optimization process.

High alloyed stainless steel was prepared using a nano-sized powder of the elements shown in tab. 3 and

Table 3. Chemical composition ratios of high alloyed stainless steel [11]

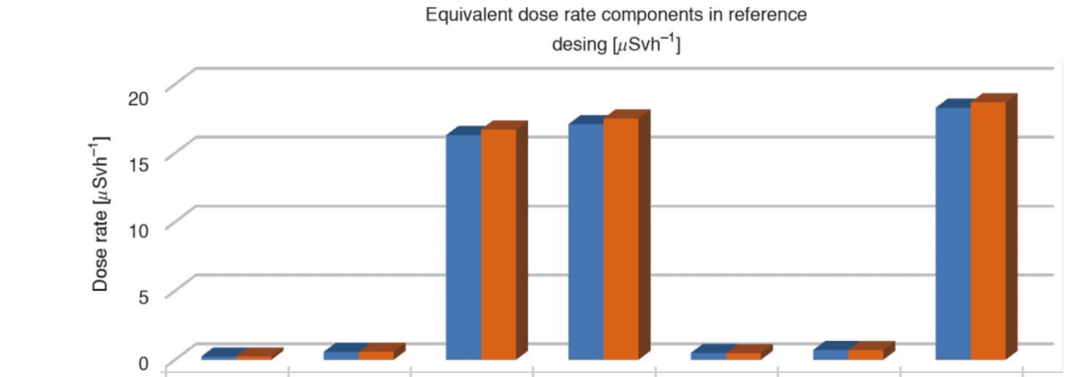
Element	wt %
Ni	30
Cr	15
Mn	1
C	0.5
Mo	1
W	20
Fe	31.47
V	0.015
S	0.015
Re	1

the powder-metallurgy method. Materials were mixed for 15 minutes. When it became homogeneous, the mixture was heated at 350 °C and then pressurized at 600 MPa pressure. The formed sample was then annealed at 1300 °C for 3 hours, then hardened by a faster cooling process. This alloy has a density of 10.45 gcm^{-3} [11].

The three main components of this alloy are Ni, Cr, and W. Based on the percentages of these elements, this alloy is closer to the steels of the austenitic class. The high content of these elements has given this steel increased heat resistance and radiation shielding properties. Stainless steels tend to crack after welding but this has not been observed for this new alloy [11].

Simulation

In order to calculate the total equivalent dose rate, the MCNPX Monte Carlo code and its surface flux tally, F2, were used. Since Am-Be beryllium generates a mixed field of radiation, two simulations must be conducted: in the first simulation, the source was considered a pure neutron source. Neutron particles were tracked for three different energy ranges: thermal neutron (0.5 eV and below), fast neutrons (0.5 MeV and above), and epithermal neutrons between these two ranges alongside resultant gamma-ray photons from



	Thermal	Epi thermal	Fast	Total	(n, γ)	γ-source	Total
Reference	0.22721	0.5637	16.2957	17.0867	0.4865	0.7096	18.2828
Simulation	0.230523	0.571671	16.7013	17.5035	4.72E-01	0.701243	18.676914

Figure 3. Comparison between data available from the reference shield and simulation results

neutron interactions with different layers of the shield. In the second simulation, the source was considered a pure gamma source, and the photons emitted from it were tracked. To convert the calculated flux to dose, ANSI-6.1.1-1991 flux-to-dose conversion factors were used [16]. To reduce the statistical errors, both simulations were run for 10 million histories. Total dose is calculated by summing up these three values: total neutron dose, gamma dose from neutron interactions with different layers of the shield, and gamma dose emitted directly from the source itself.

RESULTS AND DISCUSSIONS

Model validation

The MCNPX input file was validated by first simulating the reference shield from the work of Moadab and Kheradmand Saadi [14] and then comparing the results with available data from the reference system. The results are shown in fig. 3.

The results are in good agreement with the available data, however, the small difference in the values may be due to discrepancies in the neutron energy spectrum as well as MCNPX data libraries. From these results, one can conclude that since the highest share of the total dose belongs to fast neutrons, the focus of the optimization process must be on better moderation because this fact points toward weakness in the neutron moderation of the shield.

Optimization process

Previous simulation runs showed that by preserving the dimensions, the best structural configuration had been achieved by replacing the primary and secondary moderators from the reference shield with a single incomplete cylinder of epoxy resine+40 % NiO, gamma absorber with high alloyed stainless steel, and the neutron absorber with epoxy resine+40 % NiO again. Table 4 shows the calculated dose rates for this configuration and its relative difference in comparison to the reference shield. All dose units are in Svh^{-1}.

Figure 4 shows the structure of this new shield with the dimensions of the reference. Layers 2, 5, 9, and 11 are made of epoxy resine+40 % NiO, and layers 1, 3, 4, 6, 10, and 12 are made of high alloyed stainless steel described in this paper. To reduce the volume of the shield, only the thickness of layer 9 could be reduced. Any further reduction in the thickness of the outer layers of the shield would compromise its ability to reduce the radiation levels. By reducing the thickness of layer 9 by 7 cm, the total equivalent dose value for this configuration was increased from 11.48 Svh^{-1} to 16.27 Svh^{-1}, 18.65 % less than the recommended value and 10.98 % less than the reference system. The shield volume experienced a 54 % reduction and went from 34164.5 cm^3 to 15720.33 cm^3, while its weight, due to the use of a steel alloy instead of a polymer-based material, was increased by 3.59 % and went from 113.8461 kg to 117.9009 kg. The shield total radius decreased from 21.75 cm to 14.75 cm, but its

Table 4. Dose rate comparison between the reference shield and the new structural configuration

Radiation type	Reference shield	New configuration	Relative difference [%]
Thermal neutrons	0.22721	0.0100497	95.57
Epithermal neutrons	0.5637	0.481299	14.61
Fast neutrons	16.2975	9.93903	39.008
Total neutron dose	17.0867	10.4304	38.95
(n, γ)	0.4865	0.738161	51.72
gamma-ray	0.7096	0.312182	56.005
Total dose	18.2828	11.480743	37.20

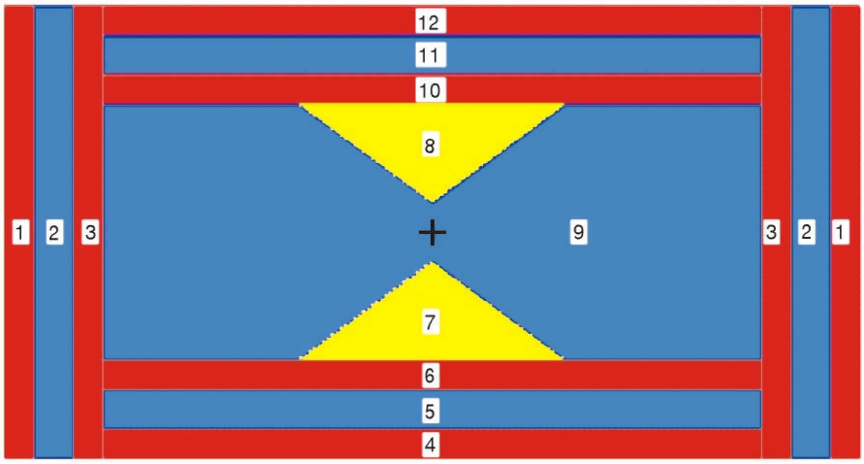
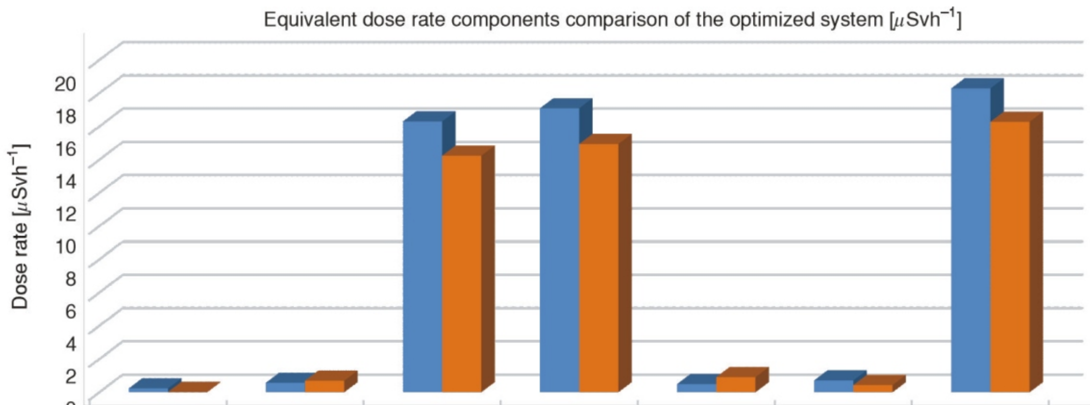


Figure 4. New structural configuration for the optimization process



	Thermal	Epi thermal	Fast	Total	(n, γ)	γ-source	Total
Reference	0.22721	0.5637	16.2957	17.0867	0.4865	0.7096	18.2828
Simulation	0.0143304	0.692327	14.2321	14.9388	9.07E-01	0.428517	16.274401

Figure 5. Comparison between equivalent dose rates of the reference system and optimized system

height remained at 23 cm. Figure 5 compares the equivalent dose rate values of the reference system with the optimized one.

CONCLUSION

In this paper, a new high alloyed stainless steel and epoxy resin polymer composite was used to optimize an Am-Be beryllium shield container. Both materials were developed with the goal of simultaneous shielding against fast neutrons and gamma-rays. Primary, secondary, and neutron absorbers of the original system were replaced by epoxy resin+40 % NiO while the gamma-ray absorber was replaced by high alloyed steel. The results showed that by using these materials, and after reducing the volume, the total equivalent dose rate and volume were reduced by 10.98 and 54 % respectively. However, due to the use of a steel alloy instead of a polymer, shield weight was increased by 3.59 %. The system provides better protection and is more mobile in this configuration.

AUTHORS' CONTRIBIUTIONS

A. Hooshmand Fini: methodology, software, investigation, resources writing – original draft; M. Kheradmand Saadi: conceptualization, methodology, software, investigation, writing – review and editing; M. Athari Allaf: methodology, investigation, resources.

REFERENCES

[1] Nasrabadi, M. N., *et al*., Neutron Shielding Design for ^{241}Am-Be Neutron Source Considering Different Sites to Achieve Maximum Thermal and Fast Neutron Flux Using MCNPX Code, *Annals of Nuclear Energy, 59* (2013), Sept., pp. 47-52

[2] Basu, P., *et al*., Compact Shielding Design for 740 GBq ^{241}Am-Be Neutron Source Transport Container, *Radiation Physics and Chemistry, 170* (2020), Dec., DOI: 10.1016/j.radphyschem.2019.018670

[3] Zhang, F., *et al*., Compact Shielding Design of a Portable ^{241}Am-Be Source, *Applied Radiation and Isotopes, 128* (2017), pp. 49-54

[4] ***, Handbook of Nuclear Engineering Volume II Reactor Design, *Springer* (2010). p. 138

[5] ***, Radaition Protection and Dosimetry: An Introduction to health Physics, Springer-Verlag New York, (2007), pp. 261
[6] Canel, A., *et al*., Improving Neutron and Gamma Flexible Shielding by Adding Medium-Heavy Metal Powder to Epoxy Based Composite Materials, *Radiation Physics and Chemistry, 158* (2019), May, pp. 13-16
[7] Ruhollah Adeli., *et al*., Neutron Irradiation Tests on B4C/Epoxy Composite for Neutron Shielding Application and the Parameters Assay, *Radiation Physics and Chemistry, 127* (2016), June, pp. 140-146
[8] Le Chang., *et al*., Preparation and Characterization of tungsten/Epoxy Composites for γ-Rays Radiation Shielding, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 356-357* (2015), 12, pp. 88-93
[9] Aygun, B., High Alloyed New Stainless Steel Shielding Material for Gamma and Fast Neutron Radiation, *Nuclear Engineering and Technology, 52* (2020), 3, pp. 647-653
[10] Chaitali, V. More., *et al*., Extensive Theoretical Study of Gamma Ray Shielding Parameters Using Epoxy Resin – Metal Chloride Chloride Mixtures, *Nuclear Technology and Radiation Protection, 35* (2020), 2, pp. 138-149
[11] Abbas, Y. M., *et al*., Gamma Attenuation Through Nano Nano Lead Lead – Nano Nano Copper Copper PVC Composites, *Nucl Technol Radiat, 36* (2021), 1, pp. 50-59
[12] Mokhtari, K., *et al*., Fabrication, Characterization, Simulation and Experimental Studies of the Ordinary Concrete Reinforced with Micro and Nano Lead Oxide Particles Against Gamma Radiation, *Nuclear Engineering and Technology, 53* (2021), 9, pp. 3051-3057
[13] Mokhtari, K., *et al*., The shielding Properties of the Ordinary Concrete Reinforced with Innovative Nano Polymer Particles Containing PbO-H_3BO_3 for Dual Protection Against Gamma and Neutron Radiations, *Radiation Physics and Chemistery, 189* (2021), Dec., 109731
[14] Moadab, N. H., Saadi, M. K., Optimization of an Am-Be Neutron Source Shield Design by Advanced Materials Using MCNP Code, *Radiation Physics and Chemistry, 158* (2019), May, pp. 109-114
[15] Aygün, B., *et al*., Experimental and Monte Carlo Simulation Study on Potential New Composite Materials to Moderate Neutron-Gamma Radiation, *Progress in Nuclear Energy, 130* (2020), Dec., 103538
[16] ***, ANSI/ANS-6.1.1-1991, Neutron and Gamma-Ray Fluence-to-Dose Factors

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Али ХУШМАНД ФИНИ, Мохсен КЕРАДМАНД САДИ, Митра АТАРИ АЛАФ

ОПТИМИЗАЦИЈА ШТИТА АМЕРИЦИЈУМ–БЕРИЛИЈУМСКОГ КОНТЕЈНЕРА ОД НОВОГ ВИСОКОЛЕГИРАНОГ НЕРЂАЈУЋЕГ ЧЕЛИКА И КОМПОЗИТНИХ МАТЕРИЈАЛА, КОРИШЋЕЊЕМ MCNPX ПРОГРАМА

Намера је да се оптимизује штит америцијум-берилијумског радиоактивног извора, који ствара мешовито поље неутрона и гама зрачења, користећи два различита материјала – нове врсте композита од нерђајућег челика и епоксидне смоле ($C_{21}H_{25}ClO_5$) са 40 % NiO адитива. У ту сврху коришћен је MCNPX програм и израчуната је еквивалентна јачина дозе за обе врсте зрачења. Након процеса оптимизације, запремина штита смањена је за 54 %, док је његова тежина, због употребе челичне легуре, повећана за 3.56 %. На удаљености од једног метра од центра извора, јачина еквивалентне дозе смањена је за 10.98 % у односу на оригинални систем.

Кључне речи: америцијум-берилијумски извор, оптимизација штита, високолегирани нерђајући челик, епоксидна смола, MCNPX програм