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Original Article

Development of gradient composite shielding material for shielding neutrons and gamma rays

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ABSTRACT

In this study, a gradient material for shielding neutrons and gamma rays was developed, which consists of epoxy resin, boron carbide (B_4C), lead (Pb) and a little graphene oxide. It aims light weight and compact, which will be applied on the transportable nuclear reactor. The material is made up of sixteen layers, and the thickness and components of each layer were designed by genetic algorithm (GA) combined with Monte Carlo N Particle Transport (MCNP). In the experiment, the viscosities of the epoxy at different temperatures were tested, and the settlement regularity of Pb particles and B_4C particles in the epoxy was simulated by matlab software. The material was manufactured at 25 °C, the Pb C and O elements of which were also tested, and the result was compared with the outcome of the simulation. Finally, the material's shielding performance was simulated by MCNP and compared with the uniformity material's. The result shows that the shielding performance of gradient material is more effective than that of the uniformity material, and the difference is most noticeable when the materials are 30 cm thick.

1. Introduction

Light weight and compact are the requirements of the transportable nuclear device [1]. Its shield always takes up a large proportion of the weight and volume. Therefore high performance shielding materials are needed. Gradient materials make it possible to improve the performance of materials. It is known that the segregation is unavoidable in preparing composite materials, especially when there is a big difference between the densities of reinforced phase particles and matrix materials [2], which, in some industries, may reduce the performance of the material in use, such as the mechanical and thermal properties, thus the uniformity material is often preferred. However, the shielding of neutrons and gamma rays often requires a gradual process, that is, the intensity and energy of neutrons and gamma rays are reduced step by step after interacting with each layer of the shielding material, and as the intensity and energy are changed, the combination and proportion of the components of the next-step layer that can reach the optimal shielding effect also alter, different from the previous [3]. Therefore, in order to minimize the radiation at each step, composite gradient materials are more effective than the uniformity material for the proportion of the components for each layer can be designed accordingly to achieve the best effect.

Fig. 1 illustrates the process in details. Generally, the energy of neutrons and gamma rays produced by nuclear reactor, accelerator or decay is high, and at this point, the elements of a high inelastic cross-section were chosen as the first layer, such as Tungsten (W), Lead (Pb), Iron (Fe) and Copper (Cu) [5]. When the energy of neutrons reached the threshold value, the elements of a high elastic cross-section were chosen as the second layer, such as Hydrogen (H), Carbon (C) [6,7]; and then the elements, Boron (B), Lithium (Li) and Gadolinium (Gd) of a high thermal neutron capture cross-section were selected as the third. Since secondary γ-rays are produced when neutrons interact with the material [8,9], the material with W or Pb was chosen as the fourth layer for shielding the primary γ-rays and secondary γ-rays [10].

As mentioned, with the interaction of the material, the energy and intensity of neutrons and γ-rays are reduced gradually, so the components of each layer and their proportions which can realize the best shielding effect are also different. Accordingly, in this

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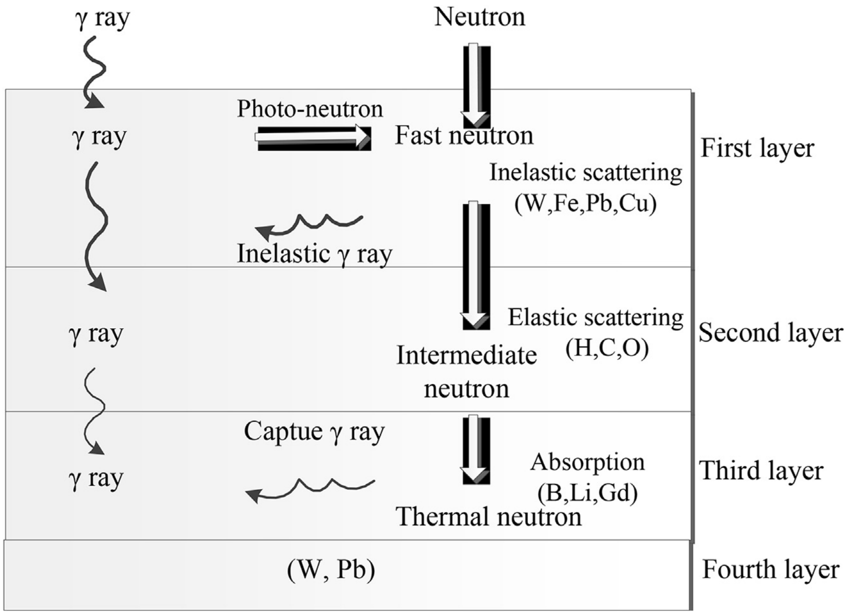


Fig. 1. Interaction of neutrons and γ-rays in the multi-layer material.

paper, the gradient shielding material for mixed neutrons and γ-rays is studied, including the component design, simulation and manufacture of the gradient material.

In our previous studies, the conclusion was obtained, the gradient composite shielding material is better than the uniform mixing and layered material [11]. Therefore, the gradient shielding material is developed in this study.

2. Material design and manufacture

2.1. Material design

The material designed in this research consists of Epoxy risen, Pb and B_4C. It was devised by using GA [12] in combination with the MCNP Code [13]. The version of MCNP is MCNP5 and the cross-section library is 6.2. The parameters, such as the thickness, density and components of the material, were allocated in cell cards and material cards in MCNP code. The data showing the dose equivalents of neutrons and γ-rays were extracted from the output file of the MCNP code, and the extracted data were placed as the objective function of GA. In the GA program, the optimal combination of structure, components and density and their proportions were sought out for a preset objective. In shielding design, the objective is the lowest dose equivalents of neutrons and γ-rays after they penetrate the shielding. The objective function is as follows:

$$\mathrm{Min}H\,(L,A) = \min[\alpha H_n(L,A) + \beta H_\gamma(L,A)] \tag{1}$$

where, $H_n(L,A)$and$H_\gamma(L,A)$are dose equivalents of the neutrons and γ-rays respectively; L is the parameter of thickness; A is the parameter of components; $\alpha$is the number of neutrons and$\beta$is the number of γ-rays in the initial source. The constraint conditions are given by the following equations,

$$\sum_{i=1}^{p} \frac{L_i}{L_{all}} = 1 \tag{2}$$

$$\sum_{i=1}^{p} A_1 + A_2 + \cdots A_i = 1 \tag{3}$$

$$\rho_x \le \rho_{eff} \le \rho_y \tag{4}$$

where, L_i is the thickness of each layer; $L_{all}$is the total thickness of the shield; A_i is the mass ratio of each component and$\rho_{eff}$is the equivalent density of the shielding.

Then, a program was written in C language to combine GA with MCNP to design the shielding materials. The flow chart shown in Fig. 2 contains five steps as follows,

1) Input the parameters of the thickness and the components of the material;
2) Produce the “inpn” file and “inpp” file for simulating the neutrons and γ-rays transmitting in the material;
3) The “inpn” file and “inpp” file are calculated by MCNP, and the “outpn” file and “outpp” are produced;
4) Extract the data showing the dose equivalents of the neutrons in “outpn” file and γ-rays in “outpp” file;
5) The program stops when the fitness value stays the same or the iteration times reaches N_0 (the generation number). If this doesn't work, new thickness and components will be produced, and then the next calculation starts.

The source is the fission neutrons and fission γ-rays. One-time fission produces 2.4 neutrons and 7.7 γ photons, so $\alpha$is 2.4 and $\beta$is 7.7 here. And in the simulation, the 6.13 MeV and 7.12 MeV gamma rays produced by ^{16}N were also studied. The thickness of the material is 30 cm; the crossover rate and mutation rate in GA are 0.3; the population size is 200 and the generation is 500; the densities of Pb, Epoxy risen, and B_4C are 11.34 g cm^{-3}, 1.20 g cm^{-3} and 2.52 g cm^{-3} respectively. It takes about 5000 min to complete a gradient material design when the thickness of the material is 30 cm.

2.2. Manufacture of the material

The material was prepared in room temperature, which mainly

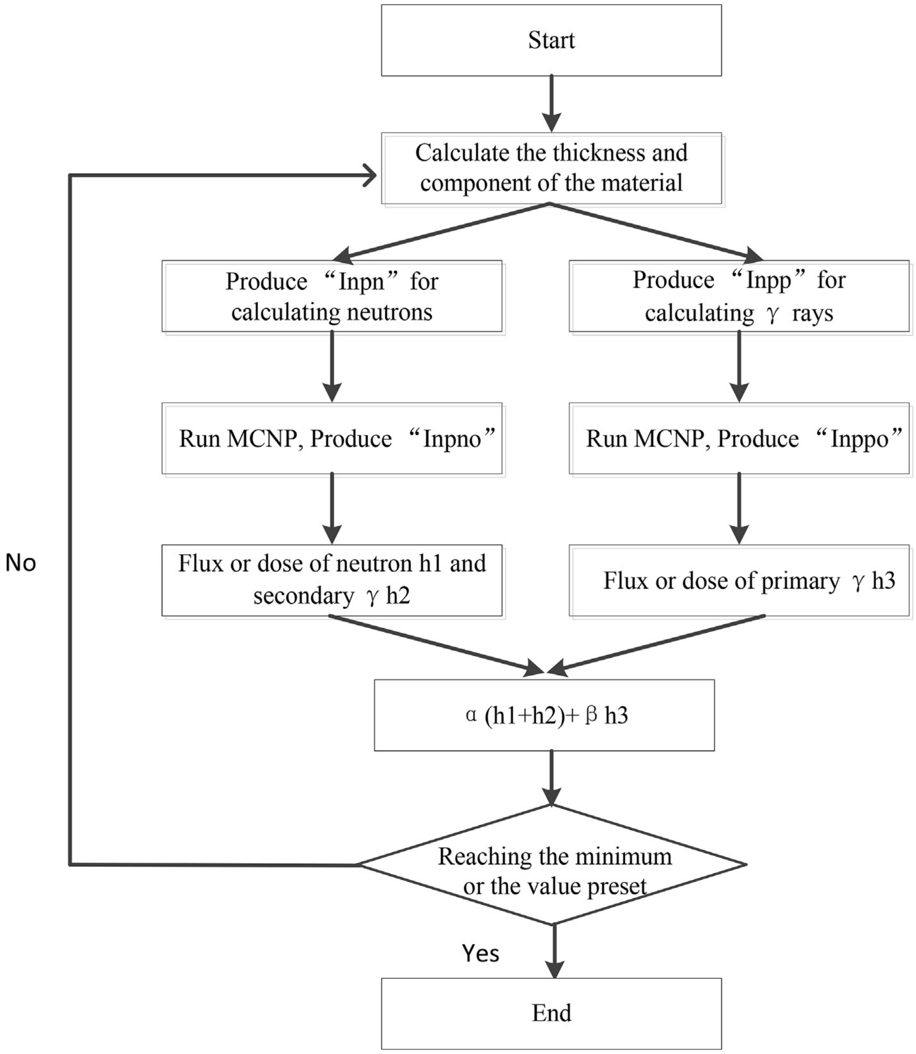


Fig. 2. Flow chart of the design of the shielding structure and components.

included five steps as follows,

1) Mix resin with the curing agent, Pb particles, B_4C particles and Fe particles. The curing agent is polyamide resin. The mass ratio of resin and polyamide resin is 100 to 80. The density of Pb, B_4C, Fe respectively is 11.34 g/cm^{-3}, 2.52 g/cm^{-3},7.86 g/cm^{-3}. The size of Pb, Fe and B_4C are 200 orders.
2) Stir the mixed material in vacuum mixer, which makes the particles equably and the air bubble in the shielding material reduced;
3) Pour the mixed material into an iron mould;
4) Cure the mixed material under the room temperature for many hours;
5) Remove the mould.

The gradient material was formed in step 4). The particles were acted upon by gravity F_g, buoyance F_b, viscous resistance F_d.

The details of gravity F_g, buoyance F_b, viscous resistance F_d are as follows,

$$F_g = mg = \rho_1 Vg = \frac{\pi}{6} d^3 \rho_1 g \tag{2-1}$$

$$F_b = \rho_2 Vg = \frac{\pi}{6} d^3 \rho_2 g \tag{2-2}$$

$$F_d = \varepsilon A \frac{\rho_2 u^2}{2} = \varepsilon \frac{\pi d^2}{4} \frac{\rho_2 u^2}{2} \tag{2-3}$$

The motion equation of the particles is

$$F_g - F_b - F_d = ma \tag{2-4}$$

$$\frac{\pi}{6} d^3 \rho_1 g - \frac{\pi}{6} d^3 \rho_2 g - u \frac{\pi d^2}{4} \frac{\rho_2 c^2}{2} = ma \tag{2-5}$$

where, m is the mass of the particles; g is the acceleration of gravity; ρ_1 is the density of the particles; V is the volume of the particles; d is the diameter of the particles; ρ_2 is the density of the resin and curing agent; u is the viscosity; c is the relative movement speed of the particles.

In formula 2-5, a connects with the diameter and the density of the particles as well as the viscosity of the resin. The diameter and density of the particles were determined before the material was manufactured.

The viscosity u of the material was tested by the experiment.

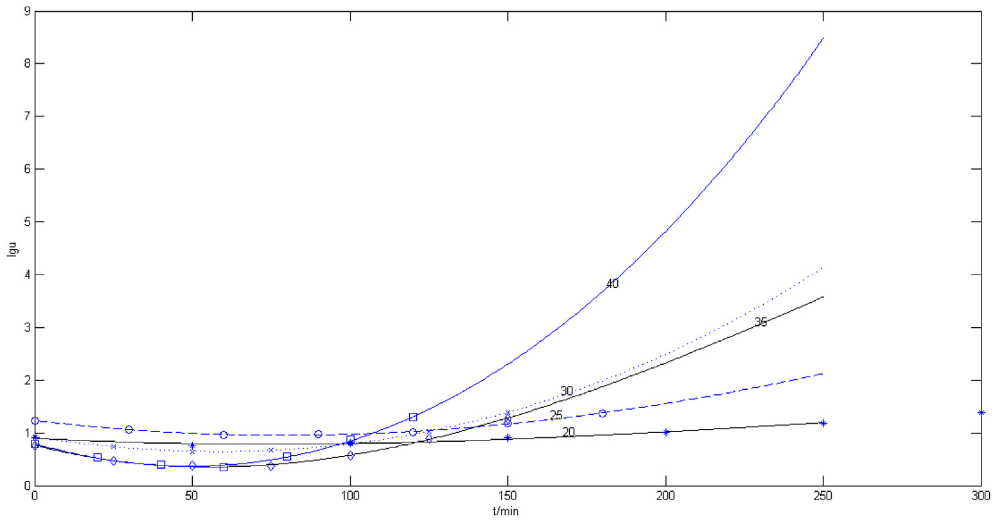


Fig. 3. Experiment data of viscosities of the resin at different temperatures.

The result is shown in Fig. 3, which were tested at 20 °C, 25 °C, 30°Cand 40 °C respectively. It is shown that, at first, the viscosity decreases, but after ten minutes, the viscosity begins to rise.

3. Result and discussion

3.1. Design result of the material

The design result is shown in Table 1. At first, the density of the material decreases with the increase of the thickness, but when reaching a certain point, it increases again. The equivalent density of the material is 3.23 g/cm^{-3}, which means in the interaction of fission neutrons and fission γ-rays with the material, fast neutrons can be firstly moderated by the elastic scattering of lead. With the decrease of the energy of fast neutrons, the lead element of the neutrons which are moderated through the elastic scattering of light elements (H, C, O) gradually decreases, the polymer gradually increases and the density of the material gradually decreases. Then in the process of moderation and absorption, secondary γ-rays will be produced, thus it is important to shield γ-rays when they reach a certain position in the material. That's why the density of the material increases again. The material in this article is designed to optimize the components of each layer of the material according to the neutrons and γ-rays which are needed to be shielded.

Table 1
Design result of the material.

Layer number	Thickness (cm)	Mass ratio of the component (W/%)			Density (g/cm^{-3})
		Resin	B_4C	Pb	
1	0.257	0.005	0.006	0.989	10.665
2	0.584	0.022	0.004	0.974	9.451
3	1.333	0.085	0.009	0.906	6.48
4	1.920	0.168	0.015	0.817	4.587
5	1.606	0.193	0.021	0.786	4.193
6	2.622	0.282	0.009	0.709	3.321
7	1.484	0.415	0.033	0.552	2.453
8	3.704	0.668	0.054	0.278	1.659
9	2.581	0.803	0.048	0.149	1.425
10	4.214	0.932	0.066	0.002	1.245
11	2.114	0.796	0.075	0.129	1.419
12	2.639	0.549	0.052	0.399	2.443
13	1.470	0.327	0.031	0.642	2.928
14	2.025	0.141	0.017	0.842	5.037
15	1.004	0.035	0.008	0.957	8.566
16	0.435	0.008	0.001	0.991	10.587

3.2. Simulation result of the forming process of the gradient material

The simulation was done by matlab software. The initial state is shown in Fig. 4 and the forming process of the gradient material is shown in Fig. 5. The size of the particle is 200 orders. It assumes that there is an upward magnetic force. The initial state of the material is uniform mixing. The particles were divided into two groups, with one group receiving upward force and the other downward force. With the action of force, the particles gradually moved up and down. Fig. 5 shows the process. Finally, with the completion of curing reaction, the particles no longer moved; at this point the material with gradient distribution was formed.

3.3. Comparison between the simulation and experiment data

The experiment was initially carried out in an environment only with gravity to verify the settlement of the lower part of the gradient material which was prepared at 25 °C with a height of 10.8 cm. As shown in Fig. 6, the prepared material was tested through mapping to detect the contents of Pb, C, and O at several positions.

In the test, the corresponding standard samples of C, Pb and O are $CaCO_3$, PbF_2, SiO_2, and they were repeatedly tested for four times. Table 2 gives the tested mass ratio and atom ratio of the material, and the results are normalized.

The comparison between the experimental and the simulation results is shown in Fig. 7. It is assumed that the entire gradient material is made up of two small pieces which are divided by the dotted line in the figure. The entire material was simulated, but due to limited experimental conditions, the part under 19.25 cm of this material was tested. It can be seen from the figure that except for the lowest position point, within the error range, the experimental results are in agreement with the simulation results at other position points. The error of the simulation mainly comes from the MCNP software, and the error of the experiment comes from the mapping test result.

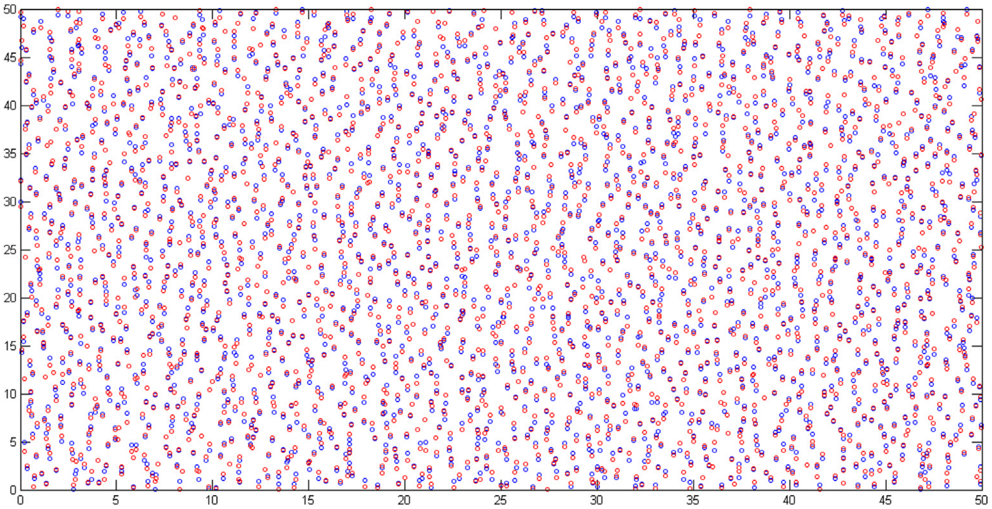


Fig. 4. Initial state of the material.

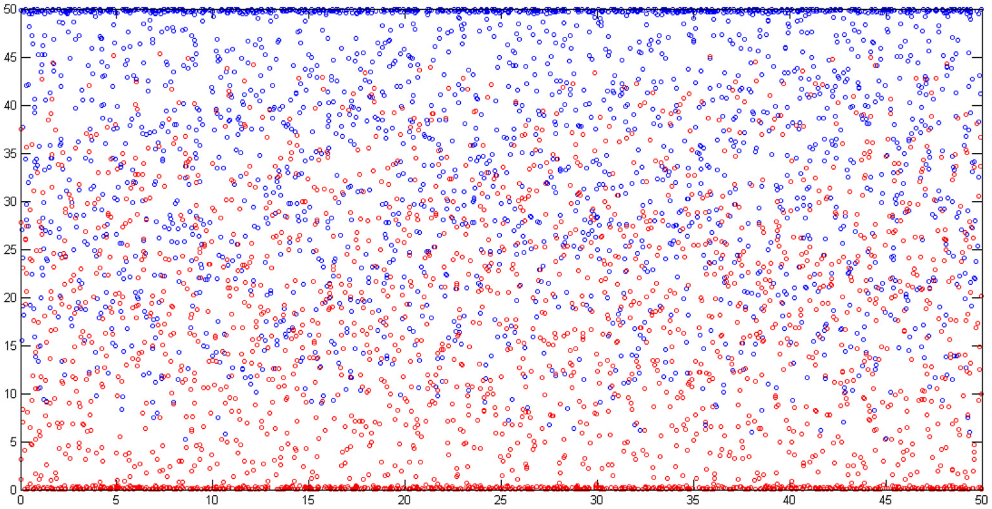


Fig. 5. Forming process of the gradient material.

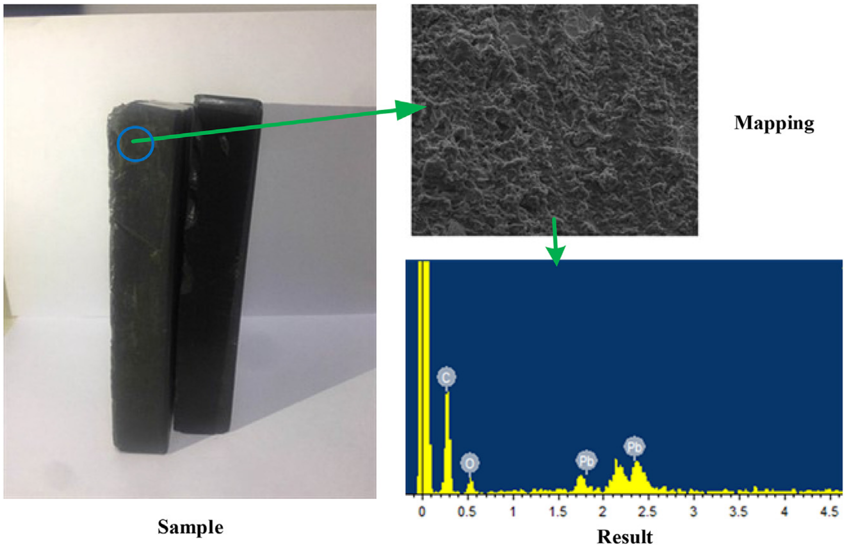


Fig. 6. Mapping result of the material at 21.36 cm.

Table 2
The result of the sample.

Element	Mass ratio	Atom ratio
C	63.85	78.42
O	22.34	20.60
Pb	13.81	0.98

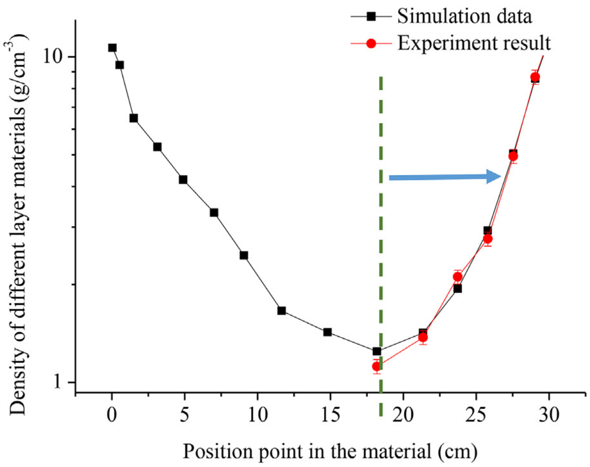
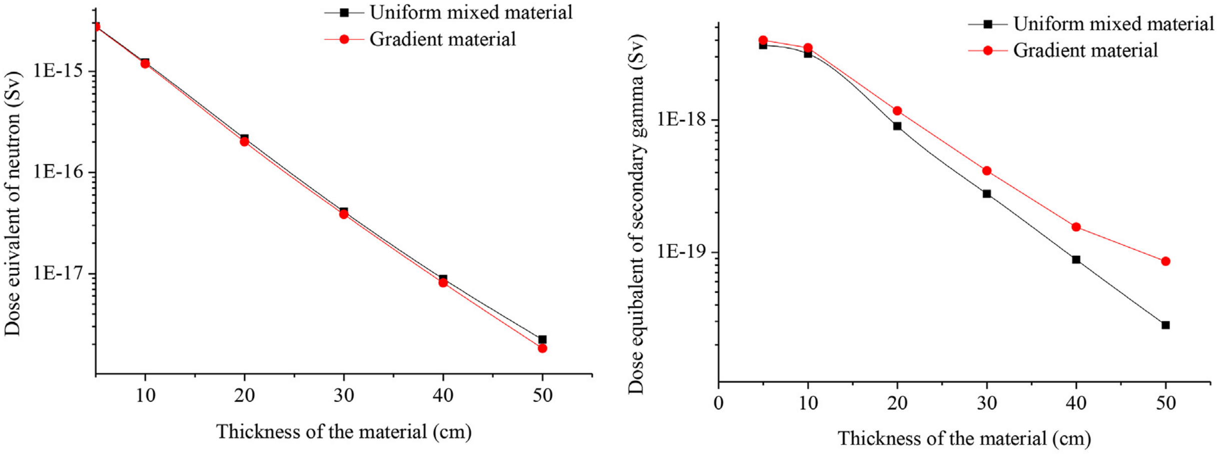


Fig. 7. Comparison between the simulation data and the experiment result.

3.4. The performance of the gradient material

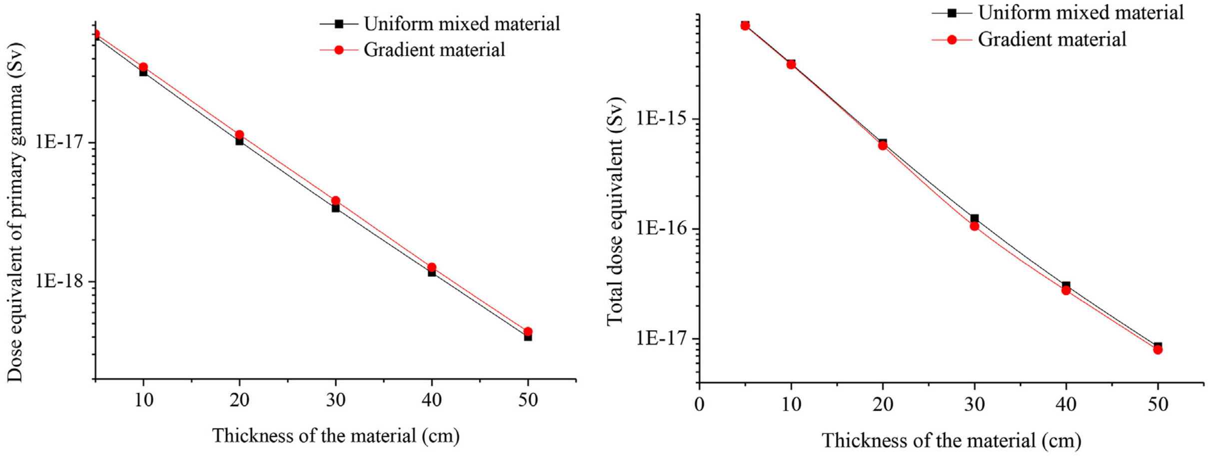
After comparing the experimental and simulation results, the shielding effects of the two materials were compared by MCNP software, for the gradient material in this paper is developed in accordance with the interaction of rays and the material to improve its shielding performance.

The two materials are uniform mixed material and designed gradient material. The results are shown in (a), (b), (c), and (d) of Fig. 8, which are respectively shielding effects of neutrons, secondary γ-rays, primary γ-rays and the total shielding effect of neutrons and γ-rays. According to the results, with the increase of thickness, the shielding effect of the gradient material is better than that of the uniform mixed material. However, in the shielding process, more secondary γ-rays are produced. Besides, in shielding γ-rays, the performance of gradient materials is not as good as uniform mixed materials, but it is not that noticeable. For the total shielding effect of neutrons and γ-rays, it can be seen from the results of (d) that the shielding effect of the gradient material is obviously better than that of the uniform mixed material, especially at about 30 cm. At 10 cm, 20 cm, 30 cm, 40 cm, 50 cm, the shielding effects of the gradient material were 1.99%, 5.21%, 18.4%, 10.6% and 6.95% better than that of the uniform mixed material.



(a) Neutron

(b) Secondary gamma



(c) Primary gamma

(d) Total

Fig. 8. Comparison between the uniform mixed material and the gradient material.

4. Conclusion

A gradient composite shielding material is developed. Its features are summarized as follows:

(a) The gradient shielding material with 16 layers was designed and the simulation of the forming process of the gradient material was done;
(b) The sample of the material was prepared and the element was tested;
(c) The gradient material is obviously better than that of the uniform mixed material, especially at about 30 cm, since the design sample is 30 cm, at which the component and its distribution of the material is optimal.

It can be concluded that this material will be a good alternative as a shielding material for transportable nuclear reactor.

Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2020.03.029.

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