Investigation of an Innovative Material for Neutron Shielding

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Abstract. The effectiveness in neutron attenuation of an innovative composite material for neutron shielding has been investigated in comparison with a conventional neutron shielding material – paraffin. For this purpose a set of calculations were performed and validated with adequate measurements. The Monte Carlo N-Particle (MCNP) computer code was used for these calculations. The measurements were performed with a certified Pu-Be neutron source. The results from the calculations show that the innovative composite material – consisting of 30 wt.% boron carbide and 70 wt.% polyethylene – is on average 13% more effective in attenuating of neutrons than paraffin, while having much better mechanical properties.

Keywords: neutron shielding, boron carbide, MCNP, composite.

1 Introduction

A well-known fact is that the best materials for neutron shielding must be able to slow down neutrons which can be fulfilled only by material containing light atoms (e.g. hydrogen atoms), such as water, polyethylene, and concrete. Thermal neutrons can be easily absorbed by capture in materials with high neutron capture cross sections (thousands of barns) like boron, lithium or cadmium. Generally, only a thin layer of such absorber is sufficient to shield thermal neutrons. It has to be noted that moderation of neutrons to thermal energy is practically a sufficient radiation protection measure in respect of the neutron component of the equivalent dose. Hydrogen (in the form of water), which can be used to slow down neutrons, has absorption cross-section of 0.3 barns. This is not enough, but this insufficiency can be offset by sufficient thickness of the water shield. In the case of cadmium shield the absorption of neutrons is accompanied by strong emission of gamma rays. Therefore additional shield is necessary to attenuate the gamma rays. This phenomenon practically does not exist for lithium and is much less important for boron as a neutron absorption material. For this reason, materials containing boron are used often in neutron shields. In addition, boron (in the form of boric acid) is well soluble in water making this combination very effective neutron shield. However, boric acid is highly corrosive, which requires the use of stainless steel tanks and piping.

2 Description of the Innovative Material

The Institute for Nuclear Research and Nuclear Energy (INRNE), Bulgarian Academy of Sciences, has been supplied with samples of an innovatively acquired composite neutron shielding material, consisting of 30 wt.% boron carbide (B_4C) and 70 wt.% polyethylene (PE). These samples are produced by a patented method [1] – powdered B_4C is homogeneously dispersed into molten polyethylene matrix and after that the mixture is cast in the desired shape for which there is almost no limitation. The presented samples are with circular and square shape and with varying thickness between 3 and 8 mm (most of them are 5 mm thick). A typical sample of the composite material is presented in Figure 1.

Figure 1. Circular sample (5 mm thick) of B_4C + PE composite material (the ruler is in cm.

The composite is very compact and at the same time very effective neutron shielding material with density of 1.16 g/cm^3. High mechanical, temperature and radiation resistive properties of the innovative material have been proved. Its radiation resistance was experimentally demonstrated by irradiating samples of the composite material placed right behind the reactor pressure vessel during campaign No.18 at Unit 6 of Kozloduy Nuclear Power Plant. The irradiated samples showed no change in their weight, density, dimensions and shape. Furthermore, the measured residual gamma-radiation was very low. All these characteristics show that this composite material can be used for neutron shielding where there is not enough space to place conventional neutron shielding materials such as paraffin or 5% borated polyethylene.

3 Summary of the Investigation

The scope of this work is to prove the effectiveness of the B_4C + PE composite in comparison with paraffin. For this purpose a set of calculations were performed and validated with adequate measurements. The Monte Carlo N-Particle (MCNP) computer code [2] was used for these calculations. MCNP is a widespread general-purpose program for transport calculations by the Monte Carlo method of almost all particles in their almost entire energy range. Geometric objects modeling in MCNP is relatively easy: the code treats an arbitrary three-dimensional configuration of materials in geometric cells bounded by first- and second-degree surfaces and fourth-degree elliptical tori.

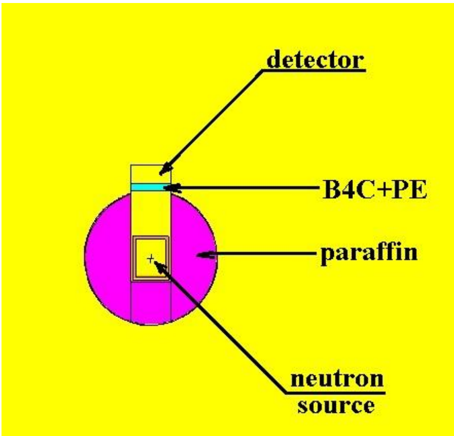

Figure 2. MCNP model visualization.

The Pu-Be neutron source with its characteristic spectrum was modelled in MCNP (see Figure 2). Although the real neutron source has a cylindrical shape with dimensions 12 mm diameter $\times$ 25 mm height, for the purpose of the calculations it was modelled as a point source. Nevertheless, the stainless steel capsule (4 mm thick) and the copper lining (1 mm thick) of the paraffin sphere are accounted for in the model. All of the areas colored in yellow in Figure 2 represent space filled with air. The detector was modelled as a cylindrical space filled with air with dimensions 60 mm diameter $\times$ 28 mm height. These dimensions were chosen in order to correspond with the dimensions of the neutron detector which was a cylinder 28 mm in diameter and 260 mm long. All the measurements were performed with the neutron detector positioned perpendicular to the axis of the cylindrical cavity of the paraffin sphere. The B_4C + PE composite material was modelled also as cylinders with 60 mm diameter and different heights. The actual samples are with diameter of 75 mm and some of them have square shape with dimensions 70×70 mm.

Pu-Be sources are fast neutron sources, where neutrons are produced mainly through the ^{9}Be$(\alpha, n)^{12}$C* reaction with a relatively small contribution from the self-multiplication effect due to the neutron induced fission on Pu isotopes and (n,2n) reactions on ^{9}Be and other nuclides present in the source construction materials. The sources include an active cylinder made of an inter-metallic compound of plutonium and beryllium, which is tightly encased in a two-fold stainless steel capsule. The intensity of typical Pu-Be sources does not exceed 10^8 n/s. Alpha-beryllium neutron sources produce approximately 30 neutrons for every one million alpha particles. The fast neutrons from Pu-Be sources are always accompanied by less intense photon radiation originating mainly from the decay of Pu isotopes and accumulated ^{241}Am as well as from the de-excitation of ^{12}C, yielding 4.44 MeV photons. The other less important sources of gamma-radiation include bremsstrahlung of secondary electrons and various $(n, x\gamma)$ activation reactions occurring in the source and its surrounding materials [3].

In order to validate the calculations, a set of measurements were performed. The measurements were performed with a certified Pu-Be neutron source used for calibration of the radiation monitoring equipment at the Nuclear Scientific Experimental and Educational Centre of INRNE. The intensity of the source is 5×10^5 n/s and it is placed in a copper sphere filled with paraffin. The measurements were performed from a distance, taking all the necessary radiation protection measures, because the shielding plug of the neutron source had to be removed. A metrologically tested system consisting of a portable wide range multi-purpose digital survey meter FH 40 G-L10 and a neutron detector FHT 752S (which contains BF_3 tube) was used to measure the count rate of all neutrons that passed through the investigated samples. Both devices are manufactured by Thermo Scientific [4]. The neutron detector was fixed on a stand at 10 cm from the upper end of the copper sphere of the neutron source. The results of the measurements are shown in Table 1.

Table 1. Results from the measurements

Thickness of B_4C + PE, [mm]	Neutron count rate, [s^{-1}]	Thickness of B_4C + PE, [mm]	Neutron count rate, [s^{-1}]
0	340 ± 2	40	120 ± 2
10	225 ± 2	45	109 ± 2
20	188 ± 2	50	105 ± 2
25	164 ± 2	55	97 ± 2
30	136 ± 2	60	94 ± 2
35	131 ± 2		

The measured and calculated results for the neutron count rate are presented in Figure 3. It is clearly shown that the calculations evaluate in a conservative manner the attenuation of the neutrons through the material's thickness. This is important when evaluating the properties of material from radiation protection point of view.

The neutron flux attenuation through the B_4C + PE composite thickness has been evaluated against that of paraffin, which is a common neutron shielding material, by means of neutron transport calculations. The results from the calculations (see Figure 4) show that the innovative

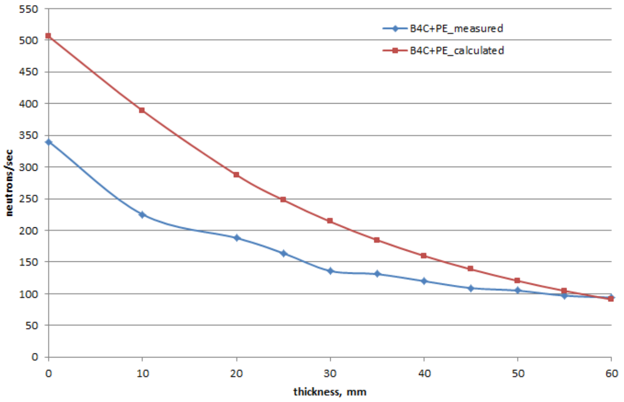

Figure 3. Comparison between calculated and measured results.

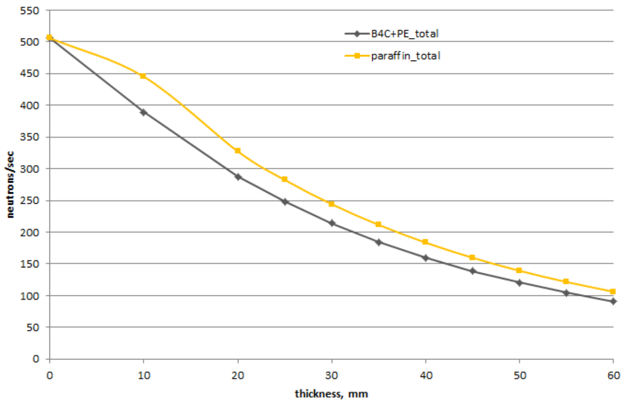

Figure 4. Results from the MCNP calculations for the total attenuation of B_4C + PE and paraffin.

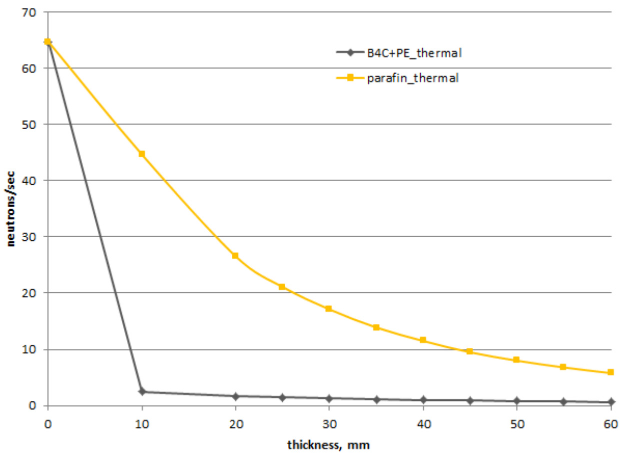

Figure 5. Results from the MCNP calculations for the thermal neutron absorption of B_4C + PE and paraffin.

composite material is on average 13% more effective than paraffin. Furthermore, 10 mm of the B_4C + PE composite absorbs more than 95% of the thermal neutrons (see Figure 5) and about 35% of the epithermal neutrons that were calculated to enter the detector area (Figure 6), while the same thickness of paraffin shields only about 30% of the thermal and about 15% of the epithermal neutrons. An evaluation of the accumulated neutron dose rate is presented in Figure 7 to assess the radiation protection properties of the investigated materials. The statistical uncer-

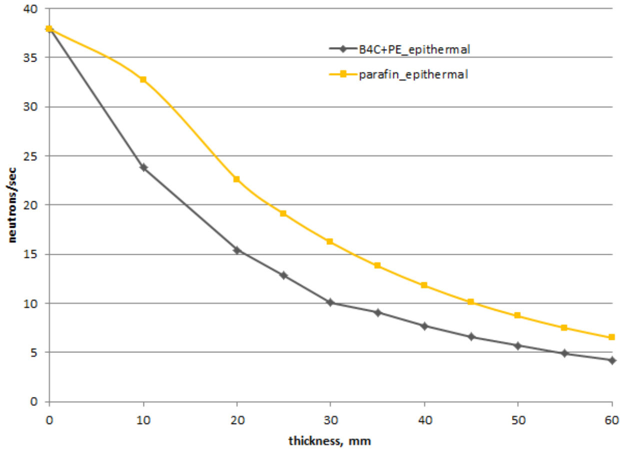

Figure 6. Results from the MCNP calculations for the epithermal neutron attenuation of B_4C + PE and paraffin.

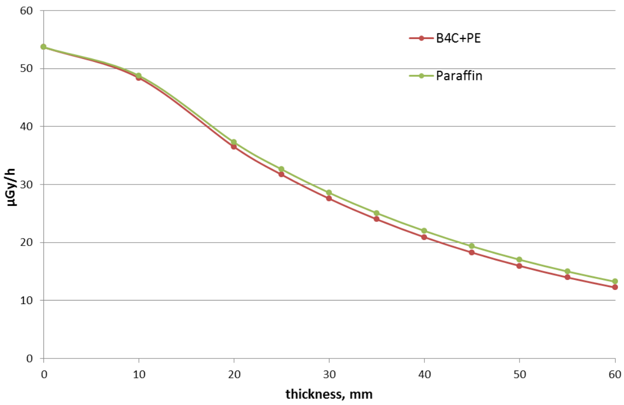

Figure 7. Results from the MCNP calculations for the total neutron dose rate on the external surface of the shielding materials (B_4C + PE and paraffin).

tainty from the calculations for both investigated materials is in the range of $0.08 \div 0.15\%$ and rises with the increase of the material thickness.

4 Practical Implementation

In 2015 10 mm thick composite neutron shielding material was installed on the side walls of a 20-feet stainless steel ISO container for storage of neutron sources on the site of the Permanent Repository for Radioactive Wastes – Novi Han. The effectiveness of the shielding was conservatively assessed by MCNP calculations for the total (neutron and gamma) equivalent dose rate at 1 m from the ISO container. The results from these calculations showed that 10 mm PE+B_4C would reduce the total equivalent dose rate with 13%. After the shielding was installed the actual measurements showed that the reduction of the equivalent dose rate was almost 30%. The measurements were performed again with Thermo FH 40 G-L10 and FHT 752 which is a neutron dose rate probe with range of 1 nSv/h $\div$ 0.4 Sv/h acc. to ICRP 60 [5]. The internal proportional counter tube (measuring range: 10 nSv/h $\div$ 100 mSv/h) of the portable survey meter was used for the gamma ambient equivalent dose rate.

5 Conclusion

Results for the neutron flux attenuation through innovative neutron shielding material, consisting of 30% B_4C + 70% PE composite have been obtained by means of neutron transport calculations. The results have been verified experimentally. The attenuation through the composite material was compared with the attenuation through common neutron shielding material – paraffin. From the results it can be concluded that the B_4C + PE composite material is on average 13% more effective than paraffin in attenuation of neutron radiation, and has similar radiation protection properties while having much better mechanical properties. In order to increase the cost efficiency of the neutron shielding based on this material a layered structure from pure polyethylene ($20 \div 40$ mm thick) and B_4C + PE ($10 \div 20$ mm thick) is going to be investigated as a future work.

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