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Monte Carlo simulations of gamma-rays shielding with phthalonitrile - tungsten borides composites

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► ***Original article***

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ABSTRACT

***Background*:** Recently, it has been demonstrated that thermosetting polymer composites have excellent gamma-rays shielding properties. Among them, phthalonitrile composites are the best suitable materials to replace the traditional shielding materials such as lead and concrete. Furthermore, tungsten-boride as high Z-material is an effective ionizing radiation shield. ***Materials and Methods*:** The gamma rays shielding properties of the phthalonitrile matrix (PH) reinforced with tungsten-boride (WB) at 661, 1172, and 1332 keV photon energies were investigated using MCNPX code and XCOM tool kit and compared to those of concrete as conventional shielding material and epoxy composites. MCNPX geometry was defined along the z-axis and described in the input file. The number of emitted photons was fixed at 10^7 at the source, which is supposed to be a monoenergetic point. ***Results*:** The simulated mass attenuation coefficients results are in good agreement with those calculated using the XCOM tool kit. Also, it was observed that at 661 keV photon energy, the shielding performances in term of Half-Value-Layer (*HVL*) are enhanced by3.08% and 22.01% for 30% and 50% of tungsten-boride concentrations compared to 30% of PbO concentration in the Epoxy-Clay composite and concrete respectively. ***Conclusion*:** In this study, the outstanding results of gamma-rays shielding properties of Phthalonitrile/WB composites (PHWB) obtained using MCNPX code and XCOM can be used for future experimental gamma-rays shielding approaches at a wide range of energy.

INTRODUCTION

The gamma rays are high penetrating electromagnetic radiations in the matter, unlike alpha and beta rays, they can pass easily through lightweight materials such as the human body. Hence, the gamma rays shielding constitutes a great challenge to the radiation shielding field [(1)]. In recent years, the development and utilization of gamma radiations have gained significant interest in the fields of space technology, dosimetry, radiotherapy, nuclear medicine, nuclear energy, and civil engineering [(2–6)]. However, the hazardous use of these radiations can expose the human being and its environment to the various harmful effects [(7–9)]. In order to enhance the gamma rays shielding properties of the materials, the traditional shielding materials such as concrete and lead are being gradually replaced by the new lightweight shielding materials such as thermosetting polymer composites [(10–13)]. Indeed, using the MCNPX code, Tekin and Manici (2017) simuated the mass attenuation coefficients of cement plaster, bricks, and concrete and the results were compared with the experimental and XCOM data ones at 59.5, 356, 662, 1173, 1274 and 1333 keV energy [(14)]. Moreover, Epoxy/Clay/PbO composites were prepared by Amin *et al*., in 2019 and they found that the experimental gamma rays shielding properties agreed well with the theoretical results (XCOM database) by using the ^{137}Cs and ^{60}Co sources [(15)].

Phthalonitrile resin is considered as a type of high -performance polymer thermosetting because of its interesting properties such as ease of processing, high glass transition temperature (Tg), good thermal stability, excellent mechanical properties, and excellent fire-resistant performance [(16–18)]. In the recent decades, many authors were interested in the preparation and characterization of the phthalonitrile resin and its related composites. The thermal and mechanical properties of the phthalonitrile reinforced with the ceramic Ti_3SiC_2 fillers were investigated by Derradji *et al*., in 2018 [(19)]. The gamma irradiation, chemical structure, mechanical, thermal and thermomechanical properties of benzoxazine/phthalonitrile copolymers were

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investigated using FTIR (Fourier-transform infrared spectroscopy), DMA (Dynamic mechanical analysis) and TGA (Thermogravimetric analysis) by Qadeer *et al.*, in 645^2 (a) [20]. The mechanical properties as well as gamma-rays shielding properties of the phthalonitrile reinforced with basalt fibers were improved by Derradji *et al.*, in 2018(c) [21]. The eugenol based phthalonitrile (EPN) monomer/ benzoxazine (P-a and BA-a) were copolymerized and the impacts of EPN loading (10 to 40%) on mechanical, thermomechanical and thermal properties were evaluated by Qadeer *et al.*, in 2018 (b) [22]. Recently, besides the thermomechanical, thermal and mechanical properties for phthalonitrile composites, its gamma rays shielding and irradiation properties have been investigated experimentally, and outstanding improvements were observed by Derradji *et al.*, in 2018(d) and Medjahed *et al.*, in 2019, respectively [23, 24].

Tungsten-boride is the mixture of a light-element compounds and a transition-metal. It has displayed great interest due to its good electronic conductivity, electrical resistance at high temperature, wear resistance, excellent mechanical properties, strong resistance to oxidation, chemical inertia, high melting points, and ultra-hardness that is equal or superior to those of traditional super hard materials (high pressure synthesized expensive materials) such as boron nitrides or diamond [25–27]. These exceptional characteristics favor them in many applications as polishing, cutting, coatings and abrasive [28, 29].

Monte Carlo simulations are based on the probabilistic methods (stochastic methods) and widely used in many studies. MCNPX (Monte Carlo Neutral-Particle eXtended) is developed in Los Alamos National Laboratory and among the most used software to simulate the gamma rays shielding properties [30]. This code can be used to simulate and solve the problem of radiation transport from neutrons, photons, and electrons or a mixture of neutrons, photons, and electrons [31].

The current work focused on the investigation of the gamma ray shielding properties for the phthalonitrile (PH) matrix reinforced with tungsten borides (WB) using the MCNPX code and the XCOM database. To confirm the reliability of our findings, these numerical data were compared to somes recent experimental results in the field of this study.

MATERIALS AND METHODS

In the current study, MCNPX code (version 2.7.0) provided by Jordan Atomic Energy Commission, Jordan Research and Training Reactor and XCOM database [32] were used to simulate the gamma rays shielding properties of WB micro-particles reinforced phthalonitrile matrix (PH) [33]. Indeed, the PH matrix has a density of 1.238 g/cm^3 while WB has a density of 15.2 g/cm^3 and the radius of one micrometer. These characteristics of WB fillers were provided by the Institute of Composite Materials, College of Materials Science and Chemical Engineering, Harbin Engineering University, People's Republic of China.

Theoretical calculations

XCOM is an online database using a mixture of pre -existing databases for attenuation interaction mechanisms processes as photoelectric absorption, coherent and incoherent scattering, and pair production in the range from 1keV to 100GeV photon energy [32]. The quantities tabulated of the compounds or mixture, such as the partial and total mass interaction coefficients, are the product of the corresponding cross-sections multiplied by the number of target molecules per unit mass of the considered material. The complete attenuation coefficient is equal to the sum of the interaction coefficients for the individual process. In the present investigation, the total attenuation coefficient without the coherent dispersion used in the gamma-rays transport calculations is expressed in the equation

$$\mu_m = \sum_i W_i \left(\frac{\mu}{\rho}\right)_i \qquad (1)$$

Where μ_m represents the mass attenuation coefficient, $\left(\frac{\mu}{\rho}\right)_i$ is the elemental mass attenuation coefficient in the corresponding compound, μ is the linear attenuation coefficient, ρ is the density and W_i is the fractional weight of the ith element in the compound.

In the case of suitable geometry, the following equation named the Lambert-Beer's law can be used calculating The shielding properties such as the linear attenuation coefficient (μ), the transmittance Ln(I_0/I), HVL and TVL [34]:

$$I = I_0 \exp(-\mu x) \qquad (2)$$

With I_0 the incident photon energy, I the transmitted photon energy and *x* the thickness of the shielding material. The calculation of the composite density can be performed using equation (3) [15]:

$$\rho_c = \frac{100}{\frac{100 - W_{WB}}{\rho_{PH}} + \frac{W_{WB}}{\rho_{WB}}} \qquad (3)$$

Where, ρ_c is the composite sample's density, ρ_{PH} is the phthalonitrile density, ρ_{wb} and W_{WB} the tungsten-boride's density, and it is fractional weight, respectively.

The Radiation Protection Efficiency (RPE), often called the Screening ratio(S), is the parameter that measures the performance of the shield to reduce the ionizing rays at a reasonable level. It can be calculated using equation (4).

$$RPE = 100 \times (1 - \exp(-x / mfp)) \qquad (4)$$

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MCNPX simulations

MCNPX is a Monte Carlo computer code using Fortran90 that solves the radiation transport at almost all particles and all energies [35]. Moreover, MCNPX is an extension of the MCNP4C3 version and consequently has many features beyond MCNP4C3. In this study, the MCNPX code is used to simulate the photon attenuations across different samples of Phthalonitrile/WB composite. The model geometry in figure 1 is defined as a 50x10x2 mm^3 cubic sample inserted into an air sphere and enclosed by an external vacuum. The MCNP Tally F4 was used in the detector for the completion of the simulations. The input file for each sample was run until all statistical checks have been passed.

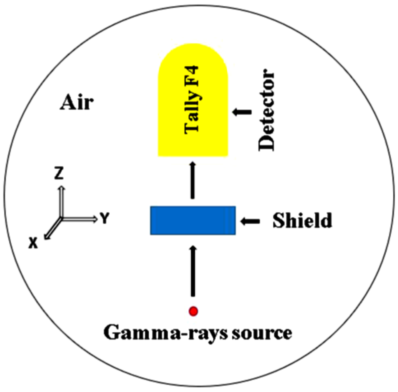

Figure 1. MCNPX simulation geometry (the figure is not in scale).

RESULTS

Elements and density determination

The composites' densities were calculated according to equation (3) and by using table 1 data which is obtained from XCOM mixture of PHWB elements. According to table 1, the density of PHWB composite increases with increasing of the WB fillers due to the high density of tungsten-boride fillers.

Table 1. Elemental concentration by weight fractions of PHWB composites.

Composite	PHWB0	PHWB10	PHWB20	PHWB30	PHWB40	PHWB50
H	0.03	0.028	0.025	0.022	0.019	0.016
B	0	0.006	0.011	0.017	0.022	0.027
C	0.75	0.675	0.6	0.525	0.45	0.375
N	0.22	0.197	0.175	0.153	0.131	0.11
W	0	0.094	0.189	0.283	0.378	0.472
Density (g/cm^3)	1.238	1.363	1.517	1.709	1.957	2.289

PHWB(X): Phthalonitrile (PH)/Tungsten-Boride (WB) sample; X means concentration of WB

Validation of MCNPX simulations

To validate MCNPX geometry modeling and simulations, the output showed that all the ten statistical verifications were passed with relative errors about less than 0.1%. Furthermore, in table 2, the relative deviation in the range of (0.17 %-12.54 %) between MCNPX and XCOM results for mass attenuation coefficients at 661, 1172, and 1332 keV photon energy showed that the MCNP modeling could be confirmed as input standard for any type of composite samples.

Figure 2 presents the XCOM calculated mass attenuation coefficients of the phthalonitrile/WB composite as a function of ^{137}Cs and ^{60}Co photon energy. The results showed that the mass attenuation coefficient is increasing with the Tungsten-boride concentration going up at ^{137}Cs energy and decreases at ^{60}Co energy photon.

The transmittance Ln (I_0/I) of gamma-rays versus different thicknesses is presented in Figure 3, 4, and 5 respectively at 661, 1172, and 1332 keV photon energy. The linear attenuation (μ) obtained from equation (2) is the slope of Ln(I_0/I) and identified to increase with increasing the thickness as reported in these figures. For instance, μ were recorded for PHWB0 samples as of 0.0965 cm^{-1} and increased to a maximum value of 0.2264 cm^{-1} for the PHWB50 sample in figure 3.

performance composites (Epoxy-Clay-PbO) [14] and traditional shielding material (concrete) [15] at 661keV, 1172keV and 1332keV energy source, respectively. As the results showed, an increase in WB content leads to a decrease in both *HVL* and *TVL* in figure 4. In can be noted also from that figure that at Co-60 source, the *HVL* of PHWB0 sample is closed to those of ECP0, while at Cs-137 source, *HVL* of PHWB50 sample is comparable to those of concrete.

In figure 5, the *RPE* was obtained by varying the thickness of the samples and introducing the simulated mean free path (*mfp*) into equation (4). Subsequently, the μ_m values obtained by MCNPX code and XCOM for each PHWB micro composites are compared in table 2, and the deviations show that the simulated results are in good agreement with that of the XCOM database.

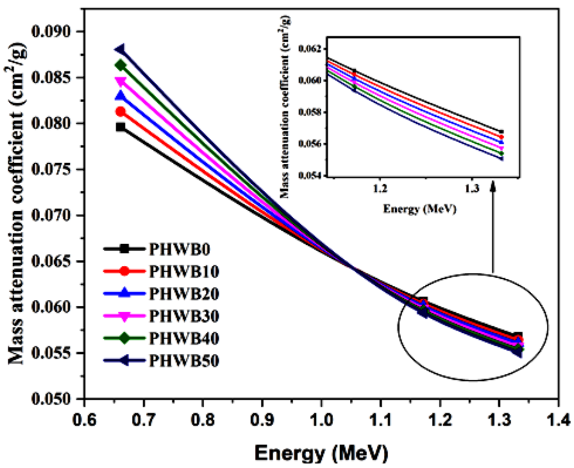

Figure 2. Mass attenuation coefficients using XCOM for PHWB samples versus energy sources.

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Table 2. Comparison of Mass attenuation coefficients obtained using MCNPX and XCOM.

Composite	661 keV			1172 keV			1332 keV		
	MCNPX	XCOM	Dev.	MCNPX	XCOM	Dev.	MCNPX	XCOM	Dev.
PHWB0	0.0779	0.0796	2.18	0.0593	0.0606	2.19	0.0556	0.0568	2.16
PHWB10	0.0910	0.0813	10.65	0.0593	0.0604	1.85	0.0547	0.0564	3.10
PHWB20	0.0949	0.0830	12.54	0.0594	0.0601	1.18	0.0545	0.0561	2.93
PHWB30	0.0966	0.0847	12.32	0.0592	0.0599	1.18	0.0544	0.0558	2.57
PHWB40	0.0979	0.0863	11.85	0.0594	0.0596	0.34	0.0543	0.0554	2.02
PHWB50	0.0989	0.0880	11.02	0.0595	0.0594	0.17	0.0545	0.0551	1.10
Dev. = 100 × $\left|(\mu_{m(MCNPX)} - \mu_{m(XCOM)}) / \mu_{m(MCNPX)}\right|$									

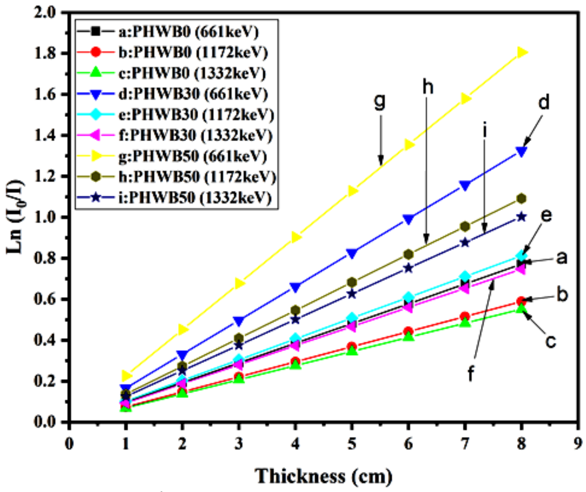

Figure 3. Ln (I0/I) against thicknesses for the phthalonitrile/WB samples at Cs-137 and Co-60 energy sources.

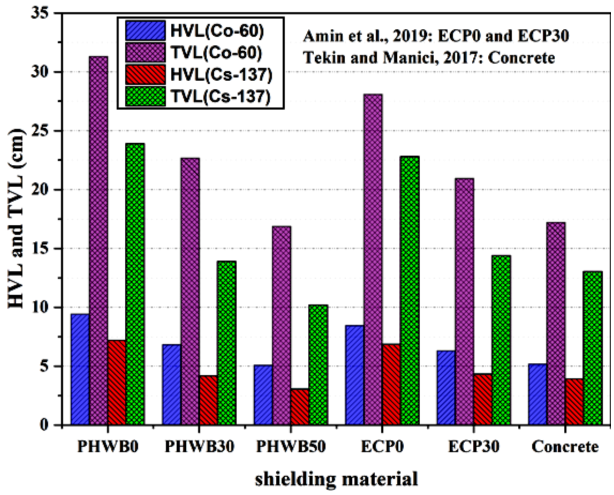

Figure 4. Comparison of HVL and TVL of Phthalonitrile/WB samples with some materials at Cs-137 and Co-60 energy sources.

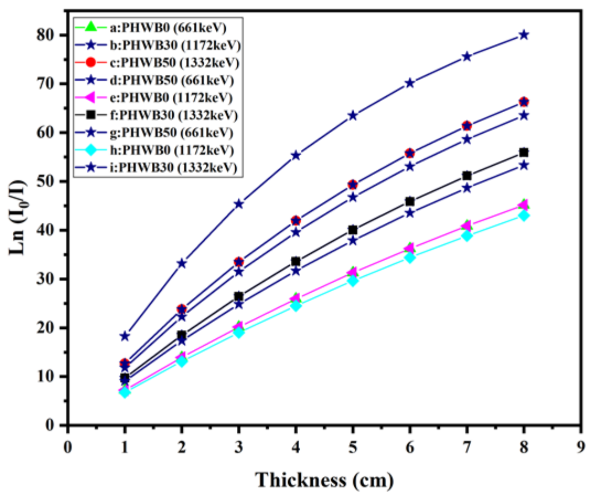

Figure 5. The radiation protection efficiency as the function of phthalonitrile/WB composites' thickness different energies.

DISCUSSION

As shown in figure 2, adding WB microparticles to the phthalonitrile matrix increases the attenuation coefficients of the PHWB composites, and the assessment is more significant at ^{137}Cs gamma ray photon energies. These behaviors of the mass attenuation coefficients are due to the Z-dependence cross-section of different gamma ray interaction processes [36]. Figure 3 showed that the transmittances are improved due to the high degree of crosslinking of the phthalonitrile matrix [23, 24] and the exceptional properties of the WB micro-particles, which absorb a significant quantity of photons gamma incidents. Figure 4 showed that HVL shielding performance improved by 3.08% and 22.01% respectively for PHWB30 and PHWB50 compared to ECP30 composite and concrete composite. Furthermore, in this figure, the shielding capabilities (*HVL*) of PHWB94 micro composite are slightly superior to those of concrete. Besides, it is essential to point out that, with 3 cm thick samples, the PHWB50 composite displayed the *RPE* values of 33.54% against 18% for the reinforced Epoxy matrix composites with the same thickness at the same photon energy by other researchers [37, 38]. These improvements of gamma rays shielding results are due to many factors, among which the use of the WB fillers which are a class of superhard materials with high density [39–41]. Recent experimental studies confirm that phthalonitrile composites attenuate gamma rays better than some traditional shielding materials [23, 24]. Thus, during this research the results determined using MCNPX and the XCOM database (study of the gamma ray protective properties of phthalonitrile/WB) were compared with the experimental results at the same energy sources (Cs-137 and Co-60).

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CONCLUSION

In this study, in the energy range of 661, 1172 and 1332 keV, the MCNPX code and the XCOM database were used to simulate and calculate the mass attenuation coefficients, transmittance Ln (I_0/I), and Half-Value-Layer, Ten-Value-Layer and radiation protection efficiency of the phthalonitrile/WB composites against gamma-rays. The WB concentrations were successively 10, 20, 30, 40, and 50 wt%. The MCNPX simulated and theoretical results of the mass attenuation coefficients were compared. Moreover, there was a satisfactory agreement between MCNPX simulations and the XCOM data. The investigation agrees well with other experimental results and includes more outstanding results at the same range of photon energy.

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