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Validating experimental data for attenuation coefficients of developed polymer composites in shielding applications through Monte Carlo simulation

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Abstract: The widespread use of composites in an ionizing environment raises durability concerns, particularly when performance is required over a longer period, in a radiation environment. The goal of this study was to measure the attenuation coefficient for the polymer composite developed for the radiation shielding application and validate it with the well-known Monte Carlo simulation. A radiation-hardened epoxy resin was developed in the laboratory by utilizing various compositions which is capable of being employed as a shielding material in a nuclear environment. To develop a radiation-hardened layer, to some extent, raised the density of the material, using suitable fillers. Polymer stabilizers were used single or in combination. The specimens were irradiated to determine the developed material's shielding properties. The prepared composites' radiation absorption is evaluated using an absorption coefficient. Experiments with gamma radiation using an Am Be Neutron source were conducted to determine the linear absorption coefficient. Monte Carlo simulation was utilized to predict the absorption coefficient for developed composites and the results were compared with experimental data.

Keywords: radiation hardened; polymer; shielding application; Monte-Carlo simulation; radiation

1 Introduction

1.1 Objective

The extensive application of composites in ionizing environments has brought about concerns regarding their durability, especially when long-term performance in a radiation-exposed ambience is required. Consequently, the objective of this study was to enhance the material's resilience by reducing the number of free charge carriers generated within it after radiation exposure, thus increasing its longevity [1].

The research endeavours aimed to develop radiation-resistant epoxy resin composites using diverse compositions and to produce a radiation-resistant polymeric shielding material suitable for use in a nuclear environment. The material's durability was improved by reducing the number of free charge carriers through crosslinking with the assistance of fillers and stabilizers, followed by subjecting it to radiation to assess any degradation in its properties. It's important to note that in this context, radiation was employed to assess the absorption capabilities of the developed material, rather than as a means to cure the material.

1.2 Linear absorption coefficient using Monte Carlo simulation

Monte Carlo simulation serves as a powerful method for replicating numerical processes, especially those involving the interactions of nuclear particles with materials. It offers significant advantages for tackling intricate problems that defy deterministic computer modelling. This approach involves sequentially simulating individual probable events that constitute a process, with these events governed by numerical representations of probability distributions. Given that achieving a fully representative portrayal of the phenomenon often demands a substantial number of trials, Monte Carlo simulations are typically conducted on digital computers [2–4].

Monte Carlo simulation emerges as a vital tool for obtaining critical material parameters, particularly when conducting experiments is unfeasible. With the increasing use of radiation across diverse fields, it becomes essential to ascertain a material's radiation shielding properties to determine its suitability for use in nuclear environments. Furthermore, researchers working with gamma or cosmic rays are increasingly concerned about their potential

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hazardous effects. An added benefit is that MCNP (Monte Carlo N-Particle) software comes preloaded with data for numerous conventional materials, simplifying the comparison between conventional and unconventional materials without the need for an extensive series of experiments, ultimately saving time.

However, in the context of this paper, Monte Carlo simulation is employed to compare experimental results with MCNP-generated results to validate the concurrence between virtual and practical outcomes. This validation aids in determining the suitability of MCNP for further experimental work.

Epoxy is a highly significant material used in the production of devices intended for nuclear environments. However, the process of formulating a robust shielding material requires determining the optimal composition. Therefore, when MCNP is employed to distinguish the combination of photons, neutrons, and electrons present in the material, simulated data may not provide an accurate prediction of the final material properties. To address this challenge, it is necessary to consider each atom within the material's composition to determine its precise molecular weight and subsequently input this data into MCNP for simulation. Consequently, the validation of results against experimental data becomes essential in ensuring the reliability and accuracy of the simulation outcomes.

1.3 Experiments with a nuclear source for shielding and calculating the linear absorption coefficient

The attenuation coefficient is a measure of how much the incident energy beam is weakened by the material it is passing through a substance or medium. A significant attenuation coefficient indicates that the beam is "attenuated" (weakened) quickly as it passes through the medium, whereas a small attenuation coefficient indicates that the material is relatively transparent to the beam. [5–8] In most cases, the words "attenuation" and "absorption" are interchangeable. Experiments on neutron detection were carried out in this study. The experiment on Epoxy polymer filled with three different fillers using an Am–Be neutron source with a flux of ~3×10^7 n/s [9–11].

2 Materials and methods

2.1 Materials

Epoxy resin was selected due to its commendable mechanical, thermal, and corrosion resistance attributes [12], and this research involved the creation of three unique composite materials, each using distinct fillers: graphite [13], lead [14], and Boron Nitride nano powder [15].

The fabrication of epoxy composite sheets employed the gravity casting method, which relies solely on the force of gravity, without the application of additional pressure [16]. It's important to note that the characteristics of these composites undergo continuous changes during the manufacturing process [17]. Therefore, achieving an even and consistent dispersion of fillers within the polymer resin is imperative to ensure optimal performance [18]. The dispersion of these fillers in various specimen types was assessed using scanning electron microscopy.

For achieving optimal results with irradiated specimens, the fundamental approach to guard against post-irradiation degradation involves employing stabilizer blends [19].

This study involved investigations on cured composites, including Epoxy/graphite powder, Epoxy/Lead powder, and Epoxy/Boron Nitride Nano powder, each blended with two distinct types of stabilizers. The materials employed in these experiments encompassed Bisphenol A-type Epoxy resin and Polyamide as the hardener. Stabilizers utilized were Hindered Amine Light Stabilizer (HALS) and Tri Vinyl Phosphate (TVP), while the composites incorporated fillers such as graphite, lead, and Boron Nitride Nano Powder (BNNP).

Four sets of samples were prepared using the following compositions;

(i) Pure epoxy resin, (ii) Epoxy/Graphite Powder, (iii) Epoxy/Lead powder, (iv) Epoxy/boron nitride nano powder.

Along with the above developed specimens, conventional radiation shielding materials, pure Lead, and tantalum specimens were also tested for comparison.

2.2 Methods

2.2.1 Monte Carlo simulation of the linear absorption coefficient: MCNP, which stands for Monte Carlo N-particle, is a versatile code designed for simulating the transport of various particles, including neutrons, photons, electrons, and combinations of these, such as neutron/photon/electron interactions. The algorithm is capable of handling complex three-dimensional arrangements of materials in geometric cells, utilizing first and second-degree surfaces, as well as fourth-degree circular tori to define boundaries. MCNP can be configured to simulate the transport of neutrons, photons, electrons, or their combinations, including:

(1) Neutrons only
(2) Photons only
(3) Electrons only
(4) Combined neutron and photon transport (including the creation of photons by neutron interactions)
(5) Combined neutron, photon, and electron transport
(6) Combined photon and electron transport

The energy ranges covered by MCNP for different particles are as follows:

– Neutron energy ranges from 10^{-11} MeV to 20 MeV, with certain isotopes extending up to 150 MeV.
– Photon energy spans from 1 keV to 100 GeV.
– Electron energy ranges from 1 keV to 1 GeV [20–22].

The weight fraction of each sample was filled in Monte Carlo simulation to predict the absorption coefficient for developed composites (Figures 1 and 2).

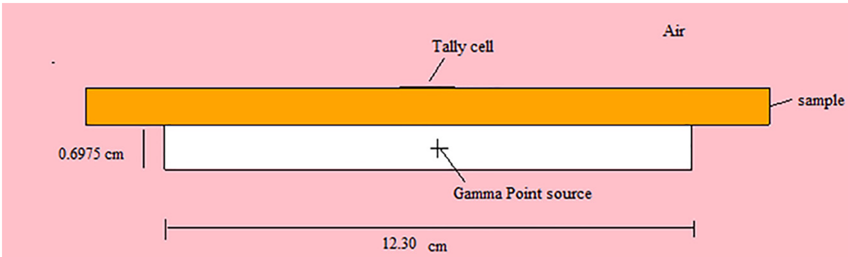

Figure 1: Gamma attenuation with MCNP simulation set-up.

The following data is used to generate gamma attenuation with MCNP simulation results:

The present activity of ^{60}Co source = 55 kci
Source Yield: 2.035E+15 (=55 kci) gamma/sec
Branching: 0.999826
Lambda: 2.97509E-06
Gamma energy: 1.332 keV

Using MCNP data cards, the Gamma Source was specified as a single energy 1.332 keV point source (although in an experimental set-up, there must be some Physical dimension of a source). Tally was calculated at the rear of the specimen material in a small volume to construct the Linear Coefficient Parameter, which measures the drop in intensity in units of Flux after adding the thickness of the test material.

The fractional drop, or attenuation, of the gamma-ray beam intensity in geometry per unit thickness of absorber, is defined by the following equation:

$$\mu = 1/t \ln (I_0/I) \tag{1}$$

The linear attenuation coefficient is defined as the chance of radiation interacting with a substance per unit travel length. This is how the mass attenuation coefficient is defined.

$$\mu/\rho = 1/\rho t \ln (I_0/I) \tag{2}$$

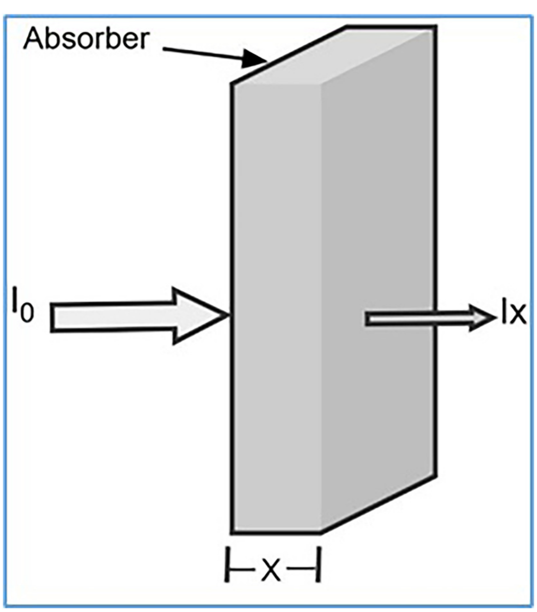

Figure 2: Attenuation of gamma radiation from absorbing material.

For each thickness variable, $\ln(I_0/I)$ values are calculated, where I_0 is the Gamma Flux calculated in a small volume without introducing the specimen, and I is the Gamma Flux calculated in a small volume after adding the variable thickness of the test specimen. Then, using $\ln(I_0/I)$ versus thickness in cm, the Gamma Linear Coefficient value in cm^{-1} for each specimen is calculated using equations (1) and (2) [23–25].

2.2.2 Gamma irradiation using an Am–Be source to test for shielding and estimate the linear absorption coefficient: The attenuation coefficient signifies the total reduction in intensity of a narrow beam, encompassing the impact of scattering, whereas the absorption coefficient quantifies the rate at which intensity diminishes due to absorption [26, 27].

Absorption occurs when high-frequency radiation traverses through materials, and as the thickness of the absorbing material increases, the radiation strength diminishes. Given the material's intended use for shielding, it becomes imperative to determine the Linear Absorption Coefficient for all generated specimens [28–30].

An experiment was conducted employing an Am–Be neutron source with an intensity of approximately 3×10^7 neutrons per second on Epoxy polymer laden with three distinct fillers, all with equivalent filler content. This experiment aimed to measure the linear absorption coefficient for neutrons. The Neutron activation technique was employed, which relies on detecting gamma rays emitted from a sample irradiated by neutrons and using Gold (Au) foils to detect transmitted neutrons.

In this technique, Au foil becomes activated due to neutron interaction, specifically through the $Au^{197}(n,\gamma)Au^{198}$ reaction, in which Au^{197}, when exposed to gamma rays, may convert to Au^{198}. These gamma rays are detected using a well-calibrated High Purity Germanium (HPGe) Detector, and the presence of induced 412 keV gammas on the activated foil is used to determine Au^{198} production for each specimen thickness, both with and without the specimen.

Neutron detection experiments were conducted using five specimens, each measuring 150 × 50 × 3 mm, and tightly clamped together without any intervening air gaps. Subsequently, during testing, one specimen was successively removed after each measurement, resulting in thicknesses of 12, 9, 6, and 3 mm, along with a measurement for the absence of the specimen (0 mm thickness).

The linear absorption coefficient was calculated by plotting $\ln(I_0/I)$ against the specimen thickness (X). The obtained graph provides a comparative assessment of the linear absorption coefficient (μ) for the various composite materials. The experimental setup for determining the absorption coefficient was employed.

The following expression is the mathematical expression for intensity (I).

$$\ln[I_0/I] = \mu \cdot X \tag{3}$$

where,

I_0 = Au^{198} produced without absorber material

I = Au^{198} produced with absorber material of thickness X

μ = linear absorption coefficient for the absorbing material

Eq. (3) was used to calculate the linear absorption coefficient. A straight-line plot between $\ln(I_0/I)$ and thickness X yields the linear absorption coefficient, and the slope of this graph gives the linear absorption coefficient. The relationship between the intensity, $\ln(I_0/I)$, and the thickness, X, is investigated. The linear absorption coefficient (μ) is determined by the slope of this graph.

Table 1: Value of $\ln(I_0/I)$ for gamma irradiation using Monte Carlo simulation.

Shielding material	Specimen thickness in mm				
	3	6	9	12	15
Epoxy	0.00	0.447	0.823	1.65	1.42
Epoxy/Gr	0.00489	0.447	0.823	1.14	1.39
Epoxy/Pb	0.00	0.447	0.823	1.14	1.42
Epoxy/Bn	0.00	0.447	0.823	1.14	1.42
Lead	0.0493	0.597	1.08	1.51	1.91
Tantalum	0.0687	0.651	1.17	1.65	2.1

3 Results and discussion

3.1 Monte Carlo simulation results for gamma radiation attenuation

For an in-depth analysis of shielding materials, please refer to Figure 3 and Table 1, which provide a comparison of $\ln(I_0/I)$ versus thickness. These results were obtained through the irradiation of epoxy and epoxy specimens loaded with fillers using gamma radiation and were compared with conventional materials such as Lead and Tantalum. The data was obtained using Monte Carlo simulation, which offers data for traditional shielding materials within the software.

It's noteworthy that pure epoxy exhibits a notably high absorption coefficient for gamma radiation. However, these findings suggest the potential for developing an epoxy-based polymer composite with a higher filler content. The values of these specimens were found to be comparable to those of conventional materials, indicating the promising potential of such composite materials in shielding applications.

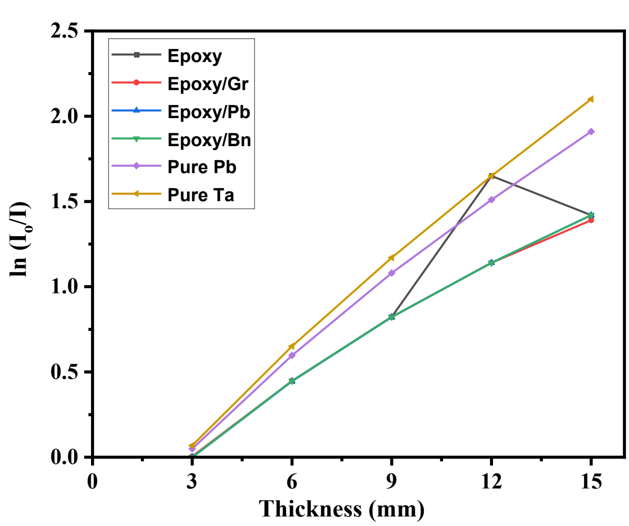

Figure 3: Comparison of absolute absorption by gamma irradiation using Monte Carlo simulation.

3.2 Using the experimental method, determine the effect of gamma radiation absorption

LAC was computed and compared to pure epoxy specimens based on the results of irradiation testing.

Table 2 and Figure 4 illustrate the linear attenuation coefficient values of the shielding material specimens. In this experiment, gamma particles were generated using a neutron source, with gamma particles representing the secondary radiation produced in the Am–Be source. The challenge in this context lies in constructing a protective barrier that effectively absorbs both neutron particles and secondary gamma radiation while providing shielding against neutron particles.

When used as a shield for neutron particles, lead exhibits an exceptionally low neutron absorption rate and virtually no secondary gamma radiation. In contrast, pure epoxy possesses a lower absorption coefficient for neutrons.

The analysis reveals that the linear absorption coefficient for gamma rays is notably higher compared to that for the neutron source. Furthermore, it is on par with the absorption characteristics of pure lead and pure tantalum materials.

According to the results obtained from the Monte Carlo simulation (MCNP), there is minimal variation in the

Table 2: The value of $\ln(I_0/I)$ for gamma irradiation, using the experimental technique.

Shielding material	Specimen thickness in mm				
	3	6	9	12	15
Epoxy	0.03659	0.079462	0.294236	0.360603	0.452677
Epoxy/Gr	0.021401	0.09798	0.356604	0.496892	0.65995
Epoxy/Pb	0.037631	0.098491	0.223681	0.31586	0.395671
Epoxy/Bn	0.23172	0.336167	0.44	0.552095	0.65939
Lead	0.0684	0.1442	0.221	0.33431	0.44
Tantalum	0.592639	0.975962	1.330946	1.654773	1.95112

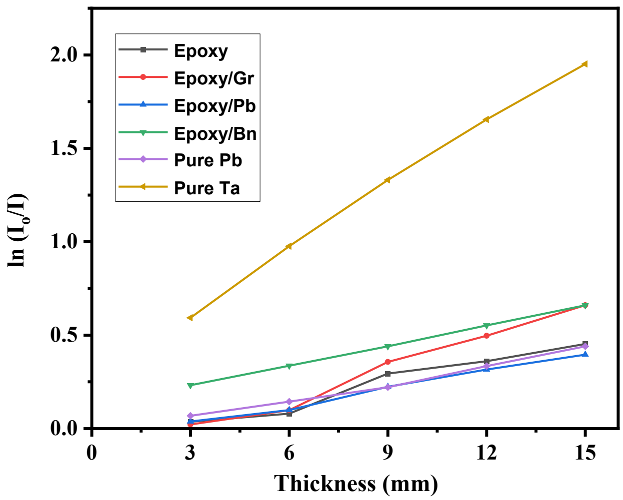

Figure 4: Comparison of intensity versus thickness for gamma irradiation.

absorption coefficient values across the developed samples. This observation could be attributed to the fact that all the samples are composed of the same material with the addition of small quantities of fillers and possess relatively thin thicknesses, ranging from 3 mm to 15 mm. Consequently, these samples exhibit minor variations in the $\ln(I_0/I)$ versus thickness curve, as the atomic weight changes are negligible when comparing one sample to another. Monte Carlo simulation calculates the absorption coefficient based on the differences in atomic weight changes.

It's worth noting that the study also highlights that pure epoxy itself exhibits favourable absorption properties for gamma radiations. These findings provide promising indications for the potential development of materials with higher filler content within epoxy-based polymer composites.

4 Conclusions

Table 2 and Figure 5 illustrate the observations that both tantalum and lead exhibit superior absorption coefficients compared to epoxy-based polymer composites. This suggests that epoxy itself may permit neutron passage, and since the composite fillers are present in relatively low proportions, their impact on neutron absorption is limited. Nonetheless, even with lower filler content, graphite has demonstrated more favourable results compared to pure epoxy, as well as epoxy with boron nitride Nanopowder and epoxy with lead filler, respectively. These findings offer encouragement for the development of materials with higher filler content in epoxy-based polymer composites (Table 3).

It is worth noting that the linear absorption coefficient values for the tested materials are significantly higher than those for the neutron source. These values are also comparable to those of pure lead and pure tantalum.

Upon comparing the experimental results with the Monte Carlo N-particle (MCNP) simulation, it was noted that the absorption coefficient values were higher in the simulated samples than in the experimental results, as indicated in Figure 4. This discrepancy could be attributed to the fact that all samples are composed of the same material mixed with fillers in relatively small quantities, and their thickness varies from 3 mm to 15 mm. Consequently, the absorption coefficient results exhibit minor variations due to negligible differences in the weight fraction of each sample relative to the others. The Monte Carlo simulation calculates the absorption coefficient based on the differences in atomic weight changes.

In conclusion, for samples with a lower proportion of fillers, the Monte Carlo simulator approximates nearly the same weight fraction for all samples, regardless of their different filler contents. Therefore, increasing the filler-to-

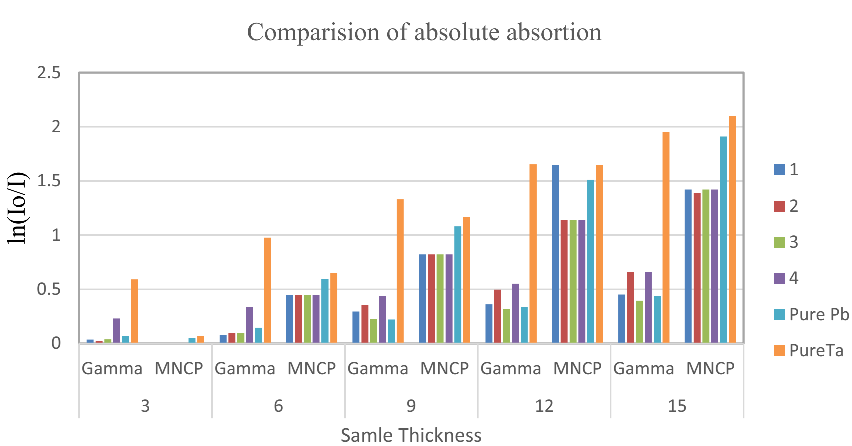

Figure 5: Comparison between Monte Carlo simulation and experimental methods for calculating the linear absorption coefficient.

Table 3: Comparison between Monte Carlo simulation and experimental methods for calculating linear absorption coefficient.

Shielding material	Sample thickness in mm									
	3		6		9		12		15	
	Gamma	MCNP	Gamma	MCNP	Gamma	MCNP	Gamma	MCNP	Gamma	MCNP
Epoxy	0.03659	0	0.07946	0.447	0.294236	0.823	0.3606	1.65	0.452677	1.42
Epoxy/Gr	0.021401	0.00489	0.09798	0.447	0.356604	0.823	0.49689	1.14	0.65995	1.39
Epoxy/Pb	0.037631	0	0.09849	0.446	0.223681	0.822	0.31586	1.14	0.395671	1.42
Epoxy/Bn	0.23172	0	0.33617	0.447	0.44	0.823	0.5521	1.14	0.65939	1.42
Lead	0.0684	0.0493	0.1442	0.597	0.221	1.08	0.334	1.51	0.44	1.91
Tantalum	0.592639	0.0687	0.97596	0.651	1.330946	1.17	1.65477	1.65	1.95112	2.1

resin ratio and ensuring significant differences in atomic weight could enable a more accurate comparison with experimental results and other conventional materials using Monte Carlo Simulation.

However, considering factors like weight and ease of fabrication, the material developed with graphite filler has the potential for modification to potentially replace pure lead and pure tantalum, provided that the composition is optimized accordingly.

Abbreviations used in the paper

BNN Powder	Boron Nitrided Nano Powder
^{60}Co	Cobalt 60
SEM	Scanning Electron Microscope
DMA	Dynamic Mechanical Analysis
TGA	Thermo Gravimetric analysis

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Author contributions: The authors have accepted responsibility for the entire content of this manuscript and approved its submission.
Competing interests: The authors states no conflict of interest.
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