Research article

Investigation of the gamma ray shielding properties for polyvinyl chloride reinforced with chalcocite and hematite minerals

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ABSTRACT

Polyvinyl chloride (PVC) is the most widely produced synthetic plastic polymer in the world; it has a variety of applications due to its low cost, elasticity, light weight, good mechanical characteristics and corrosion resistance. In order to protect living beings from harmful radiation such as gamma rays, novel low-cost chalcocite and hematite-based PVCs were fabricated for shielding purposes. The mass attenuation coefficient \( \mu_m \) for various fabricated hematite and chalcocite-based PVCs was calculated using MCNP-5 code. The results were compared with the values calculated theoretically using XCOM software between 0.015 and 15 MeV. Moreover, the simulated \( \mu_m \) parameter for chalcocite/PVC and hematite/PVC was used to calculate other shielding factors, such as the half value layer (HVL), the mean free path (MFP) effective atomic number \( Z_{eff} \), the geometric-progress (G-P) fitting parameters and the exposure buildup factor (EBF). The simulated data of \( \mu_m \) for all composites is comparable to that obtained from a theoretical calculation. The results showed that the addition of hematite and chalcocite enhance the \( \mu_m \) of PVC polymers. We also found that the \( \mu_m \) of chalcocite/PVC is higher than that of hematite/PVC due to the copper content in the former.

1. Introduction

Polyvinyl chloride (PVC) is a chlorinated hydrocarbon polymer originally produced by the USA and Germany in 1930. PVC has become an important material due to its low manufacturing cost, elasticity, light weight, and corrosion resistance. It is a solid white material formed during the polymerization of polyvinyl chloride monomers with a chemical composition of \( (C_2H_3Cl) \). The pure PVC polymers are insoluble in water, acids, and organic solvents, but they are slightly soluble in tetrahydrofuran at room temperature. Over the last few decades, PVC has become the world's mostly widely produced synthetic plastic polymer: it has a variety of applications due to its low cost, elasticity, light weight, good mechanical characteristics, and corrosion resistance [1, 2, 3].

Enhancement of the physico-mechanical properties of PVC polymers depends on the type of filler used. Numerous works have reported the enhancement effect of various filler materials on the properties of PVC. \( \text{BaTiO}_3/\text{NiO} \) were found to enhance the conductivity and dielectric constants of PVC [1], while \( \text{La}_{0.99} \text{Bi}_{0.01} \) \( \text{FeO}_3 \) nanoparticles were found to enhance the transition temperature, dielectric constant, and optical characteristics of the synthesized PVC [2]. In \( \text{ZnO}/\text{PVC} \) nanocomposite films, the glass transition temperature was found to increase with the addition of \( \text{ZnO} \) nanoparticles. This enhancement of the nanocomposite film's properties can facilitate the storage performance of a polymer battery [3]. Additionally, a few works have also reported enhancement of the strength of building materials; for example, the compressive stress of concrete can be advantageously modified by replacing some of the natural fine and coarse aggregates with PVC aggregates [4].

Today, radiation shielding is a very crucial component in radiation protection programs. It is utilized to optimize the dose of human radiation exposure in ionizing radiation practices (for example, radiation medicine, nuclear power plants, research accelerators and others). Conventionally, concrete is the most prevalent and commonly used material for radiation shielding in most facilities, such as hospitals. The cost effectiveness and vast available quantities of concrete are its main advantages as a shielding material [5, 6]. However, concrete has some disadvantages: cracks can occur after long periods of exposure to nuclear...
radiation and it is difficult to transport. Synthetic polymers can be utilized to fabricate new materials that can provide radiation shielding. Moreover, their other advantages (low manufacturing cost, durability, and high thermal and chemical stability) are among the favored traits and qualities for superior shielding materials. Recently, many works have reported various kinds of polymers, such as nylon-6, have novel shielding properties and can be used for protection against neutrons [7]. Moreover, other works have noted the effects of various fillers on the neutron shielding properties of polymers; for instance, the shielding properties of siloxane-based polymers were enhanced by the addition of boron particles [8]. Furthermore, the efficiency of epoxy resin was improved with molybdenum [9], barite [10] and a ferrochromium slag additive [11].

Hematite and chalcocite are two minerals rich in iron and copper, respectively. They are natural, cheap, abundant, and have suitable mass attenuation coefficients for low and high gamma rays [12]. In the present work, PVCs were synthesized and fabricated with chalcocite and hematite. The mass attenuation coefficient $\mu_m$ for five various polymer samples was calculated using MCNP-5 Code between 0.015 and 15 MeV. Gamma ray interaction and penetration were evaluated by calculating other shielding parameters such as the half value layer (HVL), the effective atomic number $Z_{\text{eff}}$, the effective electron density $N_{\text{el}}$, and the exposure buildup factor (EBF). In order to test the reliability of the results obtained from the MCNP simulation, the $\mu_m$ obtained by MCNP simulation code were compared to those calculated theoretically using the XCOM database.

2. Materials and methods

2.1. Theoretical aspect

Shielding materials can be characterized through their mass attenuation coefficient $\mu_m$. The theoretical calculation of $\mu_m$ for PVC, PVC/hematite and PVC/chalcocite can be reached via Eq. (1) [13]:

$$\mu_m \left( \frac{\text{cm}^2}{\text{g}} \right) = \sum_{i=1}^{n} \omega_i \left( \frac{\rho_i}{\mu_i} \right)$$

(1)

Where $\omega_i$ and $\left( \frac{\rho_i}{\mu_i} \right)$ are the fractional weight and partial mass attenuation coefficients, respectively, for the $i^{\text{th}}$ constituent element in multielement modified materials.

| Table 1. Densities and chemical composition of hematite and chalcocite-based PVC. |
|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| Chemical composition of hematite and chalcocite-based PVC (%) | PVC | PVC-H10 | PVC-H20 | PVC-H30 | PVC-C10 | PVC-C20 | PVC-C30 |
| SiO$_2$ | 0.000 | 0.093 | 0.186 | 0.279 | 0.009 | 0.018 | 0.027 |
| Fe$_2$O$_3$ | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 |
| Al$_2$O$_3$ | 0.000 | 0.016 | 0.032 | 0.048 | 0.000 | 0.000 | 0.000 |
| CaO | 0.000 | 0.009 | 0.018 | 0.027 | 0.000 | 0.000 | 0.000 |
| MgO | 0.000 | 0.101 | 0.202 | 0.303 | 0.000 | 0.000 | 0.000 |
| C$_2$H$_5$Cl | 99.900 | 89.910 | 79.920 | 69.930 | 89.910 | 79.920 | 69.930 |
| TiO$_2$ | 0.000 | 0.004 | 0.008 | 0.012 | 0.000 | 0.000 | 0.000 |
| FeO | 0.000 | 8.853 | 17.706 | 26.559 | 0.000 | 0.000 | 0.000 |
| Cu | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 7.967 | 15.934 |
| S | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 2.016 | 4.032 |
| Density (g/cm$^3$) | 1.192 | 1.649 | 2.102 | 2.551 | 1.830 | 2.467 | 3.105 |

Figure 1. Screen shot for MCNP geometry.
The mean free path (MFP) is the shielding parameter which describes the distance that gamma ray photons travel inside the shielding material between two successive collisions. The half value layer (HVL) is the thickness required to decrease gamma ray intensity to half of its initial value (HVL). The MFP and HVL can be described by Eqs. (2) and (3):

\[
MFP \ (cm) = \frac{1}{\mu \ (cm^{-1})}
\]

\[
HVL \ (cm) = 0.693 \frac{1}{\mu \ (cm^{-1})}
\]

The effective atomic number \(Z_{eff}\) is an important factor in shielding materials: it describes any multi-element shielding material in terms of its equivalent element and is defined through the following equation [14]:

\[
Z_{eff} = \frac{\sum f_i A_i (\mu_{m1})}{\sum \mu_{m1}}
\]

Where \(f_i\), \(A_i\) and \(Z_i\) refer to the fractional abundance, atomic weight and atomic number of the \(i^{th}\) constituent element, respectively.

Effective electron density \(N_{eff}\) is also a factor in radiation shielding and is defined through Eq. (5):

\[
N_{eff} = \frac{N_i}{M} Z_{eff} \sum n_i
\]

Where \(M\) is the atomic mass of the material.

### 2.2. PVC preparation

Hematite and chalcocite-based PVC mixtures were fabricated to produce a new material that possesses superiority shielding properties. A commercial-grade PVC was melted at a temperature of 250 °C and then reinforced with multiple ratio contents of hematite and chalcocite minerals. The mixtures were prepared in 2 different types of polymer for different fillers of hematite and chalcocite. They were labeled as PVC-H10, PVC-H20, and PVC-H30 for hematite ratios of 10, 20, and 30%, respectively, while the other series was labeled as PVC-C10, PVC-C20, and PVC-C30 for chalcocite ratios of 10, 20, and 30%, respectively. The ratio of hematite and chalcocite cannot exceed 30% if the elasticity of the PVC composites is to be maintained.

X-ray fluorescence (XRF) was used to determine the chemical composition of the fabricated samples while the density of the fabricated samples was measured using the Archimedes method. The compositions and densities are listed in Table 1.

MCNP is a radiation transport code developed by created by the Los Alamos National Laboratory (LANL). It is used to simulate the transport of electrons, neutrons, gamma, and X-rays using the Monte Carlo method [15]. The MCNP input file required accurate information about the geometry, source (SDEF card), and composition (material card) to execute a MCNP simulation. The geometry of the simulation was designed according to an arbitrary 3D setup, as illustrated in Figure 1. As shown in Figure 1, all equipment has been considered according to experimental facilities. The radioactive source was placed inside a lead collimator with a slit diameter of 2 cm. The source was placed at a distance of 10 cm from the detector. The samples were fabricated as a disk with a diameter of 5 cm placed between the source and the detector at a distance of 5 cm from the source. The detector was set up to be an F2 tally in order estimate the track length of the incident gamma ray. The simulation geometry shielded the outer space using 5cm of lead. The simulation was carried out using NPS card = 10^8 particles. The relative error estimated from the MCNP simulation is less than 1%.

### 3. Results and discussion

The simulations of the irradiation of the PVC, PVC/hematite, and PVC/chalcocite samples were carried out using MCNP radiation transport code and monoenergetic photon sources of 0.015 and 15 MeV to calculate the \(\mu_m\) of the samples. Figure 2 illustrates that the \(\mu_m\) for all the samples tends to peak at values of lower energy (i.e. 0.015 MeV) due to the photoelectric cross section, which is largely present in the low energy region. According to Table 2, the maximum \(\mu_m\) of all composites varied between 10.456 and 25.974 cm^2/g for PVC and PVC-C30 composites, respectively. The variation of the maximum \(\mu_m\) reveals the dependence of \(\mu_m\) on the composition of the materials.

Figure 2 also reveals that the \(\mu_m\) in intermediate energy (i.e. 0.06 < E < 2 MeV) has low variation with the incident gamma ray energy due to the Compton scattering that dominates in this energy interval. In addition, the \(\mu_m\) of all the composites is almost constant for high energies (i.e. E > 3 MeV) due to the pair production interaction in which the interaction cross section mainly depends on the energy of the incident gamma ray. It is clear from Figure 2 that the simulated \(\mu_m\) of the PVC polymer is enhanced due to the addition of chalcocite and hematite minerals.

It can also be seen that the \(\mu_m\) of chalcocite/PVC is higher than that of hematite/PVC due to the copper content in the former, which has a higher \(\mu_m\) than the iron content in hematite/PVC. The simulated \(\mu_m\) of the prepared PVC, PVC/hematite, and PVC/chalcocite compared with some commercial shielding materials such as ordinary concrete [16], zinc bismuth borate glass (10ZnO; 30Bi2O3;60B2O3) [17], and RS-520(SF6) glass [18]. The comparison showed that our studied samples have a \(\mu_m\) higher than concrete, while they have \(\mu_m\) lower than zinc bismuth borate glass and RS-520 glass (at low and intermediate energy). At high gamma ray energy (E > 3 MeV), the \(\mu_m\) for all studied samples and standard shielding materials are comparable. The theoretical \(\mu_m\) was computed using the XCOM database. The simulated results of the \(\mu_m\) for PVC, PVC/hematite, and PVC/chalcocite were comparable to those calculated using XCOM, which indicates the reliability of the values for the \(\mu_m\) showed in Table 2. The difference in percentage between the simulated and calculated \(\mu_m\) for all composites was found to be less than 1%.

Better shielding materials show thinner layers of HVL and MFP [19]. The energy dependence of MFP for hematite/PVC and chalcocite/PVC is illustrated in Figure 3. It is clear that the MFP of all composites tends to peak at high energies (i.e. 15 MeV) and varies between 13.101 and 38.357 cm for PVC and PVC-C30 respectively, while the MFP tends to be...
at its lowest for all composites at low energies (i.e. 0.015 MeV) and varies between 0.0123 cm and 0.0802 cm for PVC-C30 and PVC respectively. In the low energy region \((0.015 < E < 0.06 \text{ MeV})\), the MFP tends to be at its lowest for all composites and also varies slightly with the incident energy due to the photoelectric interaction in which the interaction cross section is mainly proportional to \(Z^4/C_0^5\) [20, 21, 22]. The MFP increases gradually for all composites with the increase of the incident gamma ray energy \((for \ 0.06 < E < 3 \text{ MeV})\) due to the Compton effect in which the interaction cross section is directly proportional to the incident gamma ray energy. Finally, at the high energy region \((for \ E > 3 \text{ MeV})\) in which the pair production interaction domains, the MFP increases rapidly with the energy increase. According to the previous discussion the sample

| Energy (MeV) | Mass attenuation coefficient (cm²/g) |
|-------------|--------------------------------------|
| PVC-MCNP   | XCOM                                |
| PVC-H10-MCNP | XCOM                                |
| PVC-H20-MCNP | XCOM                                |
| PVC-H30-MCNP | XCOM                                |
| PVC-C10-MCNP | XCOM                                |
| PVC-C20-MCNP | XCOM                                |
| PVC-C30-MCNP | XCOM                                |
| 0.015       | 10.456                              |
| 0.02        | 4.582                               |
| 0.03        | 1.492                               |
| 0.04        | 0.731                               |
| 0.05        | 0.456                               |
| 0.06        | 0.333                               |
| 0.08        | 0.230                               |
| 0.1         | 0.189                               |
| 0.15        | 0.148                               |
| 0.2         | 0.131                               |
| 0.3         | 0.111                               |
| 0.4         | 0.099                               |
| 0.5         | 0.090                               |
| 0.6         | 0.083                               |
| 0.662       | 0.079                               |
| 0.8         | 0.073                               |
| 1          | 0.065                               |
| 1.173       | 0.060                               |
| 1.332       | 0.056                               |
| 1.5        | 0.053                               |
| 2          | 0.046                               |
| 3          | 0.037                               |
| 4          | 0.033                               |
| 5          | 0.030                               |
| 6          | 0.027                               |
| 8          | 0.025                               |
| 10         | 0.024                               |
| 15         | 0.022                               |

Table 2. Comparison between simulated and calculated \(\mu_{\text{m}}\).

Figure 3. Comparison between the MFP of PVC/Hematite, PVC/Chalcocite and some commercial shielding materials.

Figure 4. Th variation of HVL with energy for hematite-based PVC, chalcocite-based PVC and other commercial shielding materials.
PVC-C30 is considered the most effective shielding material in our study due to its low MFP. The MFP of PVC-C30 varied between 0.0123 and 13.101 cm and found to be lower than the ordinary concrete which varied between 0.0614 and 20.489 cm between 0.015 and 15 MeV respectively. The MFP of PVC-C30 found to be higher than the heavy metal oxides and glass which varied between 0.006-8.262 cm and 0.003–4.687 cm respectively.

The mechanism in which the HVL varied with the incident gamma ray energy is illustrated in Figure 4. Figure 4 reveals that the HVL values for all the prepared composites can be described in the similar manner as the MFP. However, the maximum HVL varied between 9.080 and 26.599 cm for PVC- C30 and PVC respectively, and the lowest HVL varied between 0.008 and 0.055 cm for the same composites. The HVL of PVC-C30 found to be lower than the HVL of ordinary concrete which varied between 0.0425 and 14.199 cm while, it is higher than the HVL of heavy metal oxides and glass RS-520 which varied between 0.004-5.726cm and 0.002-3.248 cm respectively.

The effective atomic number $Z_{\text{eff}}$ is required to describe the shielding parameters of a multi-element composite as its equivalent element. The energy dependence of the $Z_{\text{eff}}$ is illustrated in Figure 5 (a and b). The $Z_{\text{eff}}$ tends to peak at values for composites of low energy (for $E = 0.015$ MeV) because of the dominance of the photoelectric effect, and varies between 16.67 and 25.2 for PVC and PVC-C30 respectively. On the other hand, the minimum values were found at intermediate energies ($0.2 < E < 3$ MeV) and varied between 12.01 and 16.34 for PVC and PVC-C30 respectively. In addition, it can be observed that the $Z_{\text{eff}}$ is nearly constant for all composites in the intermediate energy region ($0.2 < E < 3$ MeV) due to the Compton effect. The $z_{\text{eff}}$ increased slowly with the increase of the incident gamma ray energy ($E > 3$ MeV) due to the pair production interaction in which the cross section in directly proportional to $(\log E)$ [23, 24, 25].

It can also be observed that the $Z_{\text{eff}}$ for PVC increased with the addition of hematite and chalcocite, while chalcocite/PVC have higher $Z_{\text{eff}}$ than PVC and hematite/PVC composites due to the copper contents, which have a high efficiency in gamma ray shielding.

The energy dependence of the effective electron density $N_{\text{eff}}$ is shown in Figure 6 (a and b), which reveals that the variation of $N_{\text{eff}}$ with the incident gamma ray energy is similar to the variation of $Z_{\text{eff}}$, and as a result, it can be described in a similar manner. The highest $N_{\text{eff}}$ was obtained in the energy value of 0.015 MeV and varied between $4.19 \times 10^{23}$ and $4.77 \times 10^{23}$ (electron/g) for PVC and PVC-H10 respectively, while...
Figure 8. The variation of EBF with gamma ray energy for PVC, hematite/PVC and chalcocite/PVC composites.
the lowest \( N_{\text{eff}} \) was achieved at energy of 1.332 MeV and varied between \( 2.95 \times 10^{23} \) and \( 3.07 \times 10^{23} \) (electron/g) for PVC-C30 and PVC–H10. Furthermore, it can be observed that in the intermediate energy region (0.3 < \( E < 3 \) MeV) the addition of hematite increases the \( N_{\text{eff}} \) of the PVC, while the addition of chalcocite decreases the \( N_{\text{eff}} \) of the PVC.

The best shielding material is that which have high equivalent atomic number. The equivalent atomic number \( Z_{\text{eq}} \) for PVC, hematite/PVC and chalcocite/PVC was calculated between 0.015 and 15 MeV and illustrated in Figure 7. It is clear that for all composites the \( Z_{\text{eq}} \) tends to maximum values varied between 14.43 and 50.52 for PVC and PVC-C30 respectively at energy between 0.8 and 1 MeV while, it tends to minimum values at high energy. For gamma ray energy between 0.015 and 1 MeV there are a small variation of \( Z_{\text{eq}} \) with the incident energy due to Compton scattering which is domains in the intermediate gamma ray energy. In the other hand, the \( Z_{\text{eq}} \) is rapidly decrease for energy \( E > 1 \) MeV due to the pair production interaction. Furthermore, the G-P fitting parameters for PVC, hematite/PVC and chalcocite/PVC were calculated and listed in supplementary data Table S1.

The exposure buildup factor EBF of various prepared PVC composites calculated using geometric-progression fitting parameters (G-P fitting parameters) between 0.015 and 15 MeV and presented in Figure 8. It is clear that, EBF tends to maximum for all composite's values at penetrating depth 40 mfp while, it tends to minimum values at penetrating depth 0.5 mfp. Moreover, the EBF of PVC, hematite/PVC and chalcocite/ PVC increase with increasing the penetration depth. The highest EBF for all composites is achieved in the intermediate energy (i.e. \( 0.3 < E < 0.5 \) MeV) due to the Compton scattering domination. The highest value of the EBF is obtained for PVC composites and varied between 1.6 and 822 at 0.5 and 40 mfp respectively, while the lowest EBF is obtained for PVC-C30 and varied between 1.54 and 315 at 0.5 and 40 mfp respectively. Figure 8 reveals to the additive of chalcocite and hematite decrease the EBF.

4. Conclusion

The shielding parameters of PVC, hematite/PVC and chalcocite/PVC composites are studied between (0.015 < E < 15 MeV) using MCNP code. The simulated results of \( \mu_{\text{eff}} \) for all composites showed their dependence on the incident gamma ray energy. The highest \( \mu_{\text{eff}} \) obtained for PVC-C30 composite and varied between 25.98 and 0.024 cm\(^2\)/g, while the lowest \( \mu_{\text{eff}} \) obtained for the PVC has values between 10.45 and 0.022 cm\(^2\)/g and varied between 0.015 and 15 MeV respectively. The shielding parameters of the fabricated PVC compared to some commercial shielding materials. The comparison showed that the shielding parameters of PVC materials are better than those of ordinary concrete while, they are less than the shielding parameters of zinc bismuth borate glass and RS-520 glass. Furthermore, the simulated data of \( \mu_{\text{eff}} \) are close to those calculated using XCOM for all composites. The study showed that the addition of hematite and chalcocite enhance the mass attenuation coefficient of PVC polymers. But chalcocite/PVC have a higher mass attenuation coefficient than hematite/PVC composites. Moreover, the results showed that a thin layer of chalcocite/PVC composite was sufficient to shield the incident gamma ray at various energies, compared to hematite/PVC. The obtained results showed that chalcocite/PVC and hematite/PVC composites have adequate shielding properties and they will be useful in various shielding applications.

Declarations

**Author contribution statement**

K. A. Mahmoud: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

E. Lacomme: Analyzed and interpreted the data; Wrote the paper.

M. I. Sayyed: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

O. F. Ozpolat: Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

O. L. Tashlykov: Conceived and designed the experiments; Performed the experiments; Contributed reagents, materials, analysis tools or data.

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**Competing interest statement**

The authors declare no conflict of interest.

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