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Modified halloysite minerals for radiation shielding purposes

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**ABSTRACT**
Halloysite is a clay mineral found in the natural environment and it has many applications in chemistry and medical fields. The linear attenuation coefficient (\(\mu\)) for halloysite nanotubes minerals simulated using MCNP 5 code between 0.015 and 15 MeV. Moreover, the linear attenuation coefficient calculated for all selected samples using Phys-X/PDS program. Furthermore, some important shielding parameters that described the penetration and interaction of gamma rays with halloysite atoms, such as the half-value (HVL), effective atomic number (Z\(_{eff}\)), and exposure buildup factor (EBF) were calculated for gamma rays between 0.015 and 15 MeV.

**1. Introduction**
Recently, due to the development of radiation isotopes and its applications in many fields, numerous works reported the shielding properties of distinct mineral materials such as glass (Mahmoud & Rammah, 2019; Sayyed et al., 2018b), concretes (Mahmoud, Sayyed, & Tashlykov, 2019a; Mahmoud, Tashlykov, El Wakil, & El Aassy, forthcoming), alloys (Agar, Sayyed, Akman, Tekin, & Kaçal, 2018), rocks and natural materials (Abu El-soad, Sayyed, Mahmoud, Erdem, & Kovaleva, 2019; Mahmoud et al., 2019a; Obaid, Sayyed, Gaikwad, & Pawar, 2018; Oto, Yildiz, Akdemir, & Kavaz, 2015).

Due to the low cost, and abundance of natural rocks and minerals, we can use it directly or as aggregates in the radiation shielding fabrication. Many works studied the shielding properties of various rocks and natural minerals (Mahmoud, Sayyed, & Tashlykov, 2019b; Mavi, 2012; Obaid et al., 2018). The works concluded that granites, marbles, and basalt have the best shielding characteristics.

Halloysite are white clay minerals with nanoscale particles discovered by Berthier in 1823. Halloysite is the clay mineral of kaolin group with an average density 2.5 g/cm\(^3\) and a chemical formula of Al\(_2\)Si\(_4\)O\(_{10}\)(OH)\(_4\)·n·(H\(_2\)O)(Yuan et al., 2015).

In the last few decades, great attention has been directed to halloysite minerals due to their nanoscale particles. Numerous works reported the technological applications of halloysite clay minerals, such as antibacterial and colon cancer drug delivery (Prashantha, Lacrampe, & Krawczak, 2013), templates to form hollow porous Au nanotubes (Zieba, Huezo, Arruebo, Martinez, & Santamaria, 2014), the ability to enhance the optical and mechanical properties of polyimide (Prashantha et al., 2013), electrical and magnetic properties enhancement (Rozynek, Zacher, Janek, Čaplovičová, & Fossum, 2013; Zhang & Yang, 2012), and thermal energy storage applications (Sarkar & Bhattacharyya, 2012). Up until now, there are no applications for halloysite clay minerals in radiation shielding technology.

The present work aims to study the shielding parameters for five halloysite nanotube minerals to be used as concrete aggregates and building materials. Besides, the linear attenuation coefficient of the selected halloysite samples simulated using MCNP-5 code and also calculated using Phys-X/PSD program between 0.015 and 15 MeV. Furthermore, the shielding factors which describe the penetration and interaction of gamma radiation with the halloysite particles such as the HVL, effective atomic number \(Z_{\text{eff}}\) were calculated between 0.015 and 15 MeV. The Geometric-progress fitting method was used to calculate the exposure buildup factor EBF and the G-P fitting parameters for the halloysite minerals.

**2. Materials and methods**

**2.1. Sample preparation**

In the present work, five halloysite nanotube minerals were modified in order to be used in chemical extraction. The modification was performed using 1.365 g of halloysite nanotubes (HNTs) mixed in 20 ml of toluene and was stirred for 10 min, then 3.085 mmol of a silane coupling agent was added. The mixture was stirred at 25°C for 30 min, then refluxed for 4 h. The modified Halloysite nanotubes were filtered and washed with 20 ml of ethanol four times. The final composites were identified as HNT1, HNT2 HNT3, HNT4 HNT5.
chemical composition of the halloysite modified nanotubes measured using the inductively coupled plasma optical emission spectrometer (ICP-OES). The samples were introduced to the ICP-OES through a system and operating parameters described in ref (Nagdy, El, Wakil, El, & Gaber, 2016). The chemical composition and density of the halloysite samples were measured and listed in Table 1.

### Table 1. The chemical concentration in wt (%) for halloysite-modified nanotube.

| Element | HNT1 | HNT2 | HNT3 | HNT4 | HNT5 |
|---------|------|------|------|------|------|
| Al₂O₃   | 39.50| 32.57| 21.24| 30.76| 31.33|
| CaO     | 0.00 | 0.38 | 0.25 | 0.35 | 0.36 |
| Fe₂O₃   | 0.00 | 0.33 | 0.22 | 0.30 | 0.32 |
| MgO     | 0.00 | 0.04 | 0.02 | 0.03 | 0.40 |
| NiO     | 0.00 | 0.01 | 0.01 | 0.02 | 0.02 |
| SO₂     | 0.00 | 8.80 | 0.10 | 0.10 | 0.10 |
| SiO₂    | 46.50| 42.65| 37.73| 47.89| 54.44|
| SrO     | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| TiO₂    | 0.00 | 0.03 | 0.02 | 0.03 | 0.31 |
| H₂O     | 14.00| 11.20| 14.00| 20.50| 12.70|
| Density (g/cm³) | 1.87 | 1.87 | 1.33 | 1.75 | 1.87 |

2.2. MCNP simulation

Monte Carlo N-particle Transport code MCNP is a general and effective code for simulating the shielding factors of various shielding materials, such as concretes (Mahmoud et al., 2019a), glasses (Mahmoud & Rammah, 2019; Sayyed, Dong, Tekin, Lakshminarayana, & Mahdi, 2018a), polymers (Issa, Mostafa, Hanafy, Dong, & Xue, 2019) and building materials (Huang et al., 2014). The shielding properties were halloysite nanotube samples at energy ranges between 0.015 and 15 MeV using a created input file. The created input file contained all required information about the prepared samples, gamma-ray sources, detector, and the system geometry. In this study, the system geometry consists of an evacuated cylinder with a thickness 5 cm, made of lead, and used to shield the system from the surrounding environment as illustrated in Figure 1. The inner system (inside the shielding cylinder) was filled with air. A collimator of lead with a diameter of 10 cm and a slit of 2 cm was placed at the center of the system. Moreover, each prepared sample was placed with a monoenergetic gamma-ray source inside the collimator. The simulation process was run out using NPS card 10⁶ particle.

2.3. Theoretical calculation

The linear attenuation coefficient is the parameter which describes the ability of the material to resist the incident gamma ray and can be described by Beer’s law. (Mahmoud et al., 2019a).

\[
\mu (cm^{-1}) = \frac{1}{x} \ln \left( \frac{I}{I_0} \right)
\]  

(1)

The mass attenuation coefficient of the shielding material can be calculated theoretically through equation (2).

\[
\mu_m \left( \frac{cm^2}{g} \right) = \sum_{i=0}^{n} \omega_i \left( \frac{\mu_i}{\rho} \right)
\]

(2)

where \( \omega_i \left( \frac{\rho}{\rho} \right) \) are the fractional weight and the partial mass attenuation coefficient for the \( i^{th} \) constituent element in the multielement modified materials.
Half value layer (HVL) is one of the most important shielding parameters which describe the thickness that reduce the gamma-ray intensity to a half of its incident intensity and it is described by equation (2) (Agar et al., 2019).

$$MFP(cm) = \frac{1}{\mu(cm^{-1})}$$  \hspace{1cm} (3)  

$$HVL(cm) = \frac{\ln(2)}{\mu(cm^{-1})}$$  \hspace{1cm} (4)  

The effective atomic number ($Z_{eff}$) is considered an important factor that is characterized in shielding materials and describes any multi-element shielding material in terms of its equivalent element and is defined through the next equation.

$$Z_{eff} = \frac{\sigma_a}{\sigma_e}$$  \hspace{1cm} (5)  

When gamma rays penetrate a shielding material, beyond the shielding materials there are two different kinds of radiation collided photons and uncollided photons. The buildup factor is defined as the ratio of the total number of particles at a given point to the number of uncollided particles, at that same point. The EBF can be calculated by using G-P method which is adopted by American National Standard ANSI/ANS-6.4.3–1991 as the best method to evaluate EBF of composite materials. EBF of any material can be evaluated in three steps (Mahmoud et al., 2019b):

(i) 1 Calculation of $Z_{eq}$

$$Z_{eq} = \frac{Z_1(\log R_2 - \log R) + Z_2(\log R - \log R_1)}{\log R_2 - \log R_1}$$  \hspace{1cm} (6)  

where $R$ is the ratio of Compton scattering mass attenuation coefficient to the total mass attenuation coefficient of the shielding material, $Z_1$ and $Z_2$ are the atomic numbers related to $R_1$ and $R_2$, respectively.

(ii) 1 Calculation of fitting parameters.

$$C = \frac{C_1(\log Z_2 - \log Z_{eff}) + C_2(\log Z_{eff} - \log Z_1)}{\log Z_2 - \log Z_1}$$  \hspace{1cm} (7)  

where $C_1$ and $C_2$ are the G-P fitting parameters corresponding to the atomic numbers $Z_1$ and $Z_{eff}$, respectively.

(iii) 1 Calculation of the exposure build up factor EBF

$$B(E, x) = 1 + \left(\frac{bC - 1}{(K^* - 1)}\right) (K^* - 1), K \neq 1$$  \hspace{1cm} (8)  

$$B(E, x) = 1 + (b - 1)x, K = 1$$  \hspace{1cm} (9)  

where $x$ refers to the distance between the detector and the radioactive source in unit of mfp, $E$ refers to the incident energy, $K(E, x)$ is the dose multiplicative factor and ($X_i$, b, a and d) are the G-P fitting parameters.

3. Results and discussion

The linear attenuation coefficient of some halloysite modified samples were simulated using MCNP 5 code and calculated theoretically using program Phys-X/PSD (Erdem, Özgür, Al, Sayyed, & Kurudirek, Forthcoming) for gamma-ray energies between 0.015 and 15 MeV. Figure 2 depicts that the simulated and calculated results are closed together. It is also clear that the linear attenuation coefficient of all selected halloysite samples depended on the incident gamma-ray energy. Moreover, according to Table 2, the linear attenuation coefficient reaches maximum values for low gamma-ray energies (i.e. 0.015 MeV) and varies between 6.715 and 9.854 cm$^{-1}$ for halloysite samples HNT 3 and HNT 2, respectively, while the minimum values of the linear attenuation coefficient were obtained for high energies (i.e. 15 MeV) and varied between 0.028 and 0.039 cm$^{-1}$ for halloysite samples HNT 3 and HNT 2, respectively. Figure 2 and Table 2 depict that there is a small variation between the simulated data of halloysite samples due to the small variation in the chemical composition between the selected halloysite samples. It is also clear that at low energies (0.015 < E < 0.04 MeV) the linear attenuation coefficient of the selected halloysite samples decreases rapidly with increased gamma-ray energy due to the photoelectric cross-section. In the intermediate energy region with energies between 0.04 and 3 MeV, the linear attenuation coefficient of all selected halloysite samples decreases gradually with increased gamma-ray energy due to the Compton scattering cross section. For high gamma-ray energy range (i.e. E > 3 MeV), the linear attenuation coefficient increases slowly with the incident gamma-ray energy due to the pair production cross section. Which is proportional to log E (Mahmoud et al., 2019b; Oto et al., 2015).

The half-value (HVL) in the present work is used to describe the thickness of halloysite samples required to reduce the gamma radiation to half of its incident values. Figure 3 reveals that the variation of the half-value of the selected samples with incident energies between 0.015 and 15 MeV. It is clear that from Figure 3, the HVL tends to reach maximum values for all samples at high energy (i.e. E = 15 MeV) and varies between 16.637 and 25.186 cm for samples HNT2 and HNT 3, respectively, while the HVL tends to reach minimum values at low energy (i.e. E = 0.015 MeV) and varies.
between 0.0703 and 0.1032 cm also for samples HNT2 and HNT3, respectively. For gamma rays with a low energy range (i.e. $0.015 < E < 0.04$ MeV) the HVL increases rapidly with the energy increase due to the photoelectric cross section is proportional to $E^{-3.5}$. For gamma-ray energy between $(0.04 < E < 3$ MeV), the HVL increases gradually with the energy increase due to Compton scattering cross section which is proportional to $E$. For high gamma-ray energies (i.e. $E > 4$ MeV) the HVL increases slowly with the energy increase due to the pair production cross section which proportional to $\log E$ (Sayyed, Tekin, Kilicoglu, Agar, & Zaid, 2018c). Moreover, the half-value layers calculated for the selected halloysite samples compared to those for a standard radiation shielding concrete (Bashter, 1997). Figure 3 illustrates that the best HVL is obtained for the standard heavyweight concrete and varied between 0.01336 and 14.1990 cm while the best HVL in the present study is obtained for HNT 2 and varied between 0.0703 and 16.637 cm at gamma-ray energy between 0.015 and 15 MeV, respectively.

Figure 4 depicts the dependence of the $Z_{\text{eff}}$ on gamma-ray energy. It is clear that the $Z_{\text{eff}}$ tends to reach maximum values at low energy regions (i.e. $0.015$ MeV), which varied between 11.639 and 12.245 for samples HNT4 and HNT2, respectively, while the minimum $Z_{\text{eff}}$ was obtained at intermediate energies (for $E = 1$ MeV) and varied between 6.586 and 7.959 also for...
samples HNT4 and HNT2, respectively. The $Z_{\text{eff}}$ tends to reach maximum values at low energies (for $0.015 < E < 0.04$ MeV) due to the photoelectric cross section, while in the intermediate energy region (for $0.04 < E < 3$ MeV) the $Z_{\text{eff}}$ decreases with the energy increase. The minimum values of the effective atomic number achieved for intermediate energy regions due to the Compton cross section. On the other hand, $Z_{\text{eff}}$ slowly increases with increased incident energy (for energy $E > 4$ MeV) due to the pair production cross section (Sayyed et al., 2018a; Sayyed et al., 2018b; Sayyed et al., 2018c).

The exposure buildup factor was also calculated for the selected samples between 0.5 and 40 penetration depth in units of $mfp$ and illustrated in Figure 5. It is clear that the exposure buildup factor has low values for low and high gamma-ray energy due to the photoelectric and pair production effects. For low gamma-ray energy, the photons are totally absorbed in the material without formation secondary photons due to the photoelectric cross section which proportional to $Z^4-5$. In intermediate gamma-ray energy ($0.04 < E < 3$ MeV), the buildup factor increases with increase the incident energy due to Compton scattering in which the photon energy is not

### Table 2. Comparison between the linear attenuation coefficient simulated using MCNP-5 code and calculated using Phys-X/PSD.

| Energy (MeV) | HNT1 MCNP | Phy-X/PDS | HNT2 MCNP | Phy-X/PDS | HNT3 MCNP | Phy-X/PDS | HNT4 MCNP | Phy-X/PDS | HNT5 MCNP | Phy-X/PDS |
|-------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| 0.015       | 9.231     | 9.227     | 9.854     | 9.654     | 6.715     | 4.891     | 8.903     | 8.729     | 9.692     | 8.901     |
| 0.02        | 4.072     | 4.068     | 4.349     | 4.259     | 2.970     | 2.164     | 3.933     | 3.870     | 4.273     | 3.929     |
| 0.03        | 1.429     | 1.427     | 1.514     | 1.478     | 1.044     | 0.761     | 1.377     | 1.371     | 1.489     | 1.368     |
| 0.04        | 0.788     | 0.785     | 0.824     | 0.799     | 0.574     | 0.418     | 0.755     | 0.761     | 0.812     | 0.743     |
| 0.05        | 0.555     | 0.553     | 0.573     | 0.554     | 0.404     | 0.294     | 0.529     | 0.540     | 0.567     | 0.517     |
| 0.06        | 0.447     | 0.446     | 0.458     | 0.441     | 0.325     | 0.237     | 0.425     | 0.438     | 0.454     | 0.413     |
| 0.08        | 0.354     | 0.352     | 0.358     | 0.343     | 0.257     | 0.187     | 0.334     | 0.348     | 0.356     | 0.323     |
| 0.1         | 0.312     | 0.310     | 0.314     | 0.300     | 0.226     | 0.165     | 0.294     | 0.308     | 0.313     | 0.283     |
| 0.15        | 0.262     | 0.261     | 0.263     | 0.251     | 0.190     | 0.139     | 0.247     | 0.260     | 0.262     | 0.237     |
| 0.2         | 0.236     | 0.235     | 0.236     | 0.225     | 0.171     | 0.125     | 0.222     | 0.233     | 0.235     | 0.213     |
| 0.3         | 0.202     | 0.201     | 0.202     | 0.193     | 0.147     | 0.107     | 0.190     | 0.201     | 0.202     | 0.183     |
| 0.4         | 0.181     | 0.180     | 0.181     | 0.172     | 0.131     | 0.096     | 0.170     | 0.179     | 0.180     | 0.163     |
| 0.5         | 0.165     | 0.164     | 0.165     | 0.157     | 0.119     | 0.087     | 0.155     | 0.163     | 0.164     | 0.148     |
| 0.6         | 0.152     | 0.151     | 0.152     | 0.145     | 0.110     | 0.080     | 0.143     | 0.151     | 0.152     | 0.137     |
| 0.8         | 0.134     | 0.133     | 0.133     | 0.127     | 0.097     | 0.071     | 0.125     | 0.132     | 0.133     | 0.120     |
| 1           | 0.120     | 0.119     | 0.120     | 0.114     | 0.087     | 0.063     | 0.113     | 0.119     | 0.119     | 0.108     |
| 1.5         | 0.098     | 0.097     | 0.097     | 0.093     | 0.071     | 0.052     | 0.092     | 0.097     | 0.097     | 0.088     |
| 2           | 0.068     | 0.084     | 0.084     | 0.080     | 0.061     | 0.045     | 0.079     | 0.083     | 0.084     | 0.076     |
| 3           | 0.059     | 0.068     | 0.068     | 0.065     | 0.049     | 0.036     | 0.064     | 0.068     | 0.068     | 0.062     |
| 4           | 0.053     | 0.059     | 0.059     | 0.057     | 0.043     | 0.031     | 0.056     | 0.059     | 0.059     | 0.054     |
| 5           | 0.053     | 0.053     | 0.054     | 0.051     | 0.039     | 0.028     | 0.050     | 0.053     | 0.053     | 0.048     |
| 6           | 0.050     | 0.049     | 0.050     | 0.047     | 0.036     | 0.026     | 0.047     | 0.049     | 0.051     | 0.045     |
| 8           | 0.045     | 0.044     | 0.045     | 0.043     | 0.032     | 0.024     | 0.042     | 0.044     | 0.047     | 0.040     |
| 10          | 0.042     | 0.042     | 0.042     | 0.040     | 0.030     | 0.022     | 0.039     | 0.041     | 0.044     | 0.038     |
| 15          | 0.038     | 0.038     | 0.039     | 0.037     | 0.028     | 0.023     | 0.036     | 0.037     | 0.038     | 0.035     |
completely absorbed by the material. The formation of the secondary photons in the intermediate energy is greater than that of low and high energy which lead to high possibility of photons to build up in the materials. It is also clear that the buildup factor increases with the increase of the penetration depth and reaches to maximum values for penetration depth 40 mpf. The highest buildup factor is achieved for HNT4 while the lowest buildup factor achieved for HNT2.

4. Conclusion

The linear attenuation coefficient for halloysite-modified nanotubes simulated between 0.015 and 15 MeV using MCNP 5. The simulation results showed a dependence of the linear attenuation coefficient of the incident gamma-ray energy. The small variation in the linear attenuation coefficient between the studied samples is due to the small variation in chemical composition between the halloysite samples. The highest linear attenuation coefficient achieved for HNT2 and varied between 0.0385 and 9.854 cm⁻¹, while the lowest half-value obtained for HNT3 and varied between 0.0275 and 6.714 cm⁻¹ for gamma-ray energies between 0.015 and 15 MeV. The linear attenuation coefficient calculated using Phys-X/PSD is found close to that simulated using MCNP-5 code, which reveals to the accuracy of the simulated results. The better half-value is achieved for HNT2 and

Figure 5. The exposure buildup factor of the modified halloysite nanotube.
varied between 0.0703 and 17.999 cm. The effective atomic number \( Z_{\text{eff}} \) also calculated for the modified halloysites nanotube. The calculation showed that the highest effective atomic number achieved for HNT 2 and varied between 7.959 and 12.245 while the lowest effective atomic number achieved for HNT4 and varied between 6.586 and 11.639. The exposure buildup factor calculation showed that the HNT2 has the lowest buildup factor for penetration depth between 0.5 and 40 mfp. Finally, we can conclude that the halloysite sample HNT2 has the best shielding properties in our study.

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**Disclosure statement**

No potential conflict of interest was reported by the authors.

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