Evaluation of the performance of bentonite and red mud mixture as a shield in radioactive waste landfills

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Abstract. In the present era, due to various industrial developments, there is a need to use different applications of nuclear energy. But the leading challenge is the management of nuclear waste, which emits dangerous rays such as gamma rays. This research aim is to create a layer to be used as a protective coating against radiation in low-level waste landfills. In this paper, bentonite is used as a base material and red mud as an additive. Also, to determine the linear attenuation coefficient (μ), a combination of bentonite with 0, 15, 30, and 45% red mud with bentonite was used. To perform a linear attenuation coefficient test, the NaI (Tl) detector and the source of Co\(^{60}\) were used at two energy levels of 1173 and 1332 keV. The results of laboratory tests show that the addition of red mud has improved the parameter of the linear attenuation coefficient of radiation at both energy levels so that the energy levels of 1173 keV and 1332 keV with the addition of 45% red mud has improved by 16 and 12%, respectively.

1. Introduction
Today, based on the growth of nuclear technology, radioactive materials have wide applications in various fields such as medicine, industry, agriculture, etc. [1, 2]. Despite the many benefits of this industry, environmental pollution with these substances and excessive radiation emitted from radioactive waste can cause irreparable damage to water, soil, and living organisms, including humans. In this regard, one of the concerns related to the use of nuclear technology is the management of waste generated. Among radioactive wastes, low-level radioactive wastes (LLRW) are usually buried in surface landfills. To prevent the emission of excessive radiation from the landfills to the surrounding environment, it is necessary to use a protective layer against radiation emission around the cap of these tanks.

Leading materials and heavy concrete are used among various materials to prevent the spread of rays, but due to high cost and environmental hazards, they need to be re-examined [3-8]. Therefore, finding more efficient materials for the landfill structure has been the goal of many researchers. Meanwhile, bentonite has proven to be a natural, available, inexpensive, durable, and almost suitable material as a protection against...
radiation emission [9, 10]. On the other hand, due to the very low permeability of bentonite, this material is used in cases around landfills [11].

So far, research has been done to improve the performance of bentonite as a shield against radiation. Since the physical theory of radiation protection shows the importance of the density of the protective material on its performance, research has tried to investigate the effect of dense materials such as barite, steel slag, basalt, etc. on the performance of bentonite [12].

In 2004, Ciaravella et al. Conducted a study on the effect of X-rays on pure DNA and clay-impregnated DNA (montmorillonite and kaolinite). In this study, DNA samples were exposed to X-rays under different intensities. The results show that clay-impregnated DNA is several times more resistant to radiation than pure DNA [13].

In 2014, a study to evaluate the protective properties of gamma and neutron radiation from five different soil samples, namely clay loam, clay, loam, sandy clay loam, and sandy loam by Singh et al. Experiments were performed. In this study, the protective effect of gamma rays with the help of soil samples using physical parameters including the mass attenuation coefficient and the thickness of the absorption half layer, etc. under the energy level of 0.015 to 15 MeV for gamma rays has been evaluated and it is noted that the photon attenuation coefficient determines the penetration and propagation of gamma rays in composite materials such as two soils. In general, it can be said that all selected soil samples have good protective properties, but among the above five soils, the best protection against gamma and neutron radiation is related to loamy clay, which indicates the property of maximum absorption and minimum radiation permeability [14].

In 2015, a laboratory study was conducted by Akbulut et al. To investigate the effect of radiation and the permeability of gamma rays. In this study, the performance of clay along with several other materials in the role of protection against gamma radiation was evaluated. In this study, the samples were subjected to radiation permeability tests, standard density tests, and unrestricted compressive strength tests at the optimum moisture content. The results show that clay with white cement has the highest unobstructed compressive strength and the lowest amount of radiation permeability compared to other samples. Therefore, the combination of white clay and cement and even pure clay can be used as building materials in parts that are radioactive to prevent radiation emission and thus less damage to people, especially employees and people present in that environment [15].

In 2018, an article was published on the subject of gamma radiation protection using two types of clay located in southwestern Nigeria. In this research, the mass attenuation coefficient has been studied theoretically and experimentally with different energy levels. High Purity Germanium (HPGe) device was used to determine the mass attenuation coefficient in the laboratory and WinXCom software was used as a simulation. The experimental results were in good agreement with the simulation results. Also, the linear attenuation coefficient, the half-value layer (HVL) and tenth-value layer (TVL), and the mean free path were evaluated for the evaluated samples. From the analysis of the data, it can be concluded that the tested clay materials can be suitable options for gamma radiation protection for medical and nuclear applications [16].

Share Isfahani et al. (2019) investigated the effect of clay modification with barite powder to improve the protective performance of gamma rays while the hydraulic permeability of the clay is low. This research was carried out using experimental and simulation methods. In this research, bentonite with different percentages of barite powder has been used. In this study, the linear attenuation coefficients of the samples were calculated experimentally using the HPGe detector. Moreover, the linear attenuation coefficients were obtained using the MCNP code as a simulation method and finally were checked with the XCOM database. The simulation results are in good agreement with the experimental results and show that with increasing the percentage of barite powder, the linear attenuation coefficient and the hydraulic permeability coefficient
increase. It also shows that bentonite clay with 40% barite powder creates the optimal combination. It seems that this mixture can be a good alternative to radiation protection in radioactive waste disposal sites [17].

In another paper on the modification of clay performance used in low-level radioactive waste disposal centers by adding different percentages of steel slag (as industrial waste) to bentonite, two parameters of hydraulic permeability and gamma-ray shielding have been investigated. This study was performed using both experimental and simulation methods and the results show that adding steel slag to bentonite can improve the radiation protection performance while the mixture will be more permeable [18].

The researchers examined the linear and mass damping coefficients of new composite material, including a combination of bentonite and polyester, to investigate the protective effect of the material against gamma rays. To investigate these coefficients, a NaI (TI) detector with different energy levels has been used. This study shows that the increase in the thickness of polyester composite with bentonite has a linear relationship with gamma radiation absorption. They also found that the linear and mass damping coefficients with increasing energy are lower than those of [19].

In 2020, Share Isfahani et al. conducted a study on the additive effects of basalt fibers in four different percentages, including 0.5, 1, 2, and 5% to bentonite. Chemical and microstructural analysis was performed on the material used, using energy-dispersive X-ray spectroscopy (EDX) and scanning electron microscopy (SEM). The results concluded that, due to their radiation protection performance and permeability limitations, the 2% bentonite basalt mixture could be a new candidate covering low-level radioactive waste disposal [20].

Red mud is a waste that is produced in industries with a lot of aluminum. Of course, this material has a relatively high density due to its high percentage of iron. The production rate of this waste in the steel industry is significant. The goal of this research is to investigate the effect of red mud effluent as a residue on bentonite and to investigate the function of this compound as a protective layer against radiation emission.

2. Physical theory of radiation shielding materials

The linear attenuation coefficient is used to evaluate the properties of radiation protection. This coefficient is expressed according to Beer-Lambert law as follows [21, 22].

\[ I = I_0 \exp(-\mu t) \]  \hspace{1cm} (1)

In the above formula, the parameter “I” and “I₀” are the gamma-ray intensity with and without the presence of the sample, respectively. Also, “t” is the thickness of the shielding material and is the linear attenuation coefficient.

In order to calculate the required thickness for the protective material, two parameters of the half-value layer (HVL) and the tenth-value layer (TVL) are used. Which is introduced in equations 2 and 3 [23].

\[ HVL = \frac{0.693}{\mu} = \frac{\ln 2}{\mu} \text{ (m)} \]  \hspace{1cm} (2)

\[ TVL = \frac{2.3026}{\mu} = \frac{\ln 10}{\mu} \text{ (m)} \]  \hspace{1cm} (3)

Also, to better understand the effect of the additive on the efficiency of the shielding material, the mean free path coefficient (MFP) is used, which indicates the average distance that the photon particle changes direction or energy level after continuous collision [24].
\[ MFP = \frac{1}{\mu} \text{ (m)} \] (4)

It is important to use a material with a high atomic number because the shielding material with a higher atomic number has better radiation absorption properties.

3. Materials and methods
In this section, the utilized materials including bentonite and red mud, which are known as base material and additive, respectively, have been examined in the laboratory.

The bentonite used in the research is sodium type, which was purchased from Farzin Shimi Company in Isfahan. The characteristics of this sodium bentonite include a psychological index of 175% and a density of 2.57 $\text{g/cm}^3$. For testing bentonite passing through a sieve of 200, a grain size curve was used according to the ASTM D422-63 hydrometric test regulations and is shown in the figure 1.

![Grading diagram of sodium bentonite used by hydrometric method.](image)

Figure 1. Grading diagram of sodium bentonite used by hydrometric method.

Red mud is a substance that is the result of the process of iron production. This material was initially in the form of a slurry mixture which was used after drying by passing through a grade 10 sieve. It has a high percentage of silicon, oxygen, and iron.

The compounds made include sodium bentonite with 15, 30, and 45% Red mud powder. The type of compounds and how to name them are given in table 1.
Table 1. Specimen specifications used.

| Code   | Bentonite (%) | Red Mud (%) |
|--------|---------------|-------------|
| BS     | 100           | 0           |
| BSRM15 | 85            | 15          |
| BSRM30 | 70            | 30          |
| BSRM45 | 55            | 45          |

Given that the density of the material affects the performance of the shield against radiation. Therefore, to determine the best composition, first, the standard density test is performed according to ASTM D698 regulations. According to the standard compaction test of samples with different humidity in the form of a cylinder with a diameter of 101.6 mm in three layers and each layer with 25 blows due to the fall of a 24.4 Newton hammer released from a height of 305 mm and compression energy produces 600 (KN -m)/m³ compresses.

In the other part of the experiments, a NaI (Tl) detector is used to determine the amount of beam attenuation. This detector generally has five parts, including power supply, shielded lead plate, sample location, scintillation detector, and graphing system, as shown in figure 2. The device also uses a cesium source for use at two energy levels of 1173 and 1332 keV.

Figure 2. Schematic specifications of the NaI (Tl) detector.

In order to perform the radiation permeation test, the samples are created in the optimum humidity and specific gravity and are made in molds with a height of 2, 4, and 6 cm and a fixed diameter of 6 cm.
4. Results and discussion

The density of the compound is made of factors influencing the attenuation of gamma rays. Therefore, figure 3 shows the effect of the percentage of red mud added to the specific gravity and optimum moisture content of the composition. This figure indicated that the increasing the percentages of additives in the composition conclude the higher the specific gravity of the mixture. On the other hand, with the addition of red mud up to 30% of the composition, the optimal moisture content decreases and then increases. However, it should be noted that despite this increasing trend, the optimal moisture content in the composition has 45% less red mud powder compared to pure bentonite. Perhaps the decrease and then increase of this process can be attributed to two factors. First, clay is a water-absorbing material that the higher the amount in the composition caused the higher the optimum moisture content. Second, the presence of red mud in the bentonite turbine causes impurities compared to the pure bentonite mixture, which increases the number of cavities, and the same increase in cavities causes more water absorption (optimal moisture). Given that the two factors have opposite effects. Therefore, it causes decreasing and then increasing changes in the optimum moisture content.

![Figure 3](image)

**Figure 3.** Diagram of the relationship between density and optimal humidity with the percentage of red mud.

The results of radiation permeability measurement tests for the introduced samples are shown graphically in Figure 4. According to Beer-Lambert law, the slope of the line diagram below represents the linear attenuation coefficient of the beam. What is remarkable is that the linear regression is close to the number one, which means good accuracy of this experiment.
Figure 4. Experimental results of determining the radiation attenuation coefficient of bentonite samples with different percentages of red mud with different energy levels.
Table 2. Summary of physical coefficients.

| Parameter or sample | 1173   | 1332   |
|---------------------|--------|--------|
|                     | $\mu (1/m)$ | HVL | TVL | MFP | $\mu (1/m)$ | HVL | TVL | MFP |
| BS                  | 8.89   | 0.078 | 0.259 | 0.112 | 7.78 | 0.089 | 0.296 | 0.128 |
| BSRM15              | 9.36   | 0.07  | 0.24  | 0.106 | 9.7  | 0.07  | 0.24  | 0.103 |
| BSRM30              | 9.71   | 0.07  | 0.25  | 0.103 | 8.32 | 0.08  | 0.28  | 0.120 |
| BSRM45              | 10.32  | 0.06  | 0.22  | 0.097 | 8.69 | 0.08  | 0.27  | 0.115 |
| BCPB40              | 11.43  | 0.056 | 0.18  | 0.087 | 11.99| 0.057 | 0.19  | 0.083 |
| BCSS40              | 11.91  | 0.058 | 0.19  | 0.084 | 10.98| 0.06  | 0.21  | 0.091 |

Table 2 summarizes the results along with other physical parameters. The coefficients of $\mu$ and HVL in pure bentonite samples were 8.89 units per meter and 0.078 meters, respectively, which after adding 45% of red mud reached 10.32 units per meter and 0.06 meters. Also, compared to the work of other researchers, it can be said that the efficiency of red mud powder is 45% weaker than the sample with 40% barite powder or 40% steel slag.

5. Reference

[1] Papachristoforou, M. and I. Papayianni, Radiation shielding and mechanical properties of steel fiber reinforced concrete (SFRC) produced with EAF slag aggregates. Radiation Physics and Chemistry, 2018. 149: p. 26-32.

[2] Chegbeleh, L.P., et al., Concepts of Repository and the Functions of Bentonite in Repository Environments: A State–of–the–art review. Okayama University Faculty of Environmental Science and Engineering Research Report, 2008. 13(1): p. 1-5.

[3] NCRP, Structural Shielding Design and Evaluation for Megavoltage X Ray Radiotherapy Facilities. NCRP Report., No.151, Oxford University press, 2006.

[4] Glasgow, G.P., Structural Shielding Design and Evaluation for Megavoltage X-and Gamma-Ray Radiotherapy Facilities. Medical physics, 2006. 33(9): p. 3575-3578.

[5] Mostofinejad, D., M. Reisi, and A. Shirani, Mix design effective parameters on $\gamma$-ray attenuation coefficient and strength of normal and heavyweight concrete. Construction and Building Materials, 2012. 28(1): p. 229-224.

[6] Gencel, O., Physical and mechanical properties of concrete containing hematite as aggregates. Science and Engineering of Composite Materials, 2011. 18(3): p. 199-191.

[7] Topcu, İ.B., Properties of heavyweight concrete produced with barite. Cement and Concrete Research, 2012. (6): p. 203-203.

[8] Bashter, I., A.E.-S. Abd, and A. Makarious, A comparative study of the attenuation of reactor thermal neutrons in different types of concrete. Annals of Nuclear Energy, 1996. 23(14): p. 1189-1195.

[9] Reijonen, H.M., J. Kuva, and P. Heikkilä, Benefits of applying X-ray computed tomography in bentonite based material research focused on geological disposal of radioactive waste. Environmental Science and Pollution Research, 2020: p. 15-1.
[10] Norris, S., et al. Radioactive waste confinement: clays in natural and engineered barriers. 2017. Geological Society of London.

[11] Mann, K.S., M.S. Heer, and A. Rani, Investigation of clay bricks for storage facilities of radioactive-wastage. Applied Clay Science, 2016. 119: p. 256-249

[12] Kavaz, E., et al., Gamma ray shielding effectiveness of the Portland cement pastes doped with brass-copper: An experimental study. Radiation Physics and Chemistry, 2020. 166: p. 108526

[13] Ciaravella, A., et al., Role of clays in protecting adsorbed DNA against X-ray radiation. International Journal of Astrobiology, 2004. 3: p. 35-31

[14] Singh, V.P., N. Badiger, and N. Kucuk, Gamma-ray and neutron shielding properties of some soil samples. 2014

[15] Akbulut, S., et al., A research on the radiation shielding effects of clay, silica fume and cement samples. Radiation Physics and Chemistry, 2015. 117: p. 92-88

[16] Olukotun, S., et al., Investigation of gamma radiation shielding capability of two clay materials. Nuclear Engineering and Technology, 2018. 6(5): p. 962-957

[17] Isfahani, H.S., et al., Permeability and Gamma-Ray Shielding Efficiency of Clay Modified by Barite Powder. Geotechnical and Geological Engineering, 2019. 37(2): p. 855-845

[18] Isfahani, H.S., et al., Investigation on gamma-ray shielding and permeability of clay-steel slag mixture. Bulletin of Engineering Geology and the Environment, 2019. 78(6): p. 4598-4589

[19] Kaçal, M., F. Akman, and M. Sayyed, Evaluation of gamma-ray and neutron attenuation properties of some polymers. Nuclear Engineering and Technology, 2019. 51(3): p. 824-818

[20] Isfahani, H.S. and A. Azhari, Investigating the effect of basalt fiber additive on the performance of clay barriers for radioactive waste disposals. Bulletin of Engineering Geology and the Environment, 2020. p. 12-1

[21] Chilton, A.B., J.K. Shultis, and R.E. Faw, Principles of radiation shielding. 1984

[22] Martin JE (2006) Ch. 8: Radiation Shielding. In: Physics for radiation protection: a handbook. John Wiley & Sons, H., pp 367–423

[23] Jaeger, R.G., et al., Engineering Compendium on Radiation Shielding. Volume 1. Shielding Fundamentals and Methods. 1968: Springer.

[24] Han, I., L. Demir, and M. Şahin, Determination of mass attenuation coefficients, effective atomic and electron numbers for some natural minerals. Radiation Physics and Chemistry, 2009. 78(9): p. 760-764