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Experimental investigation of contaminated clay slabs for X-ray shielding

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Abstract. The advent of radiation is a challenge to the human race therefore; a suitable material is a solution to effective radiation shielding. This research aims to investigate the effectiveness of clay slabs mixed with some heavy metals in radiation shielding. Thereafter, absorption coefficients (mass and linear) were calculated from both incident and transmitted intensity measured with a detector at different energies: 40 keV, 60 keV, 80 keV, 100 keV, 120 keV, 150 KeV and 200 Kev. The following procedures were taken: the clay was mixed with heavy metals (Lead, Nickel Cadmium, Zinc and Copper) using the ion exchange capacity technique and then slabs were moulded using a wooden frame of dimensions 4 cm x 4 cm x 1 cm. A narrow collimated beam transmission geometry condition was used for the measurements. The measured values of these parameters were in good agreement with the literature. At selected energies, the values of linear and mass attenuation for lead were 2.29 cm^{-1} and 0.466 gcm^{-2} , 1.58 cm^{-1} and 0.392 gcm^{-2} , 1.23 cm^{-1} and 0.281 gcm^{-2} , 1.09 cm^{-1} and 0.216 gcm^{-2} , 0.88 cm^{-1} and 0.180 gcm^{-2} , 0.74 cm^{-1} and 0.168 gcm^{-2} and 0.57 cm^{-1} and 0.161 gcm^{-2} , respectively. The results obtained were compared with concrete. The clay slabs mixed with heavy metal showed good shielding properties for moderate radiation energy. Hence, these slabs are feasible and cost effective compared with traditional concrete used for construction.

Keyword: Clay-adsorption, Heavy metal, Mass attenuation, Radiation, X-ray energy

1. Introduction

The report of harmful radiation effect has caused various practitioners to suggest a variety of radiation safety materials such as Lead and concrete. Cost implication of lead (block) in shielding X-ray led to a significant search for alternative materials [1]. Now, it is necessary to search for locally available and cheap innovative shielding material for the rapid expansion of the use of medical equipment such as X-ray machine is significance. Here, clay, a geological material, can play a great role; it has better photon energy absorption characteristics than other soil types [2, 3]. Clay is a geological material composed of fine grain minerals, which shows plasticity when moist and become hard when dried or fired [4]. Clay has been used in building construction especially in the manufacturing of bricks since ancient times [5].

Most clay has the ability to exchange ions (electrically charged atoms and molecules) from a solution and release the ions later when conditions change [6]. This property of clay to adhere with ions is called cation exchange capacity [7]. Surfaces of clay minerals are negatively charged and this is why clays easily adsorb heavy metal ions (such as chromium, lead, copper, nickel, cadmium, etc.) to itself. Its negative surface is balanced with positive ions to maintain neutrality [8, 9]. However, clay may contain heavy metals depending on the prevailing geologic minerals and contact with environmental pollutants [7, 10]. These heavy metals are found as waste



products from our industries. Many industries dump their metallic waste on clay soil which in turn interacts with each other, as a result of decomposition. Hence, this research is focused on attenuation parameters of contaminated ss-labs. We shall mimic what really happened at the dump site in the laboratory and provide what the product can be used for.

The effect of grain size and voids in various soil samples on mass attenuation coefficient has received attention by researchers [11]; result showed that mass attenuation coefficient increased with the decrease of grain size. Clay bricks, translucent louver blade and verified stone tiles are other materials that have attracted a lot of attention in recent times as radiation shield [12]. Some works have been carried out in determining the radiation shielding ability of clay and some soil used for building in Bangladesh and Brazil [13, 14]. Clay also find its applications in paper foundry, building material, factories, ceramics, drilling, pharmaceutical, textile and adhesives industries [15,16]. It has also been proposed as one of the materials that shield ionizing radiation. Russian scientists used clay to protect their body while working with nuclear materials [16]. Clay deposit can be found in Nigeria: Ara-Ijero, Orin-Ekiti, Omibode, Ifon, Ikere, Itu, Kankara, Barkinladi, Lagelu (Ibadan), Ijoko, Ara-Ekiti, Eha-Alumona, Isan, Ezinachi-Okigwe, Jos, Oshiele, Enugu, Ukpok and Niger Delta [17-21].

The aim of this study was to explore the possibilities of the utilisation of contaminated clay, which was done experimentally by adding some heavy metal (solution) with fine grain clay. However, an attempt was to measure X-ray interaction parameters to assess the shielding effectiveness of the samples.

1.1 Theoretical Background

If a material of any substance is interposed in the path of a homogeneous beam of X-ray, its intensity decreases. The decrease in intensity, dI is observed to follow the equation

$$dI = -\mu I dx \quad (1)$$

Here μ is called the linear absorption coefficient which depends on the wavelength (or frequency) of X-ray used and the nature of the absorbing material. Linear absorption coefficient ' μ ' has the dimensions of reciprocal of length and its unit is therefore m^{-1} .

Mass absorption coefficient, $\alpha = \mu/\rho$ and c is the mass per unit area of the absorbing material (i.e. product of density and thickness).

By integrating equation (1)

$$\int \frac{dI}{I} = -\mu \int dx \quad (2)$$

$$\ln I - \ln I_0 = -\mu(x - 0)$$

$$\ln \left(\frac{I}{I_0} \right) = -\mu x$$

We have, $I = I_0 e^{-\mu x}$ (3)

$$I = I_0 e^{-\left(\frac{\mu}{\rho}\right)\rho x}$$

$$I = I_0 e^{-\alpha c} \quad (4)$$

2. Materials and method

The clay sample used in this research study was collected from Ibadan, southwestern Nigeria; to be precise, it was collected from the deposit at Bricks and Clay products Industry, Omi Adio, Ido Local Government Area, Oyo State. The elemental compositions of the clay used were analysed by using an energy dispersive X-ray fluorescence spectrometer. This location can be found on Longitude 3°30 and Latitude 7°32. The chemical and mineralogical information of selected sample was reported by [21, 22], respectively (Tables 1 and 2).

2.1 Preparation and analysis of digested sample

The clean (unpolluted) clay sample collected was crushed with pestle and mortar and passed through 2 micrometres sieve. The sieved sample was put into polythene bag, labelled and stored at room temperature while the pebbles were discarded. The crushing and sieving were done to make the sample homogenous and fit for the experimental work.

Samples were digested by using nitric acid extraction method. All chemicals used for this digestion were reagent grade and conform to standards established by the American Chemical Society. Prior to artificial contamination, the initial heavy metal content in the clean sample collected was determined. To determine the amount of heavy metal content of clay sample 2 g of the sample was digested according to EPASWS46 method 3959(EPA,1986).The digestate was filtered. Filtrate was analysed for Pb (0.738 ppm), Cr (2.845 ppm), Ni (1.762 ppm), Cu (2.88 ppm) and Cd (0.144 ppm) content using Atomic Absorption Spectrometer (AAS) by direct method. The concentration of these heavy metals in the clay sample were given in part per million (ppm) by the machine.

2.2 Preparation of Nitrate of Heavy Metals Stock Solutions

6400 ppm of lead, chromium, cadmium, nickel, and copper nitrate solution were prepared as the stock solution. The concentrations of nitrate of heavy metals needed for this work were prepared from stock solution using dilution method. Concentration of heavy metal nitrate solutions prepared was 400 ppm, 800 ppm, 1600 ppm, 3200 ppm and 6400 ppm. However, slab mixed with 6400 ppm concentration was presented for this work.

10.23 g of $Pb(NO_3)_2$ salt that contained 6.4 g of Pb was weighed and transferred to one litre standard flask filled. De-ionized water then poured into the flask up to the mark. This concentration is considered to be 6400 ppm stock. The method of preparation of this stock solution was based on mass volume relationship.

2.3 Formation of slabs from clay sample

Dry clay sample of mass 52.0 g (volume, 43 cm³) was put in the beaker. Prepared 70 cm³ lead solution was added to the measured powder clay. The solute (powder clay) and solvent were mixed together with the help of stirrer. Some minutes later, the mixture was gently poured into a funnel containing filter paper placed on beaker and left for two days under unpolluted atmospheric condition, just to avoid contamination. Three days later, the residue was placed and dressed in a clean frame of dimension 4cm x 4cm x 1cm. After two days, the slab was taken out of the frame and sun-dried under pollution free environment. The procedure was repeated for different concentration of lead solution (400 ppm, 800 ppm, 1600 ppm, 3200 ppm and 6400 ppm). Subsequently, nickel, cadmium, chromium and copper were added. After sun-drying, the samples were oven dried and taken to National Institute of Radiation Protection and Research Laboratory, University of Ibadan, Oyo state for attenuation capacity examination.

2.4 Experimental set up

The linear attenuation coefficient measurements of the prepared samples were performed in National Institute of Radiation Protection and Research laboratory, university of Ibadan. A PTW UNIDOS Electrometer was employed to record the counting in the control room. It consisted of two linear collimators: one at the front of the ionization chamber and another for the X-ray source. The x-ray machine is the source of radiation, ionization chamber detected the amount of radiation transmitted through sample while the lead collimator was employed to produce radiation beam of narrow parallel geometry. The clay brick was placed between two collimators each of aperture of 1.2 mm at a distance of 100 cm from the x-ray source. X-ray machine was first preconditioned for about 30 minutes to allow the machine to attain the operational temperature and voltage for good emissions of electrons having been left unused for some period of days. The counting process involves the measurement of the direct flux from the source, which was first taken without placing any sample between the X-ray source and ionization chamber and also counting was recorded when lead block was placed in between the detector and ionization chamber. Samples were subsequently placed between the X-ray source and the detector at linear collimator geometry. X-ray energies 40 KeV to 120 KeV were irradiated on each sample for 30 seconds by this procedure, the intensity of the direct (I_0) and the transmitted (I) flux were determined and the counting was recorded by the electrometer. Counting was done three times for each sample and later average value was calculated.

The measured incident and transmitted intensities together with the measured thickness (i.e. 1cm) of the samples were used in equation (5) below to calculate the linear attenuation coefficients:

$$\mu = \frac{\ln(I_0/I)}{t} \quad (5)$$

where μ is the linear attenuation coefficient in m⁻¹: and t is the thickness of the sample in m. The x-ray count without the sample is I_0 and I is the x-ray count with sample in the field of the narrow parallel beam. The mass attenuation coefficients (μ/ρ) in m²/kg for the different samples were also calculated.

3. Results and Discussion

The chemical and mineralogical compositions of clay sample for this research work are presented in Tables 1 and 2, respectively [23]. The difference in concentrations of oxide of metal and non-metal may be attributed to the difference in their chemical affinity and ion exchange capacity with respect to the chemical functional group on the surface of the adsorbent. Analytical result (Table 1) shows the dominance of SiO_2 and Al_2O_3 over all other oxides, as these alone constitute about 70% of the bulk composition of the clay. The low content of Fe_2O_3 in this clay may be attributed to geological breakdown of soil formation. Moreover, in Table 2, kaolinite and quartz are the dominant minerals while illite is in minor quantity. kaolinite is formed by the decomposition of orthoclase feldspar in granite. These minerals are the product of chemical interaction between the leached solutions from the upper layer of the soil overburden and kaolinitic clay[22].

Table 1: Chemical composition of the clays collected from selected locations

Oxide (wt%)	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	NaO	K_2O	TiO_2	P_2O_5	Cr_2O_3	LOI	Total
Omi Adio-Ibadan [22]	44.98	37.54	2.35	0.01	1.72	0.09	0.19	1.01	1.42	0	0	12.6	99.01

Table 2: Mineralogical composition of the clays collected from selected location.

Mineral %	Omi Adio-Ibadan [22]
Kaolinite	63
Quartz	21
K.Feldspar	0
Illite	3
Smectite	-

The experimental values of linear and mass attenuation coefficients for clay sample mixed with nitrate of heavy metal at 6400ppm are presented in Tables 3 and 4, respectively. The experimental values of mass attenuation coefficients for the clay slab mixed with some heavy metals are compared with another shielding material common in literature (concrete) as shown in Table 4. The results obtained showed considerable decrease in the attenuation coefficient as photon energy increases. However it reveals the interaction of photon energy in the mixed clay sample. In addition, intensity decreases as the concentration of some heavy metal increases. The ability of this sample to attenuate the x-radiation depends on the concentration of adsorbed heavy metal ions.

Table 3: Calculated value of linear attenuation coefficient for doped clay of different nitrate of heavy metals at various energies when concentration is 6400 ppm.

Energy, KeV	Linear attenuation Coefficient, cm^{-1}				
	Cod	Pb	Ni	Zn	Cu
40	2.24	2.29	2.01	2.21	2.16
60	1.55	1.58	1.36	1.52	1.49
80	1.19	1.23	1.07	1.17	1.15
100	1.01	1.09	0.89	0.98	0.96
120	0.83	0.88	0.75	0.82	0.79
150	0.64	0.74	0.59	0.63	0.60
200	0.44	0.57	0.39	0.43	0.40

Table 4: Mass attenuation coefficients of doped clay at 6400 ppm and concrete.

Energy, KeV	Mass attenuation coefficient, cm^2/kg					
	Cod	Pb	Ni	Zn	Cu	concrete
40	0.455	0.466	0.409	0.450	0.430	0.304
60	0.384	0.392	0.337	0.377	0.369	0.243
80	0.252	0.281	0.229	0.248	0.244	0.214
100	0.184	0.216	0.173	0.179	0.175	0.170
120	0.159	0.180	0.156	0.164	0.156	0.143
150	0.149	0.168	0.141	0.143	0.142	0.139
200	0.140	0.161	0.130	0.132	0.131	0.124

The graphical relationship between attenuation coefficient and photon energy for samples mixed with lead, cadmium, nickel, zinc and copper are shown in Figure 4. We deduced that the attenuation coefficient decreases exponentially as the beam energy increases. This showed sharp agreement between this work and literature. As the beam energy increases, the sample attenuates fewer photons than the lower energy photons. Figure 3 showed that lead, among others, was the best element that has the ability to reduce the penetration of radiations.

The linear attenuation coefficient of cadmium versus photon energy is presented graphically as shown in Figure 2. The graph showed exponential behaviour which in turn revealed shielding capacity of the slab mixed with cadmium. It was also observed that linear attenuation of slab mixed with cadmium decreases as a result of interaction of photon energy. Analytical result showing the dominance of SiO_2 and Al_2O_3 over all other oxides is presented in Figure 1, as a result of geological formation of the sample.

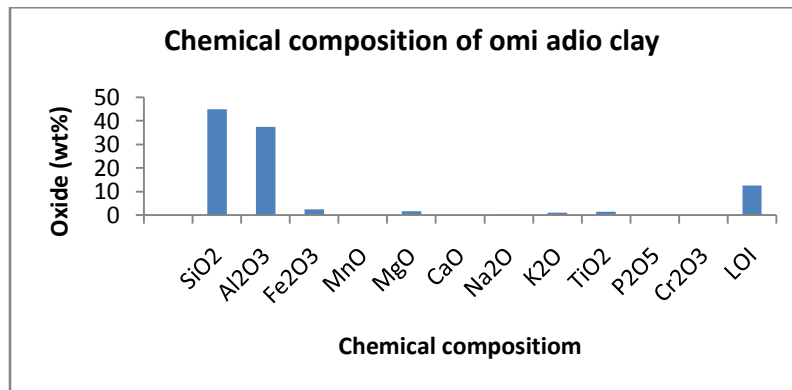


Figure 1: Analytical result showing the dominance of SiO₂ and Al₂O₃ over all other oxides, as these alone constitute about 70% of the bulk composition of the clay.

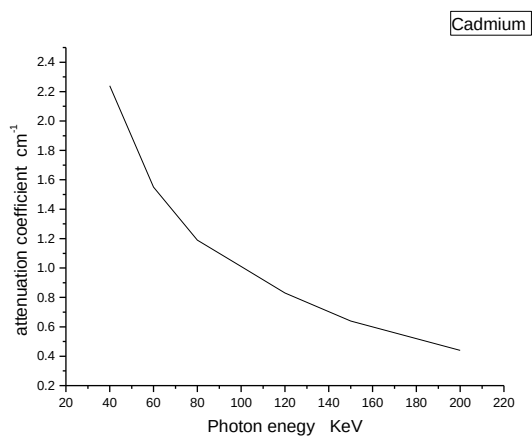


Figure2: Attenuation coefficient of clay sample mixed with cadmium versus energy of radiation.

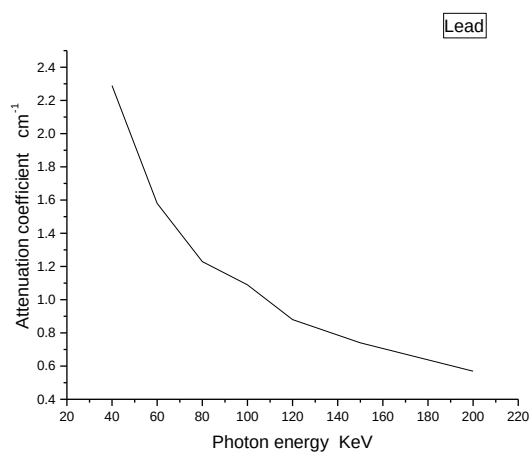


Figure3: Attenuation coefficient of clay sample mixed with lead versus energy of radiation.

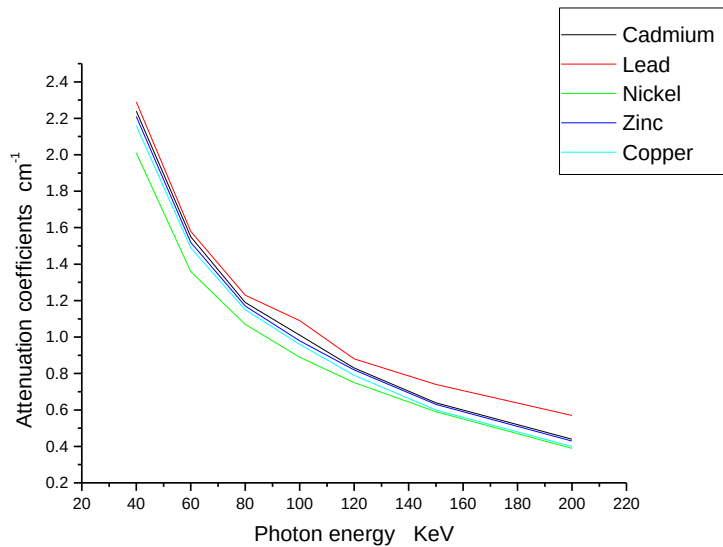


Figure4: Curves illustrating the systematic variation of linear attenuation coefficient of clay sample mixed with some heavy metals against energy of radiation.

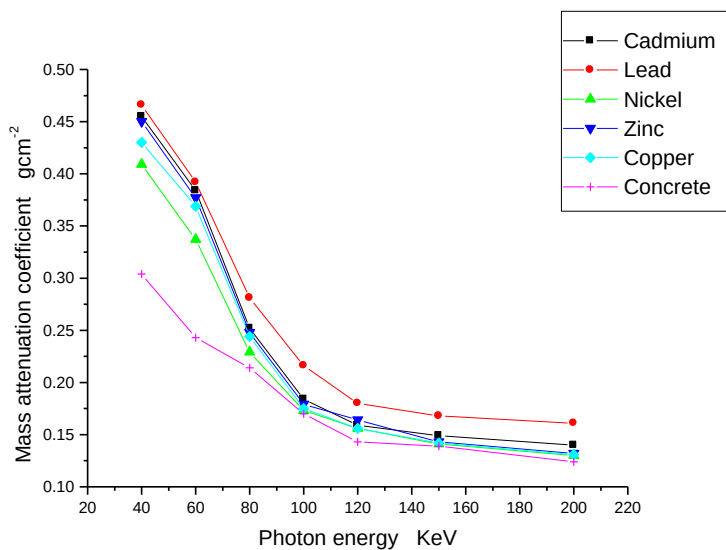


Figure 5: Curves illustrating the systematic variation of mass attenuation coefficient of clay sample mixed with some heavy metals against energy of radiation.

The attenuation properties of clay samples vary systematically with the energy of the radiation, and as well as the atomic number of the element present in the mixed clay samples, as shown in Figure 3. It was noted, however, that in region where the Compton effect predominates, the mass attenuation coefficient is almost independent of the atomic number of the sample at photon energy, 100 Kev (Figure 5).

Figure 3 showed graphically representation of linear attenuation coefficient of lead against photon energy. It was observed that linear attenuation of slab mixed with lead decreases as a result of interaction of photon energy. The graph showed shielding capacity of the slab mixed with lead which is better than cadmium

Figure 5 showed the comparison between mass attenuation coefficient values for the clay slab mixed with some heavy metals at 6400 ppm and another shielding material, concrete. The results showed an inverse relationship between the mass attenuation coefficients and the photon energy. At low energy, the mass attenuation coefficient decreases sharply due to the dominance of the photon electric effect, whereas at high energy, Compton and pair production are dominant [25]. Moreover, the linear attenuation coefficient values increase with an increase in the concentration (density) of heavy metal in the sample. By comparison with concrete, it is obvious from the results that linear attenuation coefficients are greater than those of concrete. Therefore, the experimental contaminated clay sample (clay mixed with heavy metals) would be a better shielding material, an alternative to concrete.

4. Conclusion

Experimental investigation showed that contaminated clay could be an alternative source of shielding X-rays or gamma rays. Attenuation coefficients of clay slab obtained in this study is an important data that showed a reasonable photon interaction. The sampled examined showed a reduction of the transmitted intensity of radiation as the concentrations of metallic (heavy metals) ions increase, which is in agreement for a shielding material to radiation [24, 25]. Therefore, for an improved shielding effectiveness, clay slab doped with heavy metals can be used to replace concrete or lead block at moderate radiation energy. However, clay soil contaminated with lead, nickel and copper can be an alternative for radiation shielding. This product can be lined with cement to avoid physical contact. Therefore, contaminated clay from industrial dumpsite should not be a waste product but a useful material in the building industry.

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