Attenuation Characteristic of Clay Irradiated With X-Ray in the Range between 40 Kev And 120 Kev

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Abstract: *The linear attenuation coefficient of clay at varying incident of energies is an important characteristic that needs to be studied and determined prior to using such a material in radiation protection. The linear attenuation coefficient and intensity as well as thickness were determined for natural clay (A) and artificially contaminated clay (B) to assess their use in radiation shielding. A narrow collimated beam of x-rays from sources with varying energies were passed through various thicknesses of clay slabs. The attenuation in the incident intensity of the beam was determined for each thickness of the clay slabs. Linear attenuation coefficients of A and B were compared with that of concrete. The results showed that linear attenuation coefficient value of artificially contaminated clay is higher than concrete by 37 % at 40keV.Therefore, artificially contaminated clay is a good absorber of radiation and can be used as x-ray wall material.*

I. Introduction

Clay and clay minerals are the most important industrial minerals (Murray, 1963). Clay is a geological material with many applications. Millions of tons of clays are utilized yearly in various fields of applications. These applications are used in engineering, geology, industries, agriculture, environmental remediation and medicine (Murray, 1963; Ekose 1994; Murray, 1997, 1999, 2005).

The reason for utilization of certain clay minerals in a specific application is that the physical and chemical properties of a particular clay mineral depend on its structure and composition. The structure and composition of clay minerals are very different even though they have basic building blocks. However, the arrangement and composition of these basic structures account for major and minor differences in the physical and chemical properties of clay minerals (Murray, 1999). Properties of clay such as plasticity, surface area, cation exchange capacity, and malleability are other reasons clay finds its applications in a number of industries such as plastics, paint, ceramics, ink, catalysts, pharmaceutical and fibre glass among others (Worall, 1975; Murray, 1980; Emufurieta et al., 1992).

Due to cation exchange capacity and large specific surface area, clay is used for remediation of heavy metals. When clay surface is exposed to solution of different metals its surface adsorps heavy metal ions (such as Iron, Lead, etc.) easily. Its negative surface is balanced with positive ions to maintain neutrality (Bolt et al, 1976). The large charged-ion-surface area and negative charge of clay enable it to attract metal ions (Kim et al, 2006). However, Clay becomes contaminated by the accumulation of heavy metals through emissions from the rapidly expanding industrial areas, land application of fertilizers, disposal of high metal wastes, spillage of petrochemicals, etc (Benjamin, 1983). Heavy metals such as lead (Pb), chromium (Cr), arsenic (As), zinc (Zn), cadmium (Cd), copper (Cu), mercury (Hg), and nickel (Ni) are commonly found on dumped sites and these are group of elements on the periodic table of elements, having specific gravities greater than 4.0 (Adriano, 1986).

Clay has received considerable attention especially as potential adsorbents in environmental research. Many researchers around the world, have beamed their search lights on the phase developments that occurred by sintering clay in the presence of some oxides (Kong et al,2003; and Biswajoy et al, 2010). The physical, chemical and mineralogical characteristics of Odukpani clay deposits and their brick forming properties have recently been reported (Attah et al., 2001; Oden et al., 2001). Lead adsorption in clay and its application was investigated by Adegoke and Egbeyale (2010). Variation of Thermophysical Properties of Clay with Adsorption Optimization was repoted by Adegoke et al, (2010). Effect of CEC of clay on thermal conductivity has been investigated (Adegoke et al, 2015). Some workers (Akpokodje, 1992; Adeyemi, 1994; Gbadebo, 2002) have investigated geotechnical properties of clay materials in Nigeria. Adeyemi, (1994) inferred that differences in mineralogical and engineering properties of clays are often caused by pedogenic factors such as parent rocks, relief and climate. Borode et al (2000) reported the alumina content of some Nigerian clay to be low, for their

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use alone without blending. Obikwelu, (1987) suggested blending two or more clays from different locations helps to improve the characteristics of clay materials for industrial applications.

In agriculture, clays provide some of the most catalytic surfaces in sedimentary environments, which are important to a variety of biogeochemical cycles (Kostka *et al*., 2002). The interaction of clay particles and microbial cells is dependent on the size and charge of exchangeable cations and on electrolyte concentration, just as for other negatively charged colloidal particles (Oades, 1989). Stotzky (1986) stated that surface interactions between clays and microbes in soils occur in situ. Since soil is a major habitat for microorganisms he added that, in soils with low levels of organic substrates, the clay minerals concentrate these substrates at the solid-liquid interface, where they reach levels high enough to support the growth of microbes which adhere to these surfaces in response to the nutrient enrichment.

Thus, the present study is designed to use analytical techniques to characterize and compare natural clay (A), artificially contaminated clays (B) and concrete based on their linear attenuation coefficients with the view to assess their ability to attenuate X-ray radiation at various energies and to assess their suitability for use in building x-ray rooms.

Theoretical Background
a. Interaction between clay minerals and metals:
Generally, clays and clay minerals are used for a large variety of environmental applications such as water purification, waste treatment, mineral barriers for waste deposits, and slurry walls for the encapsulation of contaminated areas. This is due to their large specific surfaces and the resulting ability to adsorb cations (Bradl, 2002). As shown in figure A.

• **Interaction of clay particles and water molecules**
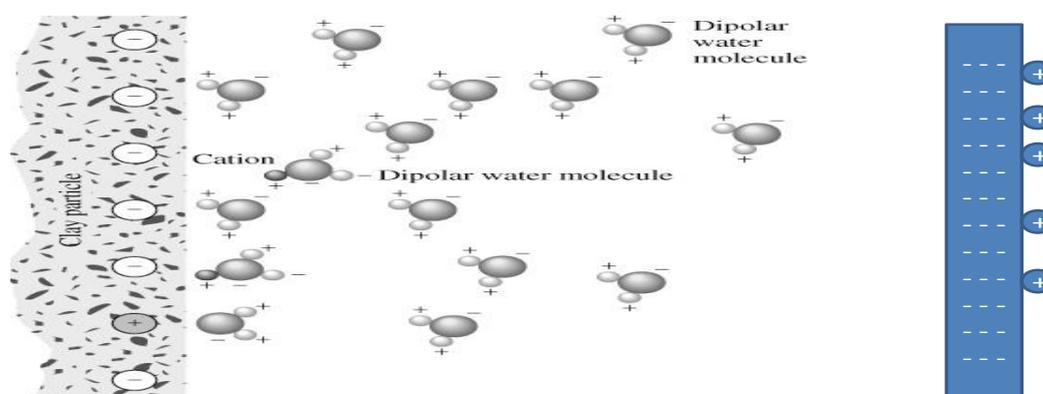
Figure A: Attraction of dipolar molecules
(Lambe and Whitman, 1979)

b. Cation Replaceability
Ion exchange is a chemical reaction in which free mobile ions of a solid, the ion exchange, are exchanged for different ions of similar charge in solution (Lucy, 2003). Many natural mineral compounds, such as clays (e.g., bentonite, kaolinite, and illite), vermiculite, and zeolites (e.g., analcite, chabazite, sodalite, and clinoptilolite) exhibit ion exchange properties (Zagorodni, 2007). Ion exchange in clays and other minerals is dependent on the crystalline structure of the mineral and on the chemical composition of any solution in contact with the mineral (Chernjaskaja, 1988). The structures of clay minerals provide a background for the ion-exchange reactions. Ion exchange in these minerals is a reversible chemical reaction that takes place between ions held near a mineral surface by unbalanced electrical charges within the mineral framework and ions in a solution in contact with the mineral. Generally the excess charge on the mineral is negative, and it attracts cations from the solution to neutralize this charge. The chemical reactions in ion exchange follow the law of mass action, but the reactions are restricted by the number of exchange sites on the mineral and by the strength of the bonding of the exchangeable cations to the mineral surface (Yamasaki et al, 1984). According to Lambe and Whitman (1979) cation exchange reaction can be summarized as follow:
i. Different types and quantities of cations are adsorbed to balance charge deficiencies in clay particles.

ii. The types of adsorbed cations depend on the depositional environment. For example, sodium and magnesium are dominant cations in marine clays since they are common in sea water. In general, calcium and magnesium are the predominant cations (Lambe and Whitman, 1979).
iii. The adsorbed cations are exchangeable (replaceable). For example,
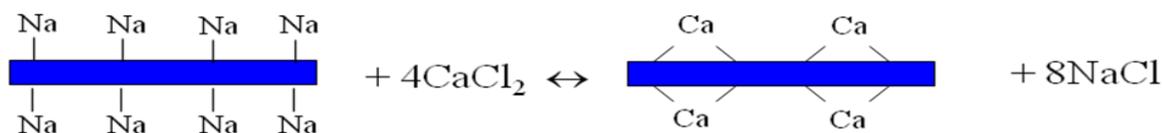
c. Attenuation coefficient

When a photon passes through an attenuator, the probability that an interaction will occur depends on its energy and the composition and thickness of the attenuator. The thicker the attenuator, the more likely an interaction will occur. Hence, linear attenuation coefficient (μ) is the actual fraction of photons interacting per 1-unit thickness of material.

If an incident beam of photons of intensity *I* is considered and the beam is directed onto an attenuator of thickness *Δx* with a photon detector recording the transmitted beam intensity. Then the fractional decrease in beam intensity is related to absorber thickness as:

$\Delta I / I \approx -\mu \cdot \Delta x$

The quantity *μ* is the linear attenuation coefficient of the attenuator material. The minus sign indicates that the beam intensity decreases with increasing attenuator thickness.

According to Lambert-Beer equation:

$$I = I_o e^{-\mu x}$$

Where *Io* is the incident intensity of the photon, *I,* is the transmitted intensity of the photon, *x* is the thickness of the absorbing medium and μ is the linear attenuation coefficient. This equation relates the intensity of radiation photons at a specified energy after attenuation coefficient. *I* is the amount of energy per unit time per cross sectional area of radiation that was able to penetrate the material thickness. The lower the value of *I*, the better the material in shielding ionizing radiation.

Radiation measurements are possible because of interaction of radiation with matter. Generally, radiation interaction with matter involves transfer of energy from radiation to the matter which it transverses. Dunn, (1957) reported that for most radiation shielding, photon energies of 10 keV to 10 MeV are important.

Geology of the study area

The area of study is located in the west of Nigeria. Omi-adio, one of the suburb villages in Ibadan falls wholly within the Pre-Cambrian rocks of Southwestern Nigeria which is part of the Nigerian Basement Complex (Oyediran and Aladejana, 2011). The rock types have undergone various episodes of tectonics depicted by foliation of the rock (Rahaman, 1976). Major rock types underlying the area include mica (mother of clay minerals), schist, quartzite and quartz schist, biotite and biotite-hornblende-gneiss, granite-gneiss and migmatite. Minor rock types are Augen-gneiss, pegmatite and amphibolites (Jones and hockey, 1964).

II. Material and method

2.1 Sample collection

Clay samples were sourced from Omi Adio Ibadan, Oyo State in the southwestern part of Nigeria. It is located on Longitude $3^0 30^1$ and Latitude $7^o 32^1$.

2.2 Sample preparation

Fresh clay samples collected was crushed with pestle and mortar and sieved. The sieved sample was put into polythene bag.

Samples were digested by using Nitric acid extraction method and Cation Exchange Capacity (CEC) was determined by using standard method.

The digested samples were analyzed using Atomic absorption Spectrophotometer. The following heavy metals were obtained; Lead (Pb), Chromium (Cr), Nickel (Ni), Cadmium (Cd), Manganese (Mn), Copper (Cu) and Zinc (Zn) (Table1). The concentration of these heavy metals in the clay sample were given in part per million (ppm) by the machine and percentage of each metal was calculated as shown in Table 1.

A mass of 10.23g of $Pb(NO_3)_2$ salt containing 6.4g of Pb was weighed and transferred quantitatively to one litre standard flask. De-ionized water was then poured into the flask up to the mark. This formed a concentration of 6400 ppm. The method of preparation of this solution was based on mass - volume relationship.

Clay sample of mass 52.0g (volume, 43cm^3) was put in a beaker; nitrate of prepared lead solution 6400ppm, volume 70cm^3, was added to the measured powdered clay. The solute and solvent were mixed together using a stirrer. Three 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, to avoid contamination. The residue was then placed and dressed in a clean frame of dimension 4cm x 4cm x X cm (X = 1, 2, 3, …10); after which the slab was taken out of the frame and oven-dried under pollution free environment. This procedure was repeated for all samples.

Samples were taken to National Institute of Radiation Protection and Research laboratory, University of Ibadan, Oyo state where attenuation capacity of both natural clay and artificially contaminated clay samples were examined. The results obtained were presented in Table 1- Table 5

2.3 *Experimental procedure for attenuation coefficient*

The experiment was carried out at the Secondary Standard Dosimetry Laboratory (SSDL) of the National Institute of Radiation Protection and Research (NIRPR), 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 was the source of radiation, ionization chamber detected the amount of radiation transmitted through the samples while the lead collimator was used 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. The counting process involved 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 the ionization chamber. Samples were subsequently placed between the X-ray source and the detector at linear collimator geometry under open wheel (without filter). X-ray source of energies 40KeV to 120KeV were irradiated on each sample for 20 seconds by this procedure, the intensity of the direct (I_o) and the transmitted (I) flux were determined and the counting was recorded by the electrometer. Counting was done three times for each sample and the later average values were calculated. A block diagram of the passage of radiation photon through shield is shown below:

X-ray Source   Collimator   Shield   Collimator   Detector
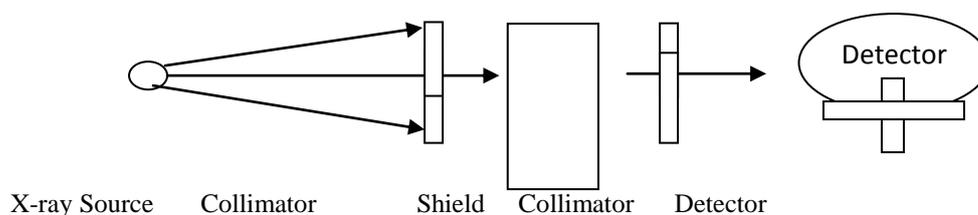
Figure 1: Block diagram of the passage of radiation photon through shield to detector.

III. Results and Discussion

Table 1, 2, 3, 4 and 5, respectively shows percentage composition of heavy metals and CEC in the clay sample used for this work, linear attenuation coefficients, transmitted intensity and absorption thickness for concrete, natural clay sample and those that were artificially contaminated with nitrate and oxide of heavy metals such as lead and Iron.

The linear attenuation coefficients calculated for all the samples at energies of 40keV to 120keV at 10 mA are shown in Table 2. As the x-ray energy increases, the linear attenuation decreases. Higher energy photons are able to penetrate a material more deeply compared to a lower energy photon as a result of interaction. This conformed to the Lambert-Beer idea that a good shielding material attenuates photons that pass through it. The attenuation characteristic behaviour of natural clay and artificially contaminated clay follow the same trends.

Table 3 shows the transmitted intensity that passed through the clay slabs at different energies. As the energy increases, the intensity that passed through the clay slabs increases. It can be said that the changes in chemical composition with the physical nature of the material is responsible for the response of the samples to the reduction in intensity of the X-ray radiation. It was also observed that as we increase the thickness of the clay slabs the amount of photon energy per unit second per unit surface area decrease. This is as a result of interaction between photons and the clay slabs. This indicates that the attenuation coefficient depends on the chemical and structural composition of clay samples. In other words, attenuation coefficient depends on the exchangeable cations in the clay which leads to increase in density of the clay material.

Table 1: Table1: Percentage composition of heavy metals present in the Omi – Adio clay sample used for this work.

Heavy metal	Cr	Ni	Cod	Mn	Pb	Cu	Zn	others
Percentage composition	5.70%	3.52%	0.28%	61.34%	1.48%	5.76%	16.98%	4.94%

Table 2: Linear attenuation coefficient of natural clay slab, clay mixed with nitrate and oxide of lead and iron at 6400ppm and concrete.

	Linear attenuation coefficient cm^{-1}				
Energy	Natural clay	Clay mixed with PbN03	Clay mixed with PbO	Clay mixed with FeNO3	Concrete
40	0.55	2.26	2.79	2.15	0.61
60	0.43	1.54	1.87	1.46	0.49
80	0.39	1.20	1.46	1.15	0.44
100	0.35	1.00	1.21	0.96	0.39
120	0.34	0.86	1.05	0.83	0.35

Table 3: Intensity that passed through the clay slabs of one centimetre thickness at different energies with machine error, ± 0.01

Energy	I_o	I_{oc}	I_{PbNO3}	I_{PbO}	I_{FeNO3}	I_{con}
40	33.03	19.05	3.46	2.06	3.84	17.94
60	117.40	76.37	25.17	18.09	27.26	71.92
80	229.90	155.65	69.25	53.39	72.79	148.06
100	369.00	260.03	135.75	110.04	141.28	249.83
120	540.00	384.35	228.51	188.93	235.47	380.53

Abbreviation: I_o - Incident intensity; I_{oc} - Intensity that passed through natural clay; I_{PbNO3} - Intensity that passed through clay contaminated with lead nitrate; I_{PbO} - Intensity that passed through clay contaminated with lead oxide; I_{FeNO3} - Intensity that passed through clay contaminated with nitrate of Iron; I_{con} - Intensity that passed through concrete.

Table 4: Variation of attenuated intensity with thickness of natural clay slabs at different energies for natural clay slabs.

Thickness	I_o=33.03 Wm^{-2} Trans. Int at 40kev	I_o =117.40 Wm^{-2} Trans. Int at 60kev	I_o = 229.90 Wm^{-2} Trans. Int at 80kev	I_o = 369.00 Wm^{-2} Trans. Int at 100kev	I_o = 540.00 Wm^{-2} Trans. Int at 120kev
1	19.06	76.37	155.64	260.03	392.12
2	10.99	49.68	105.39	183.24	284.74
3	6.34	32.32	71.35	129.13	206.76
4	3.66	21.02	48.31	90.99	150.14
5	2.11	13.67	32.71	64.12	109.02
6	1.22	8.89	22.15	45.19	79.17
7	0.7	5.79	14.99	31.84	57.49
8	0.41	3.76	10.15	22.44	41.74

9	0.23	2.45	6.87	15.81	30.31
10	0.14	1.5	4.65	11.19	22.01

Table 5: Attenuated intensity with thickness of natural clay, artificially contaminated clay and concrete at 100 keV x-ray energy.

	Transmitted Intensity (Wm^{-2}) at 100kev				
Thickness	Natural clay	Concrete	Clay with $Fe(NO_3)_3$	Clay with $Pb(NO_3)_2$	Clay with PbO
0	369.00	369.00	369.00	369.00	369.00
1	260.03	249.83	141.29	135.75	110.03
2	183.24	169.15	54.1	49.94	32.81
3	129.13	114.53	20.71	18.37	9.78
4	90.99	77.53	7.93	6.76	2.92
5	64.12	52.5	3.04	2.49	0.87
6	45.19	35.54	1.16	0.91	0.26
7	31.84	24.06	0.45	0.34	0.08
8	22.44	16.29	0.17	0.12	0.02
9	15.81	11.03	0.07	0.05	0.01
10	11.19	7.47	0.03	0.02	0.00

The fraction of the photon energy that interacts in the 1-cm thickness is presented in Table 2 and graphical representation of the value of the linear attenuation coefficient of clay material and concrete are shown in figure 1. Figure 1 shows the relationship between the attenuation coefficient for natural clay, artificially contaminated clay and concrete and photon energy. This graph shows two significant features of the relationship. One; the linear attenuation coefficient value decreases rapidly with increase in photon energy. Generally, the probability of photoelectric interactions is inversely proportional to the cube of the photon energy (Cember, 1996). This general relationship can be used to compare the attenuation coefficients at five different photon energies as shown in Table 2. The significant point is that the probability of photoelectric interactions occurring in a given material drops drastically as the photon energy is increased. More so, the rate at which photons interact is determined by the energy of the individual photons and the nature of the material.

Figure 2 describes the interaction between clay materials (natural and contaminated clay) and energy of x-ray beam. Five different beams of energy: 40 keV, 60 keV, 80 keV, 100 keV and 120 keV were used for this plot. The thickness of the attenuator was kept constant while beam energy is varied so that the attenuator thickness effect may be excluded. The figure showed that the intensity increases exponentially as x-ray energy increases. It means that photon energy can easily penetrate clay of low atomic number and low density. However as the beam energy increases, the intensity appears to increase more rapidly. This shows that intensity is affected by the interaction of x-rays with matter.
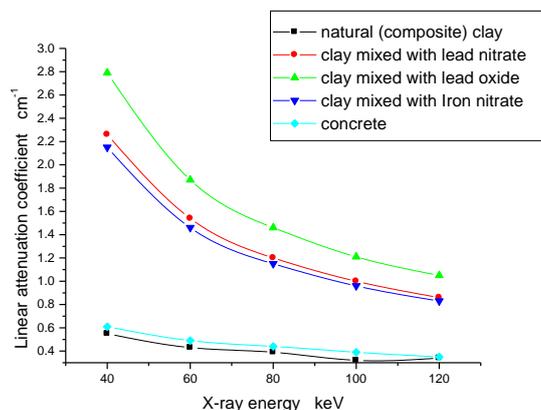
Figure 1: Linear attenuation coefficient of natural and contaminated clay versus x-ray energy for selected shielding types.
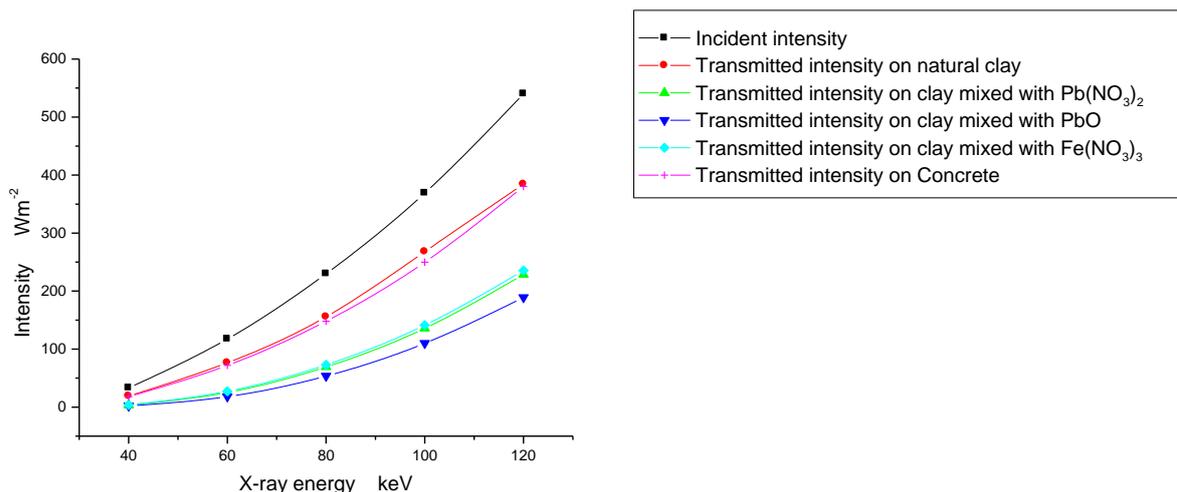
Figure 2: Graph of intensity of natural and artificially contaminated clay against x-ray energy.
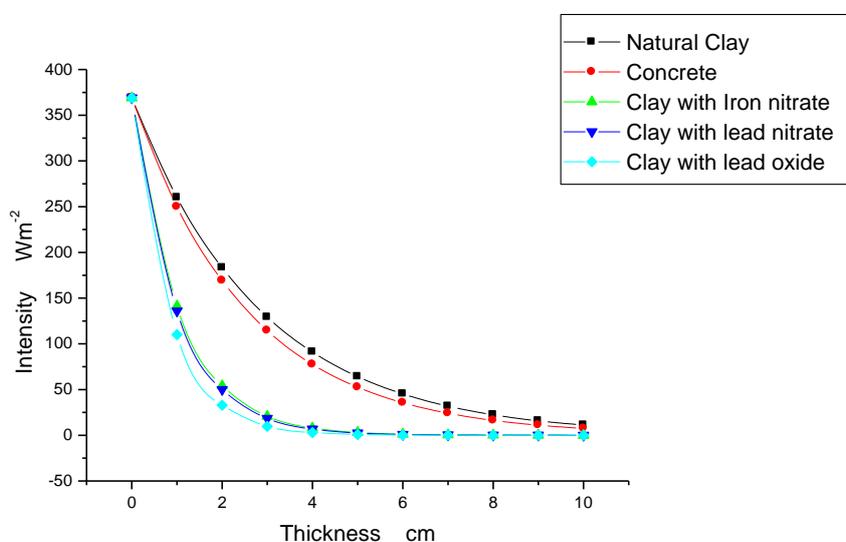
Figure 3: Graph of Intensity against the thickness of concrete, natural and artificially contaminated clay slab at 100kev

We noticed that when the linear attenuation coefficient has a low value the curve decreases relatively slowly and when the linear attenuation coefficient is large the curve decreases very quickly as shown in figure 3. It means that linear attenuation coefficient is the characteristic of individual absorbing materials. Natural clay has a small value and is easily penetrated by x-rays as a result of low density of high absorbing element such as lead. Linear attenuation coefficient can thus be associated with the distribution of intensity with the absorber thickness i.e with the steepness of the distribution curve. It is obvious that clay with lead oxide has a relatively large linear attenuation coefficient and is relatively good absorber of radiation.
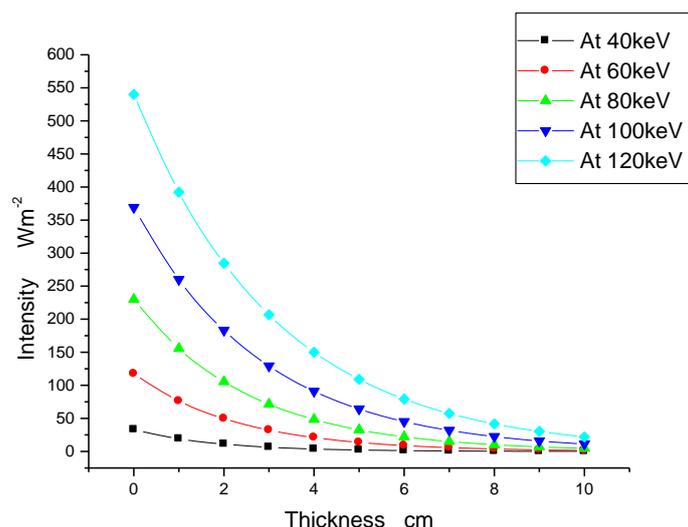
Figure 4: Graph of intensity against thickness of natural clay slabs at different energies.

Effect of Cation exchange on density of the material

Generally, clay surface adsorbs specific amount and type of cations under a given environmental conditions such as temperature, pressure, pH and pore water chemistry (Bradl, 2002). The adsorbed cations get partly or fully replaced by ions of another type subject to changes in the environmental condition. Such changes alter the physico-chemical characteristics of soil (Bradl, 2002) and hence, its mass concentration (density). This process of cation exchange was explored in this work and deduces that attenuation of x-radiations increase as the concentration (density) of cation attached at the surface of clay increases. The cation exchange capacity of clay used for this work was 15.2 meq/100g. The result of linear attenuation coefficient of artificially contaminated clay ranges from 2.15 cm^{-1} to 2.78 cm^{-1} while natural clay was 0.55cm^{-1} as seen in Table 2.

IV. Conclusion:

All materials tested in this study demonstrated effectiveness in radiation attenuation at 40 keV to 120 keV. Artificially contaminated clay was best able to shield ionizing radiation of lower and higher energy. In fact, the artificial contaminated clay material attenuated ionizing radiation better than concrete at lower (by 37 %) and higher energies (28 %). At various levels of energy, all the materials (concrete, natural and artificially contaminated clay) have been shown to predictably reduce X-ray transmission as a function of thickness. It is found that clay contaminated with lead oxide has greatest linear attenuation coefficient. Therefore, we conclude that artificially contaminated clay is an effective substitute for concrete as x-ray walling material.

Acknowledgement

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