

## Assessment of Shielding Potentials and Radiological Safety Indices of Nigerian Granite Rocks

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### Abstract

The activity concentration of primordial radioisotopes ( $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) in granite rock samples from Minna, North central Nigeria were measured using a sensitive HPGe detector for gamma spectrometric measurement. Five granite rock samples were collected from Maikunkele, Bosso, Maitumbi, Chanchaga and Paiko areas of the town. The average activity concentrations for the radionuclides in the rocks were:  $27 \pm 4$  Bq/kg;  $48 \pm 7$  Bq/kg and  $874 \pm 86$  Bq/kg for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively. The average absorbed dose rate, effective dose; radium equivalent ( $R_{eq}$ ) and internal hazard index ( $H_{in}$ ) were  $65 \pm 10$  nG/h;  $0.32 \pm 0.05$  mSv/y;  $140 \pm 21$  Bq/kg and  $0.45 \pm 0.07$  respectively. All the rock samples had  $R_{eq}$  less than the recommended safety value of 370 Bq/kg. All evaluated radiological safety indices fell within the recommended safety limits and world average values. The analysis of radionuclide content of the granite rocks showed that they do not pose environmental radiation risk to humans when used as structural shielding materials. Mass attenuation coefficients of the granite samples evaluated via the WinXcom computer code suggested that the granite rocks considered have good shielding capacity comparable to that of ordinary concrete.

**Keywords:** Radionuclide; radiation effect; shielding; dose assessment; granite

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### Introduction

Over the years, research in nuclear science and technology has been very active. This is mainly due to the wide applications of nuclear energy in diverse field such as agriculture, medicine and in industries (Olarinoye *et al.*, 2010). One major problem associated with the use of radioisotopes and the ionising radiations that emanate from them is the routine and sometimes accidental radiation exposure which subsequently results into clinical symptoms. Although, nuclear energy presents immense benefits for mankind, uncontrolled exposure of man and the biota to components of nuclear radiations can lead to clinical symptoms or even death depending on the radiation quality and dose. Consequently, for man to continue to harvest the benefits nuclear energy and technology provides, adequate protective measures would have to be in place in all nuclear facilities.

One of the cardinal principles of radiation protection recommended by the ICRP (1990) is radiation shielding. Shielding is the most reliable of all radiation protection methods. It involves confining the nuclear radiation to a volume of space. Shielding requires no administrative control unlike other forms of radiation protection. The effectiveness of any shield depends on the nature of material it is made up of and the radiation parameters such as quality factor, Linear Energy Transfer (LET) and dose (Ogundare and Olarinoye, 2016). Traditionally, high quality structural shields are usually constructed using concrete

made from weathered or quarried rocks, cement and water (NCRP, 1976; 2005; Obande, 2002). However, geological formations such as granite rocks, rivers, riverbed soils, soils and sediments have been established to contain Naturally Occurring Radionuclide Materials (NORM). These NORM are the main source of radiation exposure to man from natural sources (Olarinoye *et al.*, 2010). Thus, the use of rocks as constituents of structural shielding concretes for ionising radiation should possess two basic properties: firstly, such rocks should have enough density to produce dense concrete for the primary purpose of attenuating radiation and secondly, the rocks must not constitute a secondary source of radiation via the NORM present in them.

In Nigeria, due to their abundance especially in the Northern part of the country, granitic rocks are quarried in many places for different construction purposes. Furthermore, their texture, structural properties and mineralogical composition are other reasons why granite rocks are quarried in different part of Nigeria where the outcrops are abundant. Due to their radiological content, the use of granites for construction for homes and radiation shield may not be wise except their radiological content are confirmed to be below safety limits. It is thus important to know the relative shielding quality of available granites and their radioactive contents. This will ensure the right choice is made when considering different granites from different locations for radiation

shielding applications or for construction of homes in general. Furthermore, the increasing energy crises in Nigeria has informed suggestions and researches about alternative electrical energy sources. Nuclear energy is one of such alternatives that has been mentioned even in official quarters and presently being considered. In the future, Nigeria may eventually join the comity of nations benefitting from the peaceful use of nuclear energy. The use of local materials such as granite rocks whose radiological content is within permissible level and with high radiation shielding capacity for neutrons and photons in nuclear energy facilities will be an economic asset for the country. Although, radiological composition of some Nigerian granites has been investigated (Shittu *et al.*, 2015; Gbadebo, 2011; Odunaike *et al.*, 2008), most of these studies however have focused on granites from southern part the country and on their radiological safety when used as building material. Radiological study of rocks from the northern part of Nigeria and their shielding capacities is yet scarce in the literature. This research is undertaken to measure the NORM ( $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) concentrations and evaluate the relative photon shielding capacity of granite rocks from Minna area, north central Nigeria.

## Materials and Methods

### Study Area

Minna is the capital of Nigeria's north central state of Niger and located between longitude  $6^{\circ}43'\text{E}$  and  $6^{\circ}45'\text{E}$  and latitude  $9^{\circ}24'\text{N}$  and  $9^{\circ}43'\text{N}$ . It is located on the central portion of the Nigeria basement complex. In Minna area, five lithostratigraphic units have been recognized (Figure 1). The schist occurs like a flat laying narrow belt at the middle part of the area with small quartzite ridge parallel to it. Small suites of gneiss occur at the southern and northern section of the area forming a contact with the granite and pegmatite rich in feldspar is bounded to the east (Olarinoye *et al.*, 2010; Alabi, 2011). Granite rocks are the most dominant rock type in Minna with varying texture and composition. The rocks appear as outcrops, batholiths and ridges which are visible around the area.

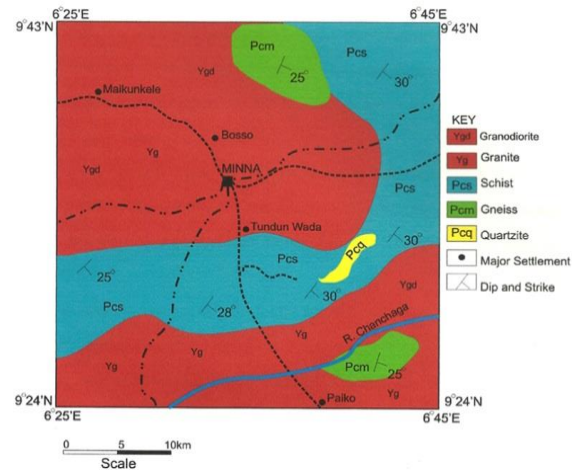


Figure 1. Geology map of Minna and environs.

### Collection and Preparation of Samples

Granite rocks were collected from 5 different locations in Minna area- Maikunkele, Bosso, Maitumbi Chanchaga and Paiko. Five samples were collected from the visible outcrops and weathered rocks in these areas. The 5 rocks were well labelled and taken to the laboratory where they were crushed and milled using a disc mill and also pulverised. The pulverised granites from the locations were labelled as MK, BS, MT, CH and PK for granite samples from Maikunkele, Bosso, Maitumbi Chanchaga and Paiko respectively.

### Gamma-spectrometry

The pulverised granite rocks were collected and stored in 1 litre Marinelli beakers. The beakers were closed using screw caps and mastering tapes were wrapped over the beaker caps to make them air tight and also prevent radon gas and its daughter from escaping out of the beakers. The sealed samples were kept for 32 days to allow for secular equilibrium to be reached. After this period, a sensitive HPGe detector was used for gamma spectrometric measurement of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in the samples. The detector was located inside a cylindrical lead shield of 24 cm internal diameter, 60 cm in height and 5 cm thick. The lead shield was lined with layers of cadmium and copper, each of 3 mm thickness to reduce background radiation from external sources. The relative efficiency of the detector was 40 per cent with a resolution of 1.85 keV (FWHM) for the 1.33 MeV gamma- transition of a  $^{60}\text{Co}$  source. The efficiency and energy calibrations of the detector system were done using standard sources from QSA Global GmbH (DKD-3) Germany. The sources which

contain 10 radionuclides ( $^{57}\text{Co}$  (122.05 keV);  $^{139}\text{Ce}$  (165.86 keV);  $^{109}\text{Cd}$  (88.03 keV);  $^{203}\text{Hg}$  (279.20 keV);  $^{113}\text{Sn}$  (391.69 keV);  $^{85}\text{Sr}$  (514.01 keV);  $^{137}\text{Cs}$  (661.66 keV);  $^{60}\text{Co}$  (1173.2 keV and 1332.5 keV);  $^{241}\text{Am}$  (59.54 keV); and  $^{88}\text{Y}$  (893.04 keV and 1836.1 keV)) with known energies and activities were prepared in a 1000 ml Marinelli beaker. The energy calibration sources were counted for 10 hrs to produce well defined photopeaks.

The samples were analysed by acquiring spectrum for each of the sample for 36000 s. A MAESTRO-32 computer program was adopted for the accumulation and analysis of the gamma energy spectra of the radionuclides. The activity concentrations of  $^{238}\text{U}$  was determined indirectly from the photopeak lines of 351.9 keV of  $^{214}\text{Pb}$  and 609.32 keV of  $^{214}\text{Bi}$  while the  $^{232}\text{Th}$  activity was estimated from the gamma ray peaks of  $^{212}\text{Pb}$  at 238.6 keV,  $^{208}\text{Tl}$  at 583.78 keV and  $^{228}\text{Ac}$  at 911.21 keV. The activity concentration of  $^{40}\text{K}$  was evaluated directly from the 1460.8 keV photon emission line. The background gamma spectrum obtained with similar conditions for both the standard and sample measurements was used for accurate corrections of the evaluated sample activity concentrations. The concentrations of activity  $A$  (Bq/kg) of the three radioactive nuclides of interest present in each granite sample was estimated using the empirical expression (Odunaike *et al.*, 2008):

$$A = \frac{N_E}{\varepsilon M t p} \quad (1)$$

where  $N_E$  is the total area under a peak at the energy of interest,  $\varepsilon$  is the efficiency of the detector at the energy,  $M$ ,  $t$  and  $p$  are the sample mass, counting time and emission probability of radionuclide of interest respectively.

### Energy Dispersive X-ray Fluorescence Analysis

The concentrations of trace and major elements in the granite rocks were determined through the Energy Dispersive X-ray Fluorescence (EDXRF) spectrometric Analysis. For this analysis, the pulverised rock samples were sundried for 96 hours until a near constant weight was obtained. Weighed 0.02 kg of the pulverised and dried granite rocks were mixed with binder (PVC dissolved in toluene) carefully and pressed into circular

pellets in a hydraulic press under a pressure of about 20 tonn. The pellets each of diameter about  $30 \pm 3$  mm were then loaded into the sample chamber of a PAN analytical Minipal4model PW4025/45B EDXRF spectrometer for trace and major elemental oxide analysis.

### Results and Discussion

The average activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  present in the 25 granite samples from 5 different locations in Minna are given in Figure 2. All three primordial radionuclides were present in all the granite samples analysed with average of  $27.1 \pm 4.1$  Bq/kg;  $48 \pm 7.2$  Bq/kg and  $574.5 \pm 86.21$  Bq/kg for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively. Being the most abundant in nature amongst the three primordial radionuclides under consideration,  $^{40}\text{K}$  presents the highest concentration in all the granite samples. The concentrations of  $^{40}\text{K}$  was more than ten times the concentrations of  $^{232}\text{Th}$  and more than 20 times greater than the concentration of  $^{238}\text{U}$  in nearly all the investigated granites. The highest concentrations of  $^{238}\text{U}$  and  $^{40}\text{K}$  were found in Chanchaga granite while the highest measure of  $^{232}\text{Th}$  was from Paiko granite. Although the mean concentrations of the radionuclides were all less than the world average concentrations in granite (Table 1). Table 1 also shows the average concentration values of the three nuclides in comparison to those obtained in other areas. It is obvious that the radionuclides concentrations in Minna is relatively lower than those from the areas included in the table. The differences in the mineral composition and conversely the geology of these areas may be part of the factors responsible for the observed differences in their radionuclide concentrations.

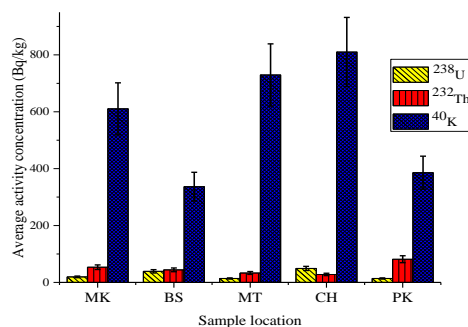


Figure 2. Activity concentration in the granite samples

Table 1. Comparison of Radiological parameters of granite rocks from other locations.

Country	<sup>238</sup> U (Bq/kg)	<sup>232</sup> Th (Bq/kg)	<sup>40</sup> K (Bq/kg)	Ra <sub>eq</sub> (Bq/kg)	D <sub>in</sub> (nGy/h)	H <sub>in</sub>	E <sub>in</sub> (mSv)	ELCR (X10 <sup>-3</sup> )	I <sub>γ</sub>	Ref.
Nigeria (Minna)	27	48	575	140	65.48	0.45	0.32	0.281	0.52	This work
Pakistan (Nagarparkar)	26	42	867	152.4	139.24	0.45	0.68	2.4	0.58	(Qureshi <i>et al.</i> , 2014)
Africa	23	42	811	145.38	132.24	0.46	0.65	2.27	0.56	(Tzortzis <i>et al.</i> , 2014)
Egypt (Wadi Karim)	56	54	4849	506.14	498.84	1.52	2.45	8.57	2.07	(El-Arabi, 2007)
Ghana (Kasoa)	76	48	731	200.8	181.2	0.75	0.89	3.11	0.74	(Otoo <i>et al.</i> , 2011)
Iran	130	83	1287	347.57	313.86	1.29	1.54	5.39	1.28	(Jahangiri and Ashrati, 2011)
Turkey	125	205	1172	558.01	480.26	1.98	2.36	8.25	2	(Orgun <i>et al.</i> , 2005)
Yemen	54	127	1743	369.51	328.82	1.14	1.61	5.65	1.4	(Abd El-Mageed <i>et al.</i> , 2011)
European Union	78	89	1049	285.84	253.58	0.98	1.24	4.36	1.05	(Trevisi <i>et al.</i> , 2012)
USA	57	69	1140	243.26	219.54	0.81	1.08	3.77	0.92	(Kitto <i>et al.</i> , 2009)
World average (Granite)	81	105	1111	317.66	279.64	1.08	1.37	4.8	1.17	(Qureshi <i>et al.</i> , 2016)
Limit values				370	84	<1	2	0.29	<1	(UNSCEAR, 2000)

The radium equivalent activity ( $Ra_{eq}$ ) can be used to compare materials having different values of <sup>238</sup>U ( $A_U$ ), <sup>232</sup>Th ( $A_{Th}$ ) and <sup>40</sup>K ( $A_K$ ). It is given as (Brekta and Matthew, 1985; Ibrahim, 1999):

$$Ra_{eq} \left( \frac{Bq}{kg} \right) = A_U + 1.43A_{Th} + 0.077A_K \quad (2)$$

Equation (2) considered the fact that 1 Bq/kg of <sup>238</sup>U (<sup>226</sup>Ra), 1.43 Bq/kg of <sup>232</sup>Th and 0.077 Bq/kg of <sup>40</sup>K yield equal absorbed (gamma) dose rates (Brekta and Matthew, 1985; Ibrahim, 1999). Consequently, the radium equivalent activity is a single and simple index that can be used to compare the specific activity concentrations in materials containing different concentrations of the three radionuclides and hence producing different radiation exposure. According to the UNSCEAR (2000) report,  $Ra_{eq}$  values is expected to be less than 370 Bq/kg if the material is to be used as a building material. The calculated  $Ra_{eq}$  for the rocks are presented in Table 1. The average measure of  $Ra_{eq}$  for the areas studied are: 142.88, 128.02, 117.46, 151.54, and 160.09 Bq/kg for MK, BS, MT, CH and PK respectively with an overall average value of  $140 \pm 21$  Bq/kg all of which are lower than the world average in granites and subsequent studies conducted in other countries (Table 1).

#### Absorbed Radiation Dose rate (D)

The rate of absorbed doses (D) in air associated with the three primordial radioactive nuclides present in the granites

were evaluated in terms of their respective concentrations according to the expression (UNSCEAR, 2000):

$$D \left( \frac{nGy}{h} \right) = 0.462C_U + 0.604C_{Th} + 0.0417C_K \quad (3)$$

where  $C_U$ ,  $C_{Th}$ , and  $C_K$  are the measured activity concentrations in Bq/kg of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K respectively in the granite samples (EC, 1999). The average values of the absorbed dose rates of the rocks are presented in Table 1. The minimum value of 56.86 nGy/h and maximum value of 73.39 nGy/h were obtained in granite samples from MT and CH respectively. The dose rates when compared with those obtained for granites in different areas of the world and the world mean dose rate value, show that the dose rates are lower in Minna granites.

#### Internal annual effective dose rate ( $E_{in}$ )

The internal effective dose rate ( $E_{in}$ ) due to the measured activities of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the granites was calculated according to the expression (UNSCEAR, 2000; EC, 1999):

$$E_{in} (mSv/yr) = D \left( \frac{nGy}{h} \right) \times 8760h \times 0.70 Sv/Gy \times 0.8 \quad (4)$$

This is the annual effective dose at 1 m high above the ground level in air using a conversion factor of 0.7 Sv/Gy and an indoor occupancy factor of 0.8 (UNSCEAR, 2000). The  $E_{in}$  estimated for Minna granite varies between 0.28 mSv/yr and 0.36 mSv/yr with an average value of 0.32 mSv/yr. The average value of the effective dose rate obtained in this study is less than the worldwide granite

average of 1.37 nSv/y and the safety limit value of 1 mSv/y for non-occupational exposure (UNSCEAR, 2000; Qureshi *et al.*, 2016).

### Internal Hazard Index ( $H_{in}$ )

Inhalation of radon gas and its short-lived progeny is the major source of internal radiation exposure (UNSCEAR, 2000). The internal hazard index is a quantity that measures the degree of internal exposure to the cancer causing radon and its daughters. The  $H_{in}$  for the considered granites were evaluated using the equation (Gbenu *et al.*, 2016):

$$H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (5)$$

For radiological safe material,  $H_{in}$  should be less than unity. Consequently, the granites under study are considered to be safe since their average  $H_{in}$  were found to be 0.45.

### Gamma Concentration Index ( $I_\gamma$ )

The  $I_\gamma$  is an index used to describe natural radionuclide in building materials. It is a value

that indicates if the annual dose as a result of exposure to external gamma radiation in a specific material goes beyond 1 mSv. The gamma index of all granite stones was evaluated through the equation (Gbenu *et al.*, 2016):

$$I_\gamma = \frac{C_U}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad (6)$$

If the annual effective dose is to be less than 1 mSv, then  $I_\gamma$  should be less than unity. The overall average value of the gamma index was 0.52 with a range between 0.46-0.58 (Table 1). According to the European Commission (EC, 1999), a building material with gamma index  $\leq 0.5$  (which corresponds to an annual effective dose of 0.3 mSv to the public) should be exempted from all restriction with respect to its radioactivity. However, if  $0.5 \leq I_\gamma \leq 1$ , then constraint should be set. The fact that the gamma index of the stones is less than unity is an indication that the stones are safe when used as building material or for radiation structural shielding.

Table 2. Major oxides (wt%) and trace elements (ppm) in the collected granites rocks.

Sample	Major Oxides Concentration (wt%)										Trace Elements Concentration (ppm)				
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	CaO	Fe <sub>2</sub> O <sub>3</sub>	MgO	MnO	P <sub>2</sub> O <sub>5</sub>	TiO <sub>2</sub>	Cr	Cu	As	Ni	Zn
MK	71.9	13.8	2.2	2.3	1.8	3.7	0.3	0.1	0.1	0.4	41.6	19.7	19.2	23.7	12.8
BS	69.6	13.8	2.1	2.3	1.9	3.5	0.4	0.1	0.1	0.2	25.5	12.66	26.3	25.9	22.6
MT	71.2	14.2	2.3	2.4	1.9	3.6	0.3	0.1	0.1	0.4	46.8	17.91	15.9	22.6	14.9
CH	69.8	14.2	2.3	2.4	1.9	3.6	0.3	0.1	0.1	0.3	28.1	10.2	18.7	36.9	17.5
PK	68.7	14	2.2	2.3	1.9	3.6	0.3	0.2	0.1	0.1	57.8	21.8	31.4	20.5	27.8

### Excess Lifetime Cancer Risk (ELCR)

Beside hereditary and exposure to background radiation, excessive exposure to ionising radiation and other cancer causing agents can increase the chances of someone developing one form of cancer or the other. The Excess Lifetime Cancer Risk (ELCR) indicates the additional risk associated with excessive exposure radiation and other cancer causing agent that could lead to cancer in an individual. The ELCR can be estimated from the indoor annual effective dose that one receives from one's indoor environment in which one spends about 80% of one's lifetime. The ELCR was calculated according to the equation (Gbenu *et al.*, 2016; UNCEAR, 2000):

$$ELCR = A \times LE \times RF \quad (7)$$

where A, LE, and RF are the indoor annual effective dose equivalent (mSv/y), life expectancy (70 years) and the fatal risk factor per Sievert (0.05) respectively. The average ELCR varies from  $0.98 \times 10^{-3}$ - $1.26 \times 10^{-3}$  with an overall average value of  $1.12 \times 10^{-3}$  for the

granite stones. The average ELCR is lower than the safety limit of  $1.16 \times 10^{-3}$  (Qureshi *et al.*, 2016). However, granite from CH and PK had ELCR higher than the limit values and presents higher risk of inducing cancer for a person living for 70 years in the dwelling made from them by a factor of 1.09 and 1.06 times respectively.

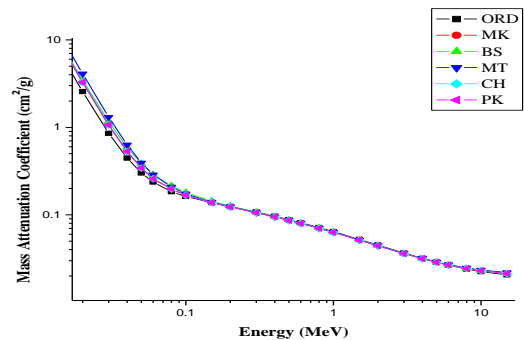


Figure 3. Mass attenuation Coefficients of the granite rocks and ordinary Concrete.

### Mass Attenuation Coefficient

The major oxides and trace elements concentrations in the granite rocks obtained from the EDXRF analysis is shown on Table 2. Generally, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O and Na<sub>2</sub>O concentrations account for more than 80% of major oxides present in the rocks, while CaO<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, MgO, P<sub>2</sub>O<sub>5</sub> and TiO<sub>5</sub> account for less than 10% of the granite oxides concentration. Only five trace elements (Cr, Cu, As, Ni, and Zn) were detected in the rocks with varying concentration (ppm). The variations in the elemental concentrations in the rocks can be attributed to the difference in their mineral contents. The chemical compositions of the granite rock samples were used to evaluate their mass attenuation coefficients for photon energies of 0.015-15 MeV through the use of the WinXcom software (Gerward *et al.*, 2004). The mass attenuation coefficients (MAC) of the 5 granite samples and that of ordinary concrete (ORD) with density of 2.30 gm<sup>-3</sup> (ANSI/ANS 6.4.3, 1991) with respect to photon energy (0.015-15 MeV) is shown in Figure 3. The elemental concentration (by weight) of the concrete are as follows: H=0.94%; C=0.09%; O=53.66%; Na=0.46%; Mg=0.12%; Al=1.32%; Si=36.74%; S=0.08%; K=0.31%; Ca=5.65% and Fe=0.63%. From Figure 3, the MAC of the concrete increased with photon energy implying more penetration in the material with increasing energy. The MAC of the rocks were comparable within the energy spectrum considered with no noticeable difference. This is majorly due to the similarities in their elemental composition. The MAC of the rocks were slightly higher than that of ORD in the lower end of the energy spectrum and almost the same beyond 0.05 MeV. Consequently, it is safe for one to conclude that the granite rock from Minna Area of Nigeria can be a choice for good structural shield material for photon beam with energy considered.

## Conclusion

The primordial radiological content, safety indices and mass attenuation coefficient of granite rocks from Minna area of Nigeria have been investigated. The average activity concentration values for the radioisotopes in the rocks were: 27 ± 4 Bq/kg; 48 ± 7 Bq/kg and 874 ± 86 Bq/kg for <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K respectively. The average absorbed dose rate, effective dose (annual); radium equivalent ( $R_{eq}$ ) and internal hazard index ( $H_{in}$ ) were 65 ±

10 nG/h; 0.32 ± 0.05 mSv/y; 140 ± 21 Bq/kg and 0.45 ± 0.07 respectively. All the rock samples had  $R_{eq}$  values less than the recommended safety limit of 370 Bq/kg. The analysis of the radionuclide content of the granite rocks showed that they do not pose environmental radiation risk to humans when used as structural shielding materials. Mass attenuation coefficients of the granite samples were comparable to one another and that of ordinary concrete due to similarity in chemical compositions. The granite rocks considered have good shielding capacity comparable to that of ordinary concrete, thus, they can be a good choice of structural shielding material for photon beams within the photon energies investigated.

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