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Natural Radioactivity Associated with Mining of Rare Metal Pegmatite of Oke-Ogun Field, Sepeteri, Southwestern, Nigeria

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ABSTRACT

Within and around major cities of Southwestern Nigeria, there are scores of mining activities whose constitute menace into the immediate environments. In this study, a combined techniques using a portable survey meter (Gamma-Scout SN 038439) for in-situ investigation and a well-calibrated NaI(Tl) detector system were used for the measurement of the average radioactivity concentration in some rocks associated with mining of rare metal pegmatite of Oke-Ogun field, Sepeteri, Oyo state of Nigeria. The average dose rate obtained for gamma and beta exposure vary between 0.1 – 0.24 μSv hr^{-1} with a mean of 0.18 μSv/hr and between 0.01 – 0.08 μSv/hr with a mean of 0.03 μSv hr^{-1} respectively. With the exception of ^{40}K and the anthropogenic ^{137}Cs, all the radionuclides detected are traceable to the pre-medial series of ^{238}U and ^{232}Th. The specific activity value obtained for ^{238}U, ^{232}Th and ^{40}K varied from 26.06 to 97.10 Bq kg^{-1} with a mean value of 58.04 ±12.78 Bq kg^{-1}, 17.65 to 85.65 Bq kg^{-1} with an average of 29.04 ± 9.49 Bq kg^{-1} and 194.87 to 1035.56 Bq kg^{-1} with an average of 586.23 ± 88.56 Bq kg^{-1} respectively. The mean value obtained for the Representative levels index (I_γ), the radium equivalent (Ra_{eq}), the total absorbed dose rate (ADR) were 1.07; 144.71 Bq kg^{-1} and 68.80 nGy h^{-1} respectively. The discrepancies of our data can be attributed to several factors such as geological formation, transport process, etc. Although our results are just some fractions of the international standard limit, but still within the same ranges when compared with those obtained elsewhere. This results also will serve as a baseline data for future investigations.

Keywords: *Radioactivity, Mining, Hazards, Gamma Spectrometry*

1. INTRODUCTION

Radiation of natural origin at the earth's surface consists of two components namely cosmic rays and radiation from the radioactive nuclides in the earth's crust. The latter component is the terrestrial radiation, which mainly originates from the so-called primordial radioactive nuclides that were made in the early stage of the formation of the solar system. Long-lived radioactive elements of interest, which are the main elements contributing to natural terrestrial radioactivity (UNSCEAR, 2000) include uranium, thorium and potassium and any of their radioactive decay products, such as radium and radon. The radioactive waste generated in mining and milling activities, especially those involving uranium and thorium (U, Th) ores, differs from that generated at nuclear power plants and most other industrial operations and medical facilities.
They contain only low concentrations of radioactive material but it is generated in large volumes (IAEA, 2002). These elements have always been present in the earth's crust and within the tissues of all living species. Studies of terrestrial natural radiation are of great importance for various reasons. They serve as useful tracers for atmospheric variation studies. It is usually realized that natural environmental radiation mainly depends on geological and geographical conditions. Also, an understanding of natural radioactivity was used to determine the age of the earth using the radioactive decay of ^{238}U to ^{206}Pb and ^{235}U to ^{207}Pb. The heat produced by radioactive disintegration is an important source of thermal energy that brings about metamorphism. Radioactivity is also employed in geological mapping, since different rock types can be recognized from their distinctive radioactive signatures(Jamal, 2002). It is also significant in its application to environmental problem such as detection of radioactive pollution in soils, groundwater, and air. The hazards to humans or to the environment posed by mining and milling waste arise not only from its radioactivity but also from the presence of toxic chemicals and other materials in the waste. Achieving a consistent regulatory approach to protect against these different hazards is a challenge for national regulators. Radioactivity and radiation levels in various environmental samples have been of great concern in many countries. Cothern et al.,(1986) reported the occurrence of Uranium in drinking water in U.S.A. Other workers(Hess et al.,1983; Horton,1984; Prylibski.1999; Prylibisk and Zebrowski, 1999; Osibote et al., 1999; Tchokossa et al., 1999; Oyawale et al., 2003; Tchokossa, 2006) have published data obtained from monitoring communities matrices in various countries.

In recent times, there has been resurgence of interest in the Nigerian pegmatite occurrence because of their associated rare metal and gem mineralisation. These have led to concentration of study on discrimination of the pegmatites into the rare metal

mineralised and barren ores in order to elucidate modes and features of mineralisation (Matheis et al.,1982; Matheis and Vachette, 1983; Garba, 2003). Nigeria has long, but discontinuous history of mining and the country is a prominent exporter of 34 minerals including Tin, Tantalite, Coal and Columbite, among others. Mining is administered through the Ministry of solid minerals development, which is promoting the participation of private sector in the exploitation of solid minerals in different parts of Nigeria. Private sector and artisanal miners driven by high prices of these minerals have made more discoveries, and as such, massive and widespread mining activities have been going on in the country. The result of this is destruction of natural landscape and soil denudation. Sepeteri is one of the places that has benefited from this mining activity.

Recently, Okunlola, (2005) also define the metallogeny of the rare metal Ta-Nb pegmatites of Nigeria outlining 7 broad fields namely, Kabba-Isanlu, Ijero-Aramoko, Keffi Nasarrawa, Dema-Ndeji, Oke Ogun, Ibadan-Osogbo and Kushaka-Birnin Gwari. The Sekpeteri pegmatites occurrences which are members of the Oke Ogun field occurrence is here studied with the aim of elucidating the radioactive features, radiation hazards and thus understanding the environmental geology of its mining.

2. MATERIALS AND METHODS

2.1 Study Area

The study area is situated in Sepeteri, adjoining the Headquarters of Saki East Local Government Area, Oyo State SouthWestern Nigeria. Sepeteri lies between latitudes $N8^{0}30'$ and $N8^{0}$ 40′ and longitudes $E03^{0}$ 37′ and $E3^{0}$ 45′. This area is located south from the main town (Sepeteri), in the humid tropical region of south western Nigeria. The temperature range within the area is 20-32˚C. Accessibilty of the area is achieved through a not-so-good network of roads but well developed footpaths that radially spring out to the main town. Sepeteri town lies in the Guinea savannah zone with tall grasses of about 1.5 m high in most parts and shrubs are present as well as many luxuriant trees. The relief of Oyo state is divided into two units; the undulating lowland terrain and hills occurring as dome shaped inselberg. Sepeteri belongs to the lowland terrain category. The main river in the area is Ogun whose tributaries drain the whole town and its environs. Some of the tributaries are Owutu, Afo, Awerole and Oje rivers. They affect the town in some localities to form a dendritic drainage pattern that springs out from Ogun river.

The pegmatite of Sepeteri belong to the Oke Ogun field of Rare-metal pegmatites (Okunlola, 2005). Although the Nigerian rare-metal pegmatites are found to be of granitic origin they are evidently not co-genetic to the more widespread Pan-African granitoids and they appear to be highly fractionated granitic pegmatites of probably remote origin(Okunlola, 2005). The pegmatites of Sepeteri area, with average dip of 60^{o} striking mainly in the N-S direction, intruded into the older lithology of amphibole schists.(Okunlola, 2005). The amphibole schists are usually greenish, fissile and in some cases weathered. In the amphibole schist major minerals include quartz, plagioclase, microcline, biotite and hornblende while accessory minerals include sphene, zircon, apatite and opaque. These amphibole schists serve as major hosts for the pegmatite intrusions. The mica schists also found in the area are mostly light grey in colour and composed mainly of muscovite and minor biotite with intergranular fine grained quartz. Pegmatite occurs as coarse inequigranular veins, milky white in appearance. Petrographic study by Okunlola, 2005, shows that the predominant mineralogical constituents include quartz, microcline, albite, muscovite and minor amounts of biotite. Microcline is the most abundant of these minerals often graphically intergrown with quartz.

2.2 Sampling

Preliminary in-situ survey of the radioactivity was done in the field using a Gamma Scout detector.

A total of eight (8) samples were collected from three open pit locations scattered over the study area(table 1). Two samples of tailings were taken from each pit, a soil sample taken about 1 km to the mining site was employed to act as control, and a sample of the ore concentrate was also taken for analysis. The samples were air dried in the room temperature for two weeks to a constant weight, then ground to fine grain size using a rock crusher and later sieved through a 2 mm sieve. Each sample of about 200 g was weighed into a plastic container and sealed firmly for at least 28 days so as to achieve secular equilibrium and to enhance Radon-226 production. All weighing was done using a digital Metler Toledo balance from the Department of Physics, Obafemi Awolowo University, Ile-Ife.

2.3 Instrumentation

The in-situ measurement was carried out using a GAMMA-SCOUT Radiation Detector
Model: GAMMA-SCOUT GmbH & Co. KG Serie: 038439 (November 2009). It displays an instant radiation and equiped with a Geiger-Muller counter tube enabling you to detect not only gamma rays, but alpha and beta rays as well. It has the ability to shift from dose rate mode to pulse counting mode and reverse.

The gamma ray spectrometric analysis was carried using a 3’’x 3’’ sodium iodide NaI(Tl) detector in laboratory located at the Centre for Energy Research and Development (CERD),

Obafemi Awolowo University (OAU), Ile-Ife. The detector was enclosed in a 100 mm thick lead shield. The energy and efficiency calibration were done using an IAEA-375 reference soil standard. Each sample was counted for 7 hours twice in order to check the reproductivity of the counting system. The photopeaks observed with regularity were belonging to those headed by ^{238}U- and ^{232}Th-series as well as the non-series ^{40}K.

3. RESULTS AND DISCUSSION

3.1 In-situ Measurement

The results of the in-situ survey are presented in Table 2. The γ dose rate ranges from 0.10 µSv/hr to 0.24 µSv/hr with an average of 0.17 µSv/hr. The ore concentrate recorded the highest values, while the lowest was obtained in air 1 m above the ground. The dose rate for γ + β range between 0.12 µSv/hr and 0.32 µSv/hr with an average of 0.25 µSv/hr. It is easily noticed that the ore concentrate shows the highest values for both readings. The Doses rates in the tailings are relatively higher than those from pit, and for each pit, it increased with the depth.

No result was recorded α + β + γ measurement. This proved the presence of β-emitters and no α- emitters. These results falls within some ranges reported elsewhere, such as 0.03 to 0.15µSv/hr on land and up to 30µSv/hr in Brazil, China, and India (Larmash, 1983). Although our results still relatively low when compared to the limit for the general public of 0.5 µSv/h and 5.7 µSv/yr for person occupationally exposed (OECD, 2011). However, activities involving the extraction, mining, beneficiation, processing, use, transport, storage, disposal, and recycling of tailings and ore concentrates as well as top soil may increase the exposure levels to workers and other individuals to level of concern. Based on the in-situ measurement, it can be affirmed partially that the mine is 'moderately radioactive' but this result is not conclusive since the in-situ survey does not indicate the activity of each of the radionuclides present.

3.2 Radioactivity Contents

The average activity concentrations in each sample analyzed are shown in Table 3. For tailings, this vary between 194.87 and 927.29 Bq kg^{-1} with an average of 511.35 ± 86.25 Bq kg^{-1} for ^{40}K; 26.06 and 68.38 Bq kg^{-1} with an average of 51.54 ± 11.84 Bq kg^{-1} for ^{238}U and between 17.65 and 21.59 Bq kg^{-1} with an average of 19.61 ± 8.19 Bq kg^{-1} for ^{232}Th; ^{40}K has the highest concentration of the three radionuclides present (about 87% of the total activity), the ore concentrate has the highest amount of ^{40}K activity concentration (1035.56±102.43Bq), while the tailings from pit 3 has the lowest amount of ^{40}K activity concentration. The high amount of ^{40}K in the ore concentrate has a bearing on their geochemical characteristics that rare-metal pegmatites have a high K:Ba and Rb:Sr but low K:Rb ratios which attest to their granitic origin. The higher concentration of ^{40}K compared to ^{238}U and ^{232}Th is consistent with Aten et al. (1961), who noted that the activity of ^{40}K in rock samples depends on the relative amounts of feldspar, mica and clay minerals that make up the mineral aggregate of the rock, so this is attributed to the presence of K-feldspar (mostly microcline perthite) and mica in Rare metal pegmatite (Okunlola, 2005). The activity of ^{40}K is generally less in control than in the tailings except in pit 3, this is in conformity with the results gotten from the in-situ survey.

The value recorded for ^{40}K in this rare metal pegmatite belonging to Oke-Ogun field when compared to values recorded for other countries, falls among the high radiation of countries like India, Brazil, Norway e.t.c (UNSCEAR, 1993).

^{238}U is the second highest contributor to the activity in the area covering about 8.5% of the total activity concentrations. The activity of ^{238}U is higher in all the samples than that of ^{232}Th and this consistency is reflected in all the samples including the ore concentrate. The ore concentrate has the highest activity of ^{238}U having a value of 97.10±18.47 Bq kg^{-1} and thus contributing 22.21% of the 8.5% from total radiation activity which is equal to about 1.8% of the total activity concentration. Except for the tailings of pit 3, the activity of ^{238}U in the control sample is generally less when compared with other samples including the ore concentrates. This reconfirm that intensity of radiation is decreasing up the pits and concentration of radionuclides is higher in the ore than the overburden. This also point to the fact that the rare pegmatite is responsible for the uranium decay.

^{232}Th makes the least contribution to the total activity in the area, with concentration ranging from 17.65±8.42 Bq kg^{-1} to 85.65±17.26Bq kg^{-1}. It constitutes about 4.5% of the total activity. The ore concentrate has the highest activity of ^{232}Th with a value of 85.65±17.26 Bq kg^{-1} while the tailings from pit 1 has the lowest value of ^{232}Th. The high value of ^{232}Th in the ore concentrate compared to control and tailings, point to the notion that the source of the ^{232}Th is the pegmatite. In contrast to ^{238}U, ^{232}Th is highly insoluble under all geochemical conditions. This means that it will have been more difficult to mobilize ^{232}Th than to mobilize ^{238}U from the source probably the pegmatite even if they have occurred with the same concentration. This agrees with NCRP (1987), that Uranium is more mobile than Thorium and would appear to explain the observation that ^{238} U is more than ^{232}Th in the samples. Generally, the activity concentration of ^{232}Th and ^{238}U in the samples are within and maybe slightly higher than the maximum permissible activity concentration of 25 Bq kg^{-1} for the general public (IAEA, 1979), except the activity concentration of the ore concentrate that is many times higher

than this threshold limit for the general public. Nasiru and Umar, 2003 observed that in Jos's Mining site (Plateau State), the higher radioactivity concentration were due mainly from monazite, xenotime and zircon which contain substantial quantities of uranium and thorium. These elements are not found in the rare metal pegmatite of Sepeteri (Okunlola and Akintola, 2005) hence, these could be the reason while tantalite mine in Sepeteri is not as radioactive as the mine in Jos plateau but considerably higher than the threshold of 25 Bq kg^{-1} for the general public. Going by the activities of these samples especially the ore concentrate, the mine at Sepeteri is not has hazardous as the mine at Jos but prolong exposure by the mine workers could pose some health danger.

3.3 Absorbed Dose Rate

The Absorbed Dose rates from the samples were calculated using the relationship derived by Beck et al. (1972) which is given as:

$$D = 0.042\ Ac\ (_K) + 0.429\ Ac\ (_U) + 0.666 Ac\ (_{Th}).$$

Where D is the Absorded Dose Rate (in nGy hr^{-1})

$Ac(_k)$, $Ac(_u)$ and $Ac(_{Th})$ are the activities concentration(in Bq kg^{-1}) for ^{40}K, ^{238}U and ^{232}Th respectively.

The results, presented in table 3, ranged between 34.00 nGy hr^{-1} in tailings from pit-1 to 142.19 nGy hr^{-1} in the ore concentrate. Normal background radiation ranges between 30 to 70 nGy hr^{-1}(UNSCEAR, 1988) hence, ore concentrate is on the high side of the normal background value as it doubles this value. Except for tailing of sample 4, all the tailings have values that are within the background value. This is in consonant with the other results on the radioactivity content of the ore concentrate.

In order to evaluate possible biological, the Dose Equivalent was derived using the weighting factor of 1 Sv Gy^{-1} for gamma radiation. The dose equivalent ranges from 0.298 mSv yr^{-1} in tailings from pit-1 to 1.25 mSv yr^{-1} in the ore concentrate. The other samples are approximately 0.5 mSv yr^{-1}, which are below the threshold value of 1 mSv yr^{-1} for the general public (ICRP, 1991). Hence, it is therefore harmful to humans handling or exposed to the ore concentrate.

4. CONCLUSION AND RECOMMENDATIONS

The measurement of the radioactivity in rare metal pegmatite in Sepeteri and the derived health hazard has being carried out hereby providing a useful information and data for establishing a comprehensive framework to investigate other mines and develop guidelines for monitoring and control of NORMs in the mining industry and the environment as a whole in Nigeria.

The results of the study indicated that the activity of radionuclides in the tailings and control sample are within the threshold limits except for the ore concentrate that shows a very high radionuclide activity concentration. The health impact assessment of the study area reveals that it is not safe for people to handle the ore because it shows a higher radioactivity than the threshold limit. It is therefore necessary to made the following recommendations

The work should be expanded to cover the whole of Oke-Ogun field of Rare metal pegmatite and also to cover the whole of the Rare-metal pegmatite in the country, elemental analysis should also be deployed to serve as check and provide more comparable and reliable data so as to know which element is responsible for the radioactivity in the ore concentrate,
regular monitoring should be conducted to take care of unforeseen eventualities especially in newly discovered field, and in setting up a mine the geology and the radioactivity contents of the area should be properly understood.

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Table 1: Sampling Location and Description

Sample No	Sample type	Sample description	Longitude(E)	Latitude(N)
1	Control sample	Darkcoloured sample containing topsoil.	E003˚39.431´	N08˚31.61´
2	Tailings	Tailings from pit 1,	E003˚39.426´	N08˚31.543´

3	Tailings	Second tailing pit - 1	E003˚39.539´	N08˚31.539´
4	Tailings	First tailing from pit2	E003˚39.949´	N08˚31.469´
5	Tailings	second tailing from pit2	E003˚39.935´	N08˚31.477´
6	Tailings	First tailing from pit3	E003˚40.286´	N08˚31.028´
7	Tailings	Second tailing from pit3	E003˚40.281´	N08˚31.033’
8	Ore concentrate	Dark coloured grain mixture of tin, tantalite ore from pit3	E003˚40.283’	N08˚31.030’

Table 2: Results of Dose Rate from the In-Situ Measurement

Sample Type	Description	Location		Dose Rate (μSv/hr) for	
		Latitude	Longitude	γ	γ + β
Control 1	1 km to the mine	08º 31.610'	003º 39. 432	0.11	0.12
Control 2	1 km away from the mine	08º 31.191'	003º 39.852'	0.12	0.13
Air	1 m above the ground	08º 31.537'	003º 39.943'	0.1	0.12
tailings-1	from pit 1	08º 31.535'	003º 39.932'	0.22	0.26
tailing-2	from pit 1	08º 31.525'	003º 39.931'	0.19	0.2
tailings-1	from pit 2	08º 31.475'	003º 39.953'	0.2	0.23
tailing-2	from pit 2	08º 31.472'	003º 39.951'	0.19	0.21
tailings-1	from pit 3	08º 31.032'	003º 40.272'	0.21	0.23
tailing-2	from pit 3	08º 31.02 9'	003º 40.275'	0.21	0.24
ore concentrate	from pit 3	08º 31.029'	003º 40.275'	0.24	0.32
depth-1	Bottom of pit 1	08º 31.537'	003º 39.940'	0.16	0.18
depth-2	5 m from bottom	08º 31.537'	003º 39.940'	0.14	0.16
depth-3	10 m from bottom	08º 31.537'	003º 39.940'	0.12	0.16
depth-1	Bottom of pit 2	08º 31.475'	003º 39.953'	0.2	0.22
depth-2	5 m from bottom	08º 31.472'	003º 39.951'	0.18	0.20
depth-3	Top of pit 2	08º 31.472'	003º 39.951'	0.14	0.20
depth-1	Bottom of pit 3	08º 31.032'	003º 40.272'	0.19	0.21
depth-2	5 m from bottom	08º 31.029'	003º 40.275'	0.13	0.16
depth-3	Top of pit 3	08º 32.035'	003º 40.275'	0.16	0.17
Average				**0.17**	**0.25**

Table 3: Activity concentration, Absorded Dose Rate and Dose Equivalent in the samples

S/N	Sample type	Activity Concentration (Bq kg^{-1})			Absorbed Dose Rate (nGy/hr)	Dose Equivalent (mSv/yr)
		^{40}K	^{238}U	^{232}Th		
Sample 1	Control	373.36±67.73	30.74±9.39	27.57±6.31	47.23	0.4140
Sample 2	Tailings from pit1	672.41±76.67	68.38±17.75	17.65±8.42	34.00	0.298
Sample 3	Tailings from pit 1	501.48±98.52	51.09±11.51	20.45±9.23	41.88	0.367
Sample 4	Tailings from pit 2	540.48±105.44	53.43±12.83	18.98±8.32	79.80	0.699
Sample 5	Tailings from pit 2	927.29±92.72	61.72±15.43	21.59±7.54	58.26	0.511
Sample 6	Tailings from pit 3	231.54±75.32	26.06±7.22	19.67±7.51	56.60	0.496
Sample 7	Tailings from pit 3	194.87±68.84	48.54±6.32	19.32±8.12	69.33	0.608
Sample 8	Ore concentrate	1035.56±102.43	97.10±18.47	85.65±17.26	142.19	1.25
Mean(Tailing)	This work	586.23 ± 88.56	58.04± 12.78	29.04 ± 9.49	49.73	0.50
	Other(Nasiru and Umar, 2003)		1247.5	986.83		