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RADIOLOGICAL ASSESSMENT OF COLTANT MINING IN NIGERIA.

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ABSTRACT:
Coltant has global demand for variety of application due to its qualities of being good heat conductor, corrosion resistant. The aim of this study was to assess radioactivity level emanating from coltant shipments from Nigeria, due to several recorded cases of delays/denials and impoundment from transit or importing countries, using RDS-31S/R Multi-purpose survey meter, Radionuclides Identifinder and Sodium Iodide detector. The dose rate measurement from the coltant samples ranges from (0.23±04 and 8.44±03) $\mu Sv/hr$ which was above maximum dose rate of 1 $\mu Sv/hr$ at 1m from the package surfaces. Also, specific activity concentration of ^{226}Ra, ^{232}Th and ^{40}K found to be (335±195 - 15,786±9,164) $Bqkg^{-1}$, (152±18 - 15,201±1,738) $Bqkg^{-1}$and (16±10 - 142±21) $Bqkg^{-1}$, respectively. The evaluated GDR was with a mean value of 7959 nGy/hr higher than world mean of 59 nGy/hr. The outdoor and indoor AEDR estimated were 9.76 mSv/yr and 39.05 mSv/yr respectively higher than world mean of 420 mSv/yr; the estimated ELCR was 3333 higher than world mean value of 0.29×10^{-3} as reported in UNSCEAR 2000. The radiological risk assessment from this investigation reveals workers and public in such mining areas and other biota will definitely be overexposed leading to radiation health effects over long term. It is recommended that there should be effective regulatory control of mines and mining Coltant products which often contains fissile material, ore and radioactive substances.

Keywords: Coltant, Mines products, Radioactivity level, Shipment and Transportation Packages

INTRODUCTION:

Coltan is a combination of columbite and tantalites which is a gloomy black metallic mineral, which has qualities of highly resistant to heat, corrosion and a good conductor, making it quite useful in several applications like in electronics, medical devises, optical lenses, aerospace engines cutting tools, capacitor, resistor, coil, transformer, mobile phones, super alloy for nuclear reactor etc (Tantalum-Niobium International Study Center, 2002)

Nigeria is endowed and has a large and exceptional deposit of tantalite /Coltant, which is found in the several parts of Nigeria found in granite complexes in Jos – Plateau, leading to commercial exploitation for economic purposes. It has contributed immensely to socio-economic benefits and accounts for 0.3% Nigeria's GDP ((Jibiri *et al.* 2011; Usikalu *et al.* 2011).

The uncontrolled and indiscriminate mining, mainly open pit type leaves behind burdens of mine tailings of technically enhanced Natural Occurrence Materials (NORMs) at the earth surface, which normally associates with high concentration of primordial radionuclides (^{226}Ra, ^{232}Th and ^{40}K), and thus increasing radioactivity levels in the environment. There is therefore a huge damaging effect of these technical enhanced NORMs to ecosystem, consequently resulting in impoverishment of agriculture farm lands, environmental and radiological health hazards of host community, leading to radiation sicknesses, through several pathways exposures (Aigbedion and Iyayi, 2007; Pasquini and Alexander, 2005; Mallo & Wazoh 2014).

This investigative study is as a result of recorded cases of delays, denials and impoundments of coltant products shipments originating from Nigeria either at transit port or importing countries Regulatory Body (RB), due to radioactive material concentration associated with coltant mining ores. The objective was to evaluate radioactivity level of coltan from Nigeria and make recommendations to exporters, in order to demonstrate adequate international compliance regulatory requirements and provisions of ICAO-TI on packages and dangerous goods declarations to forestall future contraventions (IAEA, 2010; UNECOSOC, 2017; IAEA, 2018).

MATERIALS AND METHOD

Study Areas

The study areas of Jos Plateau is located within coordinates of 09°46'02N and 008°51'39E for Latitude and Longitude respectively. It is bounded with Bauchi State – to the north east, Kaduna State – to the north west , Nasarawa State – to the south west, Taraba State – to the south east. Jos, Plateau is geographically exceptional in Nigeria due to its boundaries of elevated hills with altitude of 1279 m, and considered as highest plain amongst surrounding plains. This results in a near temperate climate with an average temperature of between 13 and 22 °C even though in the tropical zone. Harmattan winds cause the coldest weather between December and February. Plateau State geological formation is basically Precambrian basement complex rocks as well as Jurassic younger granites (Biotite granites) and the tertiary quaternary volcanic rocks (basalts, pumice, lava flows and ash deposits. The younger granite province is where mineral deposits of coltant, tin etc is normally found in Jos Plateau (Mallo & Wazoh 2014; Jibiri *et al.* 2011). The a 2-D Arc-Gis Map map describing geological formations of Jos insert in the Plateau State, Nigeria is presented in Figure 1.

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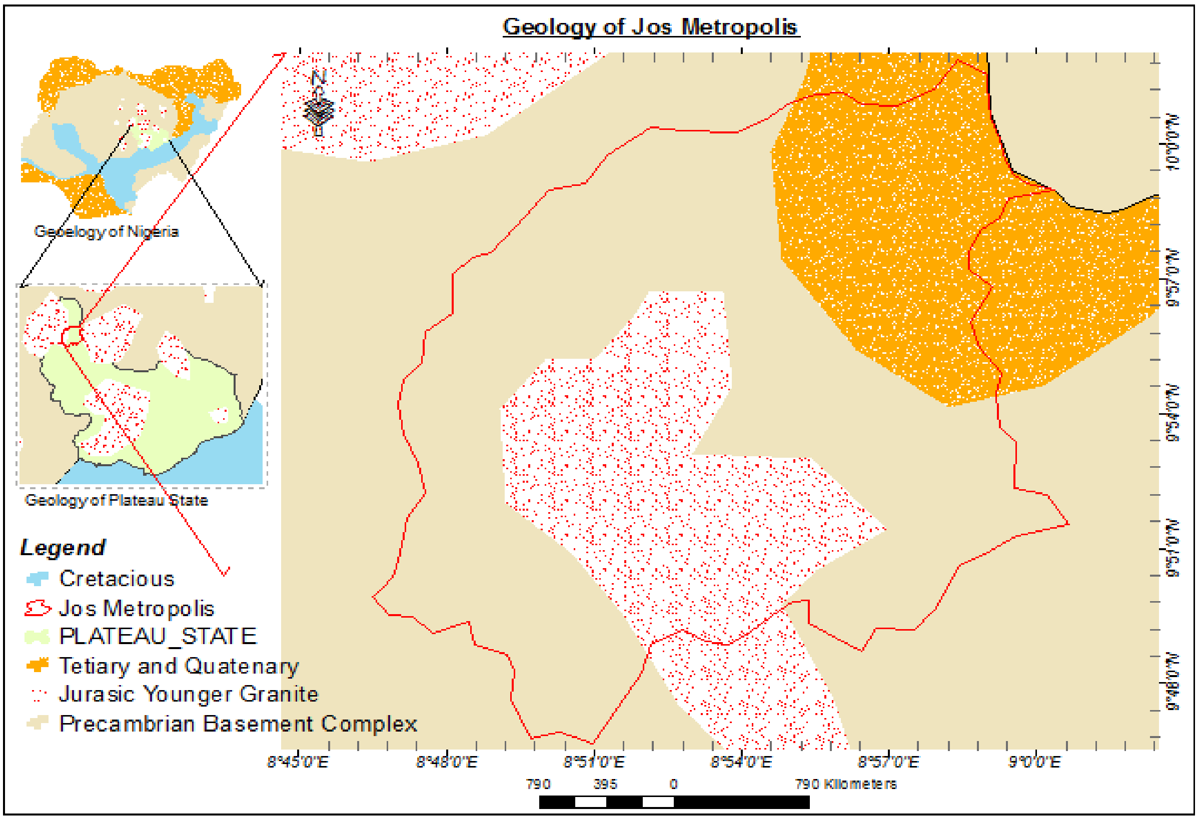

Figure 1: 2–Dimensional Arc-Gis Map presentation of geological formations and tin fields of Jos insert in the Plateau State, Nigeria.

Dose Rate/Preliminary measurements

The method employed in the investigative study was basically judgmental random sampling, since the locations were already predetermined, and in this instance the Coltant Export Company (CEC), Nigeria sites and warehouses where the impounded exporting coltant product originated from. The global positioning system (GPS) – GARMIN Etrex 10 (Serial number 3964) was used to locate coordinates of mine sites, warehouse and packages. Radiation measurements and qualitatively radionuclides identification were done with RDS-31S/R Multi-purpose survey meter with serial number 2100372, measuring a dose range of 0.01 uSv/hr to 10 mSv/hr and a Radionuclide Identifinder (RID) respectively. The RID was able identify radionuclides or elements present in coltant material ores to about 100% certainty with corresponding dose rate measurements. The preliminary result from the RID measured during investigation study collaborated with the suspected radionuclides reported from the coltant shipment impounded at transit Airport by RB (Table 1, Column 3). To ensure adequate equipment performance, a pre-operational/ functionality and quality checks prior to dose rate measurements were performed on the equipment to ensure their effective, accurate and perfect working conditions (IAEA, 1999).

Collection of coltant samples

The coltant material ores samples were collected through grab method at the same locations where the dose rates and preliminary measurements were conducted, which were mine sites, ware houses and some packages ready for export. These samples were bag into a polyethylene and sealed, carefully labelling with codes names for proper identification and conveyed to the laboratory, Pictures are given Plates 1- 4.

Analysis of coltant samples

The coltant sample were open air dried, subsequent oven dried at about 50^oc - 150^oc to obtain constant weight, stony samples crushed, pulverised through 500 μm mesh sieve for homogeneity and increase surface area and reduce much attenuation. Coltant samples were packaged in Marinelli beaker and placed vertically on a detector and counted for 18,000 seconds. Gamma spectroscopy method employing a Sodium Iodide (NaI) detector with 80% Efficiency and Resolution of 2.4 keV at ^{60}Co was used to analyzed coltant samples to acquire spectra on its Region of Interest (ROI) which identifies the radionuclides present qualitatively, and counts obtained with corresponding uncertainties to quantitatively determine the activities of radionuclides.

Plate 1: TENORM Zircon sand dump

Plate 2: Air drying of coltant at warehouse

Plate 3: Coltant products storage at warehouse

Plate 4: Coltant processing at warehouse

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Theory

Activity Concentration (A_c) can be calculated from analyzed sample data using Equation (1) (Al-Sulaiti, 2009):

$$A_c = \frac{C_{net}}{\varepsilon_\gamma \times I_\gamma \times m} \quad (1)$$

where mass of the sample is denoted as *m* in the expression. The unit of activity concentration of soil sample is given as $Bqkg^{-1}$.

In determining the absorbed dose rates (D), conversion factors were applied. These factors used in Equation 1 are of postulation that all the progenies of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their parents radionuclides (UNSCEAR, 2000; Rani & Singh, 2005).

$$\mathbf{D = 0.461A_{Ra} + 0.623A_{Th} + 0.0414A_K} \quad (2)$$

An Annual Effective Dose Rate estimations are derived from Gamma Dose Rate, a conversion factor of 0.7 Sv/Gy of absorbed dose in air to effective dose an adults receives with 20% time out-of-doors (80% indoors) is given in Equation 3 (Al-Sulaiti, 2009; UNSCEAR, 2000):

$$\mathbf{AEDE_{\mu Sv} = D_{nGy/h} \times 8760_{h/y} \times 0.2 \times 0.7_{Sv/Gy} \times 10^{-3}} \quad (3)$$

For relative purposes, assessing the hazard associated with material containing different concentrations of ^{226}Ra, ^{232}Th and ^{40}K, Radium Equivalent Activity can be estimated by the expression in Equation 4 (Mantazul, 1979; Beretka, *et al.*, 1985).

$$\boldsymbol{Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.007A_K} \quad (4)$$

Excess Life Cancer Risk is determined from the Annual Effective Dose Rate with Duration of Life (DL, 70 years for children and 50 years for adult), Risk Factor (RF, 5%) for public exposure considered to produce stochastic effect is given in Equation 5 (Taskin *et al.*, 2009; ICRP, 1990).

$$\mathbf{ELCR_{mSv/y} = AEDE_{mSv/y} \times RF \times DL} \quad (5)$$

RESULTS AND DISCUSSIONS

Results

Table 1 presents Dose rate measurements and activity concentration of coltant products collected from Coltant Export Company (CEC) Jos, Nigeria.

The dose rate measurement reported in Table 1 Column 3 range (1.29±0.9 - 8.44±0.4) μSv/hr, and higher than 0.2 μSv/hr of the global average and above 1μSv/hr recommended at 1 meter from packages during transportation (UNSCEAR, 2000; IAEA, 2018).

Also, the preliminary measurement from the RID as reported in Table 1 Column 4, from all the coltant samples except for JK-W showed qualitative description of ^{226}Ra, ^{232}Th progenies, thus also corroborating initial suspecting report from RB at the Airport that led to the coltant consignment impoundment.

Furthermore, activity concentration derived from Equation 1 from laboratory analysis of coltant samples are reported in Table 1 Column 5 for ^{226}Ra, ^{232}Th and ^{40}K were found to be (335±195-15,786±9,164) $Bqkg^{-1}$, (152±18-15,201±1,738) $Bqkg^{-1}$and (16±10 - 142±21) $Bqkg^{-1}$, respectively. These activity concentration far exceeded the global average of 33 $Bqkg^{-1}$, 45 $Bqkg^{-1}$ and 420 $Bqkg^{-1}$ for ^{226}Ra, ^{232}Th and ^{40}K respectively (UNSCEAR, 2000). These high activity concentration recorded were due to minerals ore mined from Jos, Plateau are usually associated with TENORMs.

Table 1: Dose rate measurements and activity concentration of Coltant products collected from Coltant Export Company Jos, Nigeria

S/NO	Sample Names/ Codes	Dose Rate (μSv/hr)	RID Preliminary Result from sites	Laboratory Analysis Activity Concentration (Bq/kg)		
				K-40	Ra-226	Th-232
1	J-KN (Kano)	4.10±0.5	57% Th-232; 52% Te-132	ND	5155±2993	18422±2.106
2	J-KW (Kwara)	4.35±0.7	50% Mn-54; 47% Be-7	123±11	4931±2863	7044±806
3	J-TL (Tailings)	4.70±0.8	71% Th-232; 45% Ru-103	ND	6339±3680	10830±1238
4	J-TR2 (Toro 2)	4.80±0.6	58% Na-22; 56% Bi-207	16±10	817±475	1185±138
5	J-ZS (Zircon Sand)	8.44±0.4	64% Ra -226; 55% Y-88	ND	15786±9164	7480±857
6	J-BL (Barinkin-Ladi)	3.56±0.9	60% Na-22; 56% Pb-214	ND	335±195	152±18
7	J-C63 (Coltant 63)	1.29±0.9	57% Zr-95; 54% La-140	ND	13493±7832	2407±276
8	J-C64 (Coltant 63)	1.29±0.9	59% Bi-207	95±39	7401±4296	15201±1738
9	J-TR1 (Toro 1)	2.90±0.6	58% Na-22; 56% Bi-207	142±21	3239±1881	9692±1108

Discussions of Results

It was evident that in both the preliminary measurements and laboratory analysis, ^{40}K was found in only three (3) of the coltant samples with activity concentration below admissible limits but were not found in other six (6) coltant samples as stated in Table 1, Columns (3-4), and this was attributed to perhaps geological formation (Njinga *et al.*, 2015).

The maximum dose rate measurement and activity concentration result of laboratory analysis from this investigation study, corroborates with maximum dose rate of 8μsv/hr and activity concentration ^{232}Th which was 2,400±400 Bq, ^{238}U which was 4,900±700 Bq in a 381.0 g conducted on the CEC coltant consignment, by the RB at the Airport of the impounding country. Also, these values were above Regulatory requirement of Quantity for Notification ($1x10^4$ Bq) and the aggregate of these samples put together for export/shipment likely to exceed 20 tons of the international safeguards requirements for declaring such shipment (NiBRII, 2003; IAEA, 2010; IAEA, 2018). This high radioactivity measurement was regarded as gross violations of regulatory requirements and

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ICAO-TI provisions, as regards to inappropriate labelling and declaration on packages containing fissile materials, ore and radioactive substances (IAEA, 2010; UNECOSOC, 2017; IAEA, 2018). This had led to impoundment of the five (5) pieces of CEC, Jos Nigeria coltant products at the transit Airport by RB.

A similar studies agrees with this investigative study having same elevated radioactivity level due to anthropogenic activities of mining in same city of Jos, Plateau. Soils and food crops (yam) from dump tin mine area were measure to determine radionuclides contents. The ^{40}K, ^{238}U and ^{232}Th concentrations ranged between (60 – 494) Bqkg^{-1}, between (0 – 48) Bqkg^{-1} and between (0 – 17) Bqkg^{-1} respectively. The ^{40}K was the highest contributor than other radionuclides in the food products analysed, and this high concentration was largely attributed to the mining activities (Jibiri *et al.*, 2007). Also, evaluations of tin tailings were conducted to determine the activity concentrations of the samples. The ^{40}K was below detectable limits while ^{238}U ranged between (17.1 x 10^2 - 16.6 x 10^3) Bqkg^{-1} with means of (72.2 x 10^2) Bqkg^{-1} and ^{232}Th ranged between (52.9 x 10^2 to 47.5 x 10^3) Bqkg^{-1} with means of 16.8 x 10^3 Bqkg^{-1} which exceeds world mean (Ademola, 2008).

Discussions of Radiological Health Parameters

Table 2 presents the radiological hazards considerations of activity concentration from the coltant analyzed samples of Table 1 were evaluated using Equations (2-5).

The estimated GDR arising from terrestrial gamma of activity concentration from the samples of the investigation area was with a mean value of 79 nGy/hr were found to be higher than world mean value of 59 nGy/hr (Al-Sulaiti, 2009; UNSCEAR, 2000). More so, the AEDR estimated from both terrestrial outdoor was with mean value of 9.76 mSv/yr, whereas the evaluated indoor AEDR was with mean value of 39.05 mSv/yr also higher when compared with world mean value of 460 mSv/y The rationale behind outdoor AEDR lower than that of AEDR indoor is because more time is spent indoors than outdoors. Furthermore, the estimated ELCR was with the mean value of 3333 higher than world mean value of 0.29×10^{-3} (Al-Sulaiti, 2009; UNSCEAR, 2000). This means that the possibility of infant of adult becoming a cancer patient in the study area is very high. The radiological hazards indices evaluation of the dose rate measurements from the site and warehouses translated to between (2.58 – 16.88) mSv/yr which is far above public acceptable annual effective dose without constraint of 1mSv/yr for the purpose of safety, and with constraint of 0.5 mSv/yr (Al-Sulaiti, 2009; UNSCEAR, 2000). The implication of these radiological risk assessment from the study investigation is that, there is likelihood of overexposures leading to radiation health hazards over long term. First to workers who spends more than 2000 hours in a year working without any protective measures, also to the public through external exposures on elevated radiation level, through pathways of inhalation of re-suspended TENORMs particles in air, ingestions through various food chain, and finally to other biota.

Table 2: Radiological hazards indices of Coltant products collected from Jos, Nigeria

Sample	GDR (nGy/hr)	Ra_{eq} (Bq/Kg)	AEDR (Outdoor) (mSv/y)	AEDR (Indoor) (mSv/y)	ELCR
J-KW	6666	15004	8176	32704	28616
J-BL	249	552	306	1222	5.22
J-TR1	7537	17099	9244	36974	161
J-KN	13853	31498	16989	67959	298
J-C64	12886	29139	15803	63214	275
J-C63	7719	16935	9468	37870	160
J-TL	9669	21826	11859	47434	206
J-ZS	11937	26482	14640	58560	250
J-TR2	1115	2512	1368	5472	24
Inv. Mean	**7959**	**17894**	**9761**	**39046**	**3333**
World Mean	59	370	0.46	0.46	0.29

CONCLUSION

The coltant extraction and mining in Nigeria is economically viable, but with environmental burdens due to abandon mine tailing on earth surface, thereby increasing radiation exposure level. Some coltant samples from CEC were collected and analysed to determine radioactivity level, and there were found to exceed the admissible global and regulatory limits, and which translate to radiological hazards on mining workers, public, environment and other biota. In these regards, to forestalling future contraventions of international regulations, there is need for exporters to conduct both radiometric and elemental analysis on coltant products from Nigeria to determine radioactivity level, apply appropriate labelling/ packaging and declaration prior to export. Furthermore, constant enlightenments on radiological hazards associated with coltant mining is recommended for miners/ exporters and a uniform ICAO-TI requirements be developed as guidelines/extract for exporters to be acquainted with requirements before coltant shipment.

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