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MEASUREMENT OF RADIONUCLIDES IN PROCESSED MINE TAILINGS IN JOS, PLATEAU STATE

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ABSTRACT
The Jos Plateau is situated in the central part of Northern Nigeria, on a rugged terrain of low lands at the edge of the Plateau surface (Hyde, 1986). It is the principal centre of tin and columbite mineralization, which forms the focal area of younger granites (Macleod et al., 1971). The mine tailings are associated with radioactive minerals as impurities such as monazite, zircon among others. These minerals are very radioactive and causes hazards to human health during mining and milling. A total of thirty-one (31) tailing samples were collected from a processing site in Jos at different directions and distances and analyzed for ^{40}K, ^{226}Ra and ^{232}Th concentrations using NaI (TI) detector in order to estimate the level of their radiological impact to the public. The highest and lowest concentrations were in D_8 of ^{232}Th (27930.0Bqkg^{-1}) and D_5 of ^{226}Ra (364.9Bqkg^{-1}). All concentrations ranged from 364.9Bqkg^{-1} – 27930.0Bqkg^{-1} in the tailings.

Keywords: Tailings, Radionuclides, Radiation and Dose Limit.

INTRODUCTION

A lot of mining activities have been taking place in Jos area, for several decades, with extensive commercial mining and processing starting in 1930's (Onuoba, 1992). An extensive quantity of mine tailings have been generated by the several mining companies that operated in the area and these are either in the premises of the mining companies or are dumped in and around the mining pits (Funtua, 2001).

Different kinds of radioactive wastes both in liquid, gaseous and solid form have different levels of radioactivity and therefore required different methods of management (GAEC, 2005).

Tailings also known as tailings pile, tails leach residue or slickens are the materials left over after the process of separating the valuable fraction from the worthless fraction of an ore. These are waste products that have no financial gain to a mineral operator at that particular point in time. These tailings consists of ground rock and process effluents that are generated in a mine processing plant.

Tailings characteristics can vary greatly and dependent on the ore mineralogy together with the physical and chemical processes used to extract the economic product. Tailings of the same type may possess different mineralogy and therefore will have different physical and chemical characteristics (Ritsey, 1989).

Tailings are around ten times more radioactive than typical granites. If some one were to live continuously on top of the ranger tailings, they would receive about double their normal radiation dose from the actual tailings or even more (Nasir, 1993).

Typically, the tailings consist of heavy accessory minerals that include: Zircon, monazite, xenotine, ilmenite, magnetite, some columbite and cassiterite (Funtua, 2001).

These tailings that contain radioactive elements remain active for a very long period of time and will continue to be potentially dangerous. Over the last century, the volumes of tailings being generated has grown dramatically as the demand for minerals and metals has increased and lower grades of ore are being mined. In the 1960's, tens of thousands of tones of tailings were produced each day and by 2000 this figure has increased to hundreds of thousands (Kajubick, et al., 2003).

Significantly, the hazardous nature of radioactive source depends on the radionuclides activity.

Monazite and Zircon are found to be more hazardous than the rest of the minerals that are frequently associated with cassiterite (garnet, pyrite, rutilege etc) (Umar and Rabiu, 1995). As such, all these minerals are radioactive and a major source of external radiation hazard to workers who handle them in the mines and mills as well as the general public living around the area.

Both mechanical and chemical processes are used to extract the desired product from the mine ore and produce a waste stream known as tailings (Eagel and Dixon, 2002-2007). As mining techniques and the price of minerals improve, it is not unusual for tailings to be reprocessed using new methods or more thoroughly with old methods, to recover additional minerals. Yesterday's tails can be tomorrow's resource as seen during the 1990's when the extensive tailings dumps of Kalgoorlie/Boulder in Western Australia were reprocessed profitably by kaltails mining (Wikipedia the free encyclopedia).

MATERIALS AND METHODS

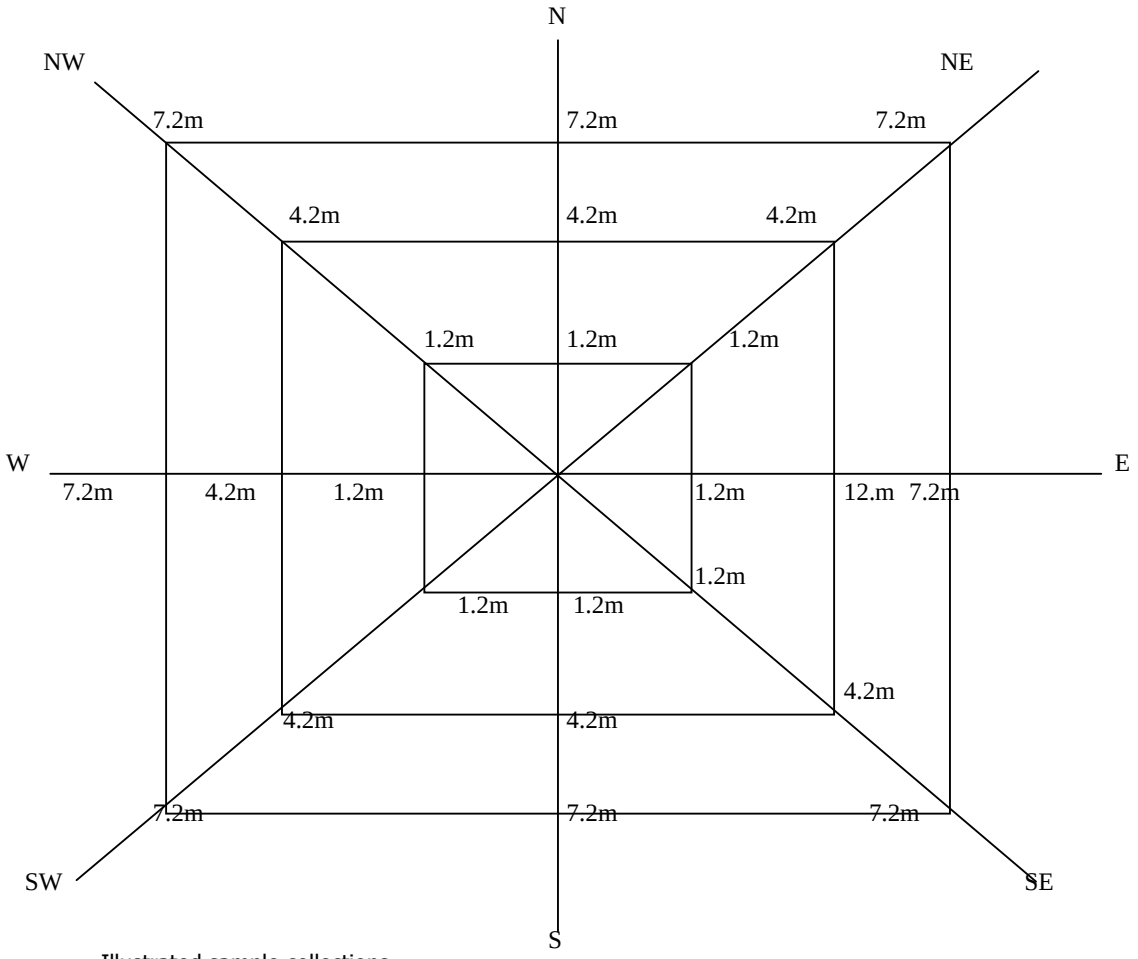
Thirty-one tailings samples were collected and emptied into rubber containers, from the tin shed mining company at Zaria Road off Rock Heaven, Plateau State.

Seven tailings samples were collected and labeled as A(s). Eight tailings samples were collected some distance of 1.2m away from the peak of the tailing and labeled B(s). Another eight tailings samples were then collected some distance of 4.2m from the peak of the tailings and labeled C(s). Finally, eight tailings samples were collected, also some distance of 7.2m from the peak of the tailings and labeled D(s).

Duration of Tailings: The collected samples have spent over six (6) years of existence.

Two (2) tailings samples labeled A_1 and A_2 were collected at the centre peak surface. Sample A_3 was collected at the centre peak of the tailings. Sample A_4 was collected 0.4m deep at the centre peak of the tailings at the depth of 0.2m. Sample A_5 was also collected at a depth of 0.6m at the centre peak of the tailings. Sample A_6 was collected at 0.8m deep at the centre peak of the tailings. Finally, sample A_7 was then collected at 1.0m deep at the centre peak of the tailings.

A distance of 1.2m away from the centre peak at the North, East, South and West labeled samples B was measured.



Illustrated sample collections

Sample Preparations

The collected tailings samples were left opened in the laboratory for a minimum of 24 hours to dry under ambient temperature. They were in fine powder as such, they were not being ground. They were then packed to fill cylindrical plastic containers of height 7cm by 6cm diameter. This satisfied the selected optimal sample container height (Ibeanu, 1999).

Each container accommodated approximately 630g of sample. They were carefully sealed with candle wax, followed by a masking tape, to prevent random escape and then stored for a minimum of 24 days. This is to allow radium attain equilibrium with the daughters.

A gamma ray spectrometry technique was employed in the spectoral collection of the prepared samples using the high energy region of the γ-lines. The measurement system consists of a 7.62cm x 7.62cm NaI (TI) detector housed in a 6cm thick lead shield and lined with cadmium and copper sheets. The samples were mounted on the detector surface and each counted for 5,000 seconds in reproducible sample detector geometry.

The chosen configuration was strictly maintained throughout the analysis. A computer based multi-channel analyzer Maestro programme from ortec was used for data acquisition and analysis of gamma spectra.

The 661.6keV x-line of GS-137 was sued in the assessment of the activity concentration of 40K while 132.5KeV 8-line of Ci-60 was used for ^{232}Th. The single 1173.2KeV 8-line of Co-60 was used for ^{226}Ra. All the obtained raw data were converted to conventional units using the calibration factors from the NaI detector to determine the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th.

RESULTS AND DISCUSSION

From Table 1, samples D_8 of ^{232}Th, B_2 of ^{226}Ra and D_8 of ^{40}K shows high concentrations in composite tailings. D_6 of ^{232}Th, D_5 of ^{226}Ra and C_7 of ^{40}K shows low concentration in the composites tailings. The highest and lowest concentrations are in D_8 of ^{232}Th (27930.0Bqkg^{-1}) and D_5 of ^{226}Ra (364.9Bqkg^{-1}). All concentrations ranges from 364.9Bqkg^{-1} to 27930.0Bqkg^{-1} in the tailings.

Due to the relatively high concentration of radionuclides in most mineral substances, the National Radiation Protection Department (NRPD) as the competent authority has established national limits (900Bqkg^{-1} for ^{232}Th, 1800Bqkg^{-1} for ^{226}Ra and 11000Bqkg^{-1} for ^{40}K) but the overall general limits for all nuclide elements is 25Bqkg^{-1} as the maximum permissible activity concentrations. Following the NRPD maximum permissible limits, it can be seen that ^{232}Th has the lowest permissible limit followed by ^{226}Ra. It shows that thorium is more hazardous or has more effects in humans than ^{226}Ra and ^{40}K.

Judging by the activity concentrations of these samples, therefore, the workers in the mine and mill are at risk of being over exposed, particularly from ^{220}Rn and ^{222}Rn which are daughter products of ^{226}Ra and ^{232}Th respectively. It is more dangerous to the smokers in the mine and mill that do smoke during mining and milling activities or around the heap tailings or environment because radon is being inhaled naturally at a very low rate but for smokers, it is twice and above higher than non-smokers.

About seventeen (17) samples have been analyzed and ^{226}Ra was not detected (ND) in them. In general, thorium is about three (3) times as abundant as uranium. It is known that thorium can be inhaled or ingested into the body. About 0.02 – 0.05% of the amount ingested is absorbed into the blood stream, through the intestine Arogunjo, 2003). Of the amount entering the blood, about 70% deposits in bone where it is retained with a biological half-life of about 22 years, 4% deposits in the liver where it is cleared with a biological half-life of 700days (Ibrahim *et al.,* 1993). Most of the remaining 10% is directly excreted (www.speclab.com/elements/thorium.htm).

Considering ^{226}Ra, ^{40}K and ^{232}Th concentrations, it could be said that, the major source of radiation in Jos Plateau State is thorite. Samples A_1 and A_2 collected at zero (0) depth, A_3, A_4, A_5, A_6 and A_7. In thorium, shows an increase in activity concentration as the depth increases starting with any of A_1 or A_2 respectively, A6 the increase in activity from A_1 to A_5 changes (decrease by 3139.4Bqkg^{-1} from A_5 instead of increasing as it started). After which it then goes up from 15604.05$\pm$194.8Bqkg^{-1} to 16975.3Bqkg^{-1}$\pm$209.4Bqkg^{-1} for the ^{232}Th. ^{226}Ra has five (5) undetected samples that goes with depth. With A_3 and A_6 showing a decrease with an increase in depth. For ^{40}K, A_1, A_3 and A_4 shows a decrease in activity concentrations with an increase in depth, at A_5 it increases to 6846.5$\pm$1302.4Bqkg^{-1}.

Thus, in ^{232}Th samples, B_4, C_4 and D_4 with activity concentration (15519.9$\pm$1189.3Bqkg^{-1} 18882.4$\pm$218.1Bqkg^{-1} and 19972.5$\pm$202.1Bqkg^{-1}) shows an increase in concentrations as the distance increases at an interval of 3m southeast. Also, B_7, C_7 and D_7 shows an increase in concentrations levels as distance increases at an interval of 3m south west. Samples B_8, C_8 and D_8 with concentrations (11483.6$\pm$1161.0Bqkg^{-1}, 14602.9$\pm$1205.7Bqkg^{-1}, and 27930.0$\pm$257.8Bqkg^{-1}) also recorded an increase in concentrations as distance increases at an interval of 3m northwest. Samples B_1, C_1 ad D_1 and B_6, C_6 and D_6 shows an increase in concentrations at an interval of 3m as distance increases but then decreases at D_1 and D_6 North and Southwest respectively. Only sample B_2C_2 and D_2, B_3C_3 and D_3 and B_5C_5 and D_5 shows a decrease in concentrations as distance increases and increases at D_2, D_8 and D_5 in the directions NE, E and S. ^{226}Ra in general shows a decrease in activity concentrations as distance increases. Samples B_4, B_5, B_6, B_7, B_8, C_1, C_2, C_3, C_5, C_6, C_7 and D_7 shows undetection of ^{226}Ra in them.

In the case of ^{40}K samples B_2, C_2 and D_2, B_4, C_4 and D_4 and B_5, C_5 and D_5 shows an increase in concentration in their respective directions as distance increases by 3m intervals, northeast, southeast and south. Samples B_1C_1 and B_6C_6 also shows an increase in concentration levels as distance increases at an interval of 3m but decreases at D_1 and D_6 in their respective directions North and South West.

Considering ^{40}K, ^{226}Ra and ^{232}Th in Table 1, 90% of the analyzed samples shows a totality of increase in their respective direction as distance increases. All these high activity concentrations point to the fact that concentration increase with distance, as such, the workers in the mine and mills as well as the general public, since most of these companies are now located in the urban areas, may be at the risk of exposure to external radiation, inhalation and ingestion. Although due to the open pit method of mining in practice, external exposure could be minimal at the mining sites, but inhalation and ingestion of radon (long-lived α-emitters and short-lived daughters) cannot be ruled out because of the dusty environment. A more serious concern should be given and also monitor in the mine tailings of Jos area before disposal. Disposal of the waste products (tailings) can be in the surface landfills or in deep geological formations depending on the levels of concentration instead of the haphazard dumping of the tailings as is being presently practiced (Funtua, 2001).

Table 1: Concentrations of ^{40}K, ^{226}Ra and ^{232}Th in tailings samples from Jos Area.

Sample No	^{40}K (Bgkg^{-1})	^{226}Ra (Bgkg^{-1})	^{232}Th (Bgkg^{-1})
A_1	60.95.1±327.0	ND	16470.4±205.2
A_2	5061.4±253.9	ND	15437.8±177.2
A_3	5809.6±298.7	1004.8±136.1	17523.6±205.1
A_4	5398.8±155.0	ND	18185.1±201.6
A_5	6846.5±302.4	ND	18743.9±202.3
A_6	6065.9±278.8	819.9±120.1	15604.5±194.8
A_7	5894.8±266.4	ND	16975.3±209.4
B_1	4772.5±230.4	661.4±171.1	14793.4±165.4
B_2	4299.4±197.2	1302.8±165.3	14954.4±170.8
B_3	6037.0±287.7	681.1±179.0	16040.9±179.3
B_4	5172.1±288.4	ND	15519.9±189.3
B_5	5892.1±258.6	ND	17487.1±209.9
B_6	5697.1±268.2	ND	14217.7±172.2
B_7	5393.5±277.9	ND	14229.1±185.2
B_8	4147.4±257.3	ND	11483.6±161.0
C_1	6986.1±315.5	ND	20509.6±215.3
C_2	4821.9±245.2	ND	14097.5±161.7
C_3	5244,2±2480	ND	14898.8±192.3
C_4	5499.9±280.0	674.2±111.6	18882.4±218.1
C_5	6374.4±298.7	ND	16842.0±229.0
C_6	6129.6±287.7	ND	16294.5±211.7
C_7	382.2±205.9	ND	18214.5±134.1
C_8	2849.0±230.3	1217.1±191.3	14602.9±205.7
D_1	6415.7±279.7	609.8±179.2	18528.8±216.1
D_2	5226.5±266.4	ND	14167.5±213.0
D_3	5535.1±266.1	564.4±106.0	14921.4±156.5
D_4	7081.6±278.8	549.3±173.7	19972.5±202.1
D_5	9021.6±373.7	364.9±176.4	22583.3±247.7
D_6	909.3±154.4	826.2±96.7	5055.5±102.0
D_7	7899.8±359.7	533.1±115.0	20222.7±218.1
D_8	10423.2±406.6	864.2±196.8	27930.0±257.8

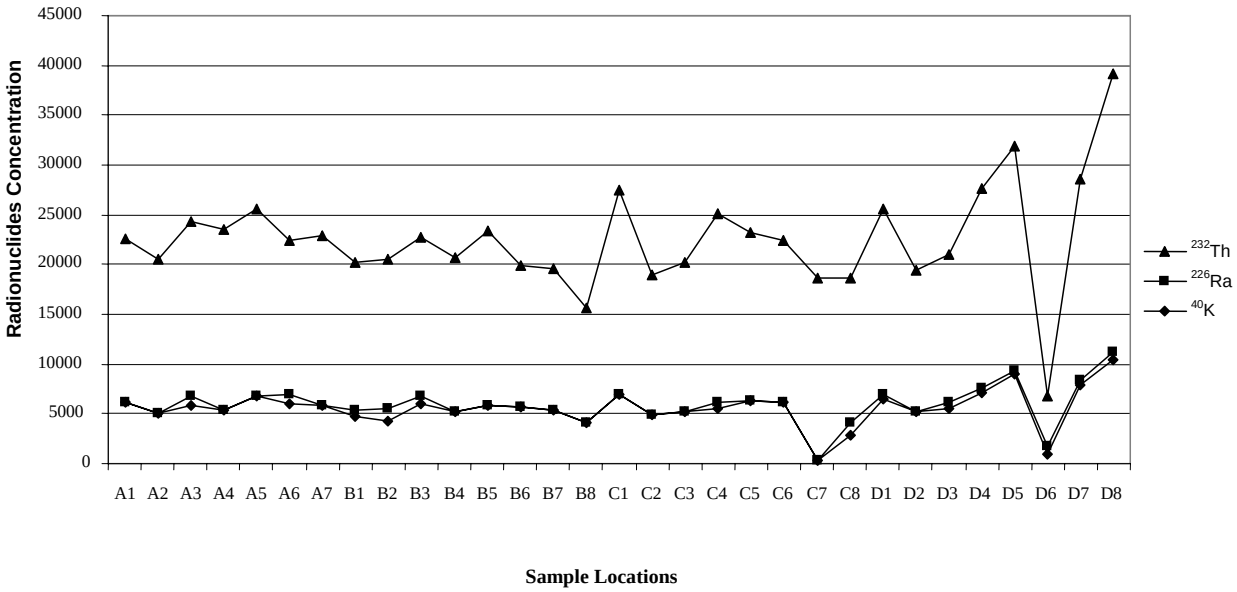


Figure 1: Graph of Concentrations of Radionuclides ^{232}Th, ^{226}Ra and ^{40}K against sample locations

From the graph above, it has been seen that as CPS (counts per seconds) increases concentration/activity increases too both in thorium and potassium. Both the increase in the CPS and concentration has no specific interval. A sharp increase was observed from 12.8039 – 24.4891Bqkg^{-1} in thorium concentration and from 12.8039 – 24.4891Bqkg^{-1}) in potassium concentrations, which showed that at C8-D8 of both thorium and potassium, there were much active radionuclide. Fig 1 also showed an increase of CPS with concentration in Radium with CB8 not detected. Also from the graph in fig. 1, it showed an increase in depth as concentration/ activity of nuclides increases, but at A_6 (15604.5) it decrease instead of increasing but then went up again to A_7 (16975.3), which goes to tell us that as depth increases concentration also increases.

CONCLUSION

The high concentrations of ^{232}Th of D_8 (27930.0$\pm$257.8Bqkg^{-1}), D_5 (6846.5$\pm$1302.4Bqkg^{-1}) and D_7 (6846.5$\pm$1302.4Bqkg^{-1}) in the mine tailings should be of serious concern to government and the environmentalist in the state as this further suggests the over exposure of the mill workers and the danger the general public tends to face.

In general, the high concentrations of these elements is in the tailings particularly, Thorium needs to be monitored before disposal. Also, there is the need for the government to advice and educate the people on the dangers or hazards associated with these by-products of tin (tailings) and also of using these tailings for building purposes. Finally, smokers should be seriously warned and educated on the dangers of smoking during and after mining and milling activities. The main health concern for environmental exposures is general bone cancer (Muazu *et al.,* 2004).

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