



Measurement of Radioactivity Concentration in Pegmatite Rock Samples and Dose Assessment from Wamba in North Central Nigeria

Taiwo Bukola Fasiku ^{a*}, Musbau Kewulere Fasasi ^b
and Adebimpe Amos Amosun ^b

^a Department of Physics, Ajayi Crowther University, Oyo, Nigeria.

^b Center for Energy and Research Development, Obafemi Awolowo University, Ile- Ife, Nigeria.

Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Aims: To evaluate the radiological impact parameters on mine workers.

Study Design: The research work was carried out by using a gamma-ray spectrometer with a NaI (TI) detector.

Place and Duration of Study: Centre for Energy and Research Development (CERD), Obafemi Awolowo University, Ile- Ife, Osun State, Nigeria between September 2016 and June 2017.

Methodology: The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K were determined in ten samples which were used to evaluate the absorbed dose rate, radium equivalent activity, external and internal hazard indices, representative level index, and annual effective dose equivalent.

Results: The average activity concentrations of ²²⁶Ra (24068.11 Bq kg⁻¹), ²³²Th (387.72 Bq kg⁻¹), ⁴⁰K (9509.24 Bq kg⁻¹) and absorbed dose rate (11720.77 nGy h⁻¹) were higher than their respective recommended world mean values of 35 Bq kg⁻¹, 45 Bq kg⁻¹, 420 Bq kg⁻¹ and 59 nGy h⁻¹ by UNSCEAR. The mean values of radium equivalent activity, external hazard index (H_{ex}), internal hazard index (H_{in}), representative level index, and annual effective dose equivalent of the area under study were determined as 25346.82 Bq kg⁻¹, 68.52, 135.43, 357.95, and 14374.36 μSv y⁻¹ respectively. This study revealed that all the radiological parameters were higher than their respective recommended world average values.

*Corresponding author: E-mail: ayenitaiwobukola@gmail.com;

Conclusion: With the high concentrations obtained in this study, it is therefore concluded that this may pose a serious health risk to the miners and the general public. It is therefore advised that necessary guidelines should be provided for the exploration of minerals in this mining site.

Keywords: Mining; radiological protection; spectrometer; pegmatite; absorbed dose rate.

1. INTRODUCTION

Human exposure to radiation can be from either natural or artificial radioactivity. Activities like the mining of natural resources such as uranium and other minerals expose a lot of people to dangers which have generated a need for regulations [1,2]. The naturally occurring radionuclide materials (NORMs) have been in existence since human creation. This has been observed to have spread across the earth's crust with the presence of many radioactive elements such as Uranium (U) and Thorium (Th) which are referred to as parent's radioisotopes and Potassium (K) which is also seen in geological formations of soils, rocks, plants, water, and air [3-5]. The level of radiation exposure can be determined by the type of rock that is mined. Higher radioactivity is observed to be mostly present in igneous rocks such as granite and pegmatite while lower levels of radioactivity are associated with sedimentary rocks with the exceptions of Shale and Phosphate rocks having a relatively high amount of radioisotopes [6]. The level of radionuclides in mined rocks helps to know the extent of radioactive pollutants infused into the surroundings [7]. Therefore, the measurement of radioisotopes in rock samples before use is very important in order to prevent radiation exposure.

Several researches have been conducted to estimate natural radiation exposure and likely radiological hazards on people [8-13]. Some of these studies have established that the accumulation of ingested radionuclides through food, water, and inhalation of tiny particles could have serious health repercussions like leukemia, bone cancer, and high blood pressure [14,15].

Other studies also revealed that the activity concentrations are not particular to a kind of rock alone but the release of these radionuclides over time causes cancer and other related health hazards which can later damage some essential organs in the body [16-19]. According to [20], it was revealed that in an environment of uranium operational mining sites, 68.33 % of the people living around this site do die before age 62.

Presently explorations of minerals in the area under study are done by small-scale miners whose activities are dangerous and are not controlled by any regulated bodies. In that case, the illegal mining currently going on in the area reveals that the Wamba pegmatites may be of economic importance. It is, therefore, necessary to unveil more pieces of information on the natural resources so that their economic mineral potential can be assessed.

This study focused on the extent of exposure of the miners to naturally occurring radionuclide materials (NORMs) by evaluating the radiological parameters and comparing with other literatures and with the United Nations Scientific Committee on Atomic Radiation Report [7] average values.

2. MATERIALS AND METHODS

2.1 Study Area

Wamba town is located mid - northern part of Nasarawa State, Nigeria (Fig. 1) between Longitudes 8° 30'E and 8° 40'E and Latitudes 9° 00'N and 9° 07' N. It has an area of 1,156 km² and a population size of 72,894 people [21]. The pegmatite field of Wamba is one of the Pan-African rare - metal pegmatite broad belt which stretches from southwestern to central Nigeria [22].

It is found covering about 45 km² within the Pan-African reactivation zone. The exploration of minerals in this area started as early as 1912 with tin-bearing pegmatites in the 40 km of Wamba area and ceased in the 1960s. The study area is characterized by hosting several hills with an average temperature of 31° C. Due to the peculiarity of this area mining activities are predominant. Therefore the collection of pegmatite rock samples was done randomly to survey the natural radiation in the mining area. A total of ten samples were collected for radiation measurement and absorbed dose rate.

2.2 Sampling

A total of 10 pegmatite samples collected randomly from the mentioned study area were

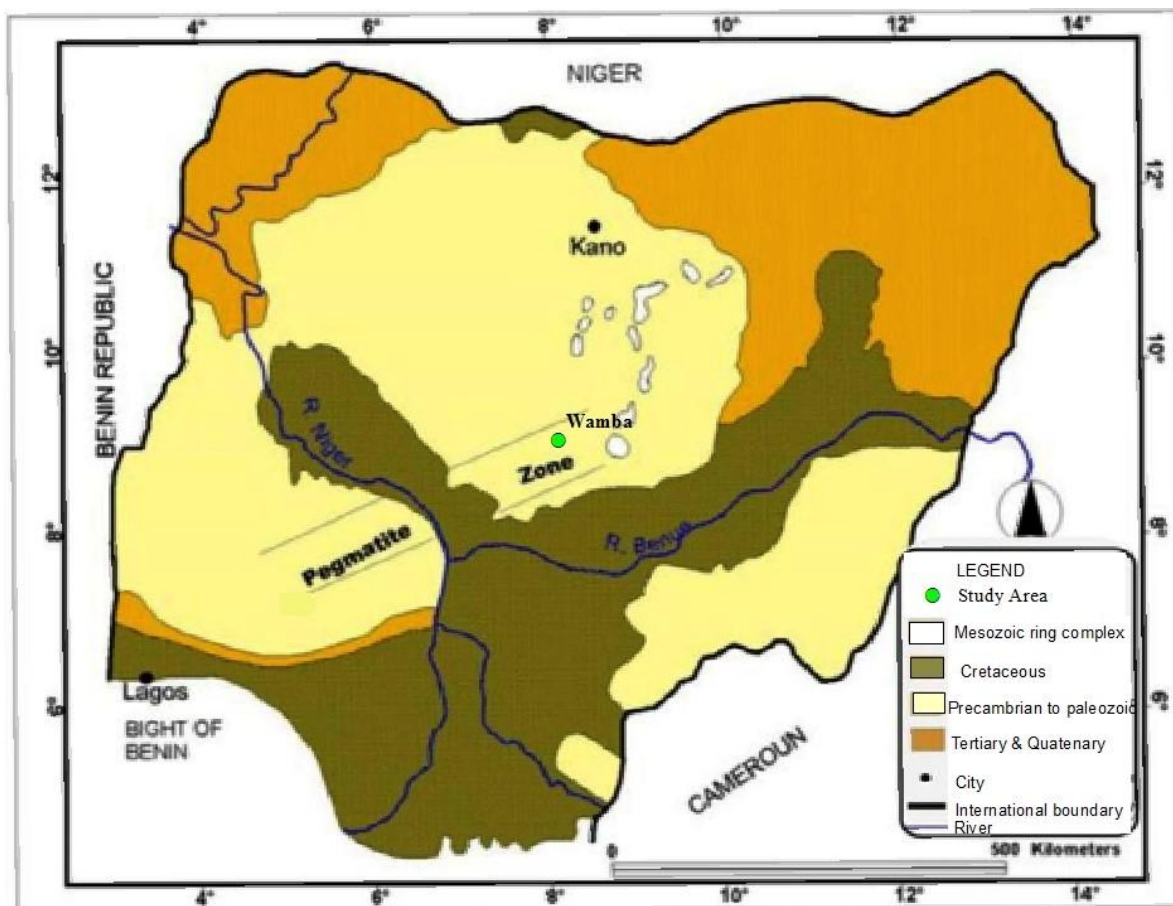


Fig. 1. General geology of Nigeria showing the location of the pegmatite zone and the study area [24]

left to dry for some days under a thorough laboratory conditions in order to remove any liquid present. These rock samples were pulverized in a Rocklab milling machine (Model C RC3E, Serial number 1288) situated at the Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile- Ife Osun State, Nigeria. The samples were oven dried at 100 °C for 24 hours to ensure that they are free of humidity. The samples were sieved with a 2 mm mesh and thereafter, 150 g of each sample was packed into a cylindrical plastic container tightly closed, sealed, and stored for 28 days to ensure secular equilibrium between the parent radionuclides ^{226}Ra , ^{232}Th , and their respective daughters [23]. An identical empty plastic container was sealed to serve for background counting.

2.3 Instrumentations

The activity measurement was carried out using a well-calibrated 76 mm × 76 mm Thallium

activated Sodium Iodide (NaI(Tl)) detector with coupled photomultiplier tube at the Centre for Energy and Research Development (CERD), Obafemi Awolowo University, Ile- Ife Osun State, Nigeria. The detector is shielded with a lead castle thickness of 5 cm and a resolution of about 8% at an energy of 0.662 MeV (^{137}Cs). The preamplifier provided a voltage of 750 V to the detector. The multichannel analyzer (MCA) connected through a preamplifier provides a stabilised spectrum which can be analyzed on an IBM personal computer. The energy and efficiency calibrations of the detector were done using a standard source which was shown in Table 1, with known gamma-ray energies prepared by the Isotope Products Laboratories, California, U.S.A. The calibration was performed by determining the sources of known radionuclides with energy ranging from 60 keV to 1332.50 keV. This was achieved by measuring the standard sources emitting known gamma-ray energies (^{137}Cs) 661.66 keV, (^{60}Co) 1173.24 and 1332.50 keV with channel numbers of 738, 1504,

and 1743 respectively. The standard sources have the same geometry with that of the sample. The gamma counting was done for 18000 s.

The efficiency of a detector measures the number of pulses that occur for a given number of incident gamma rays, i.e., the fraction of all the photons that are emitted by the source or sample, which cause an event in the detector. This depends on the following: the detector features, counting geometry used, the sample shape, volume, and distance of the sample to the detector.

As soon as the counting time started, the computer started recording the photo peaks and the same time storing them. The accumulation of counts at a specific energy range is due to emitted photons from the sample in addition to the one from background interference. However, if the background is constant, then a sample with high activity will accumulate relatively more counts in the peak compared to the background. Therefore, as the total area of the peak increases, the relative background size reduces as well as the proportional background error. After the counting elapsed, the peaks were displayed. The radioactivity analysis was carried out by spectra- analysis program, SAMPO 90, which was used to analyze the samples on the computer. The spectrum of each sample was analyzed and the identification of radionuclides were determined from the energy of the peak centroids. Also as a sample activity increases, the estimated standard error decreases, and as the activity decreases, the relative error increases. An empty container of the same geometry was sealed and counted for gamma-ray distribution count. This served as a background measurement which was done repeatedly at regular intervals for quality control. The spectrum obtained was recorded for analysis.

Table 1. Radionuclides used for Energy Calibration of NaI (TI)

Radionuclides	Energy (keV)	Channel number
¹³⁷ Cs	661.66	738
⁶⁰ Co	1173.2	1504
⁶⁰ Co	1332.50	1743

In Table 2, the activity concentration of the daughter radionuclide (²²⁶Ra) in the samples was obtained using the γ-ray peaks of 1120 keV from ²¹⁴Bi which is equivalent to the concentration of

(²²⁶Ra) in the samples. The daughter radionuclide ²²⁸Ac was determined from its 911 keV γ-ray peak which was chosen as an indicator of ²³²Th. The concentration of ⁴⁰K was determined by measuring the photo peak of 1460 keV emitted during its decay.

The background radioactivity distribution in the environment around the detector was determined by counting the empty sealed container for 18000 s. The background measurements were repeated at regular intervals for quality control. The spectrum obtained was recorded for analysis. The specific activity concentration of the natural radionuclide, A, ($Bq\ kg^{-1}$) of each sample in this study was evaluated using Eq. (1) [25, 26].

$$A = \frac{C}{p_{\gamma}wt\varepsilon} \tag{1}$$

where C is the net counts of the radioisotope in the samples, p_{γ} is the absolute gamma-ray emission probability (gamma ray yield), w is the net dry sample weight (kg), t is the sample measurement time (18000 s) and ε is the absolute efficiency of the detector.

Table 2. Spectral energy windows used in analysis using NaI(Tl) gamma spectroscopy

Element analysed	Isotope Used	Energy (keV)
²³² Th	²²⁸ Ac	911
²²⁶ Ra	²¹⁴ Bi	1120
⁴⁰ K	⁴⁰ K	1460

The specific activity concentration (C_x) of the radionuclide was obtained by direct comparison with the same radionuclide in a given standard. The specific activity in the sample and the corresponding specific activity in the standard (C_s) are related by:

$$C_x = C_s \frac{M_s A_x}{M_x A_s} \tag{2}$$

where M_s is the mass of the standard, M_x is the mass of the sample, A_s is the area of the standard and A_x is the area of the sample.

The concentration of natural radionuclides to the absorbed dose rate in the air (D_R) depends on the concentration of the radionuclides present in the pegmatite rock samples. Absorbed dose radiation dose in an exposed person per unit mass at 1m above the ground level is defined as the quantity of ionizing energy present in the

body in Gray/second... (Gy/s) . This was determined using Equation (3).

$$D_R(nGy h^{-1}) = 0.462 C_{Ra} + 0.604 C_{Th} + 0.0417 C_K \quad (3)$$

where C_{Ra} , C_{Th} , and C_K are the activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K , in the rock samples ($BqKg^{-1}$) respectively.

2.4 Evaluation of Radiological Impact Parameters

2.4.1 Radium Equivalent Activity (Ra_{eq})

The radium equivalent activity ($Bq kg^{-1}$) represents a weighted sum of activities of ^{226}Ra , ^{232}Th and ^{40}K . Ra_{eq} is related to the external gamma dose and internal dose due to radon and its daughters. In this study, Ra_{eq} was calculated using equation (4) [27].

$$Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.077 C_k \quad (4)$$

where C_{Ra} , C_{Th} , and C_K are the activity concentrations ($Bq kg^{-1}$) of ^{226}Ra , ^{232}Th , and ^{40}K , in the rock samples respectively.

2.4.2 External Hazard Index (H_{ex})

The external hazard index (H_{ex}) connotes the level of health hazard incurred from gamma radiation that comes from the materials used in building walls of a dwelling. Equation (5) was used in calculating for external hazard index [7].

$$H_{ext} = \left(\frac{C_U}{370}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \quad (5)$$

where C_{Ra} , C_{Th} , and C_K are the activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K ($Bq kg^{-1}$) in respectively.

2.4.3 Internal Hazard Index (H_{in})

The internal hazard index (H_{in}) also defined as the inhalation exposure of the respiratory organs to radon and its daughter nuclei inside a house was calculated using Equation (6) [7].

$$H_{int} = \left(\frac{C_U}{185}\right) + \left(\frac{C_{Th}}{259}\right) + \left(\frac{C_K}{4810}\right) \quad (6)$$

C_{Ra} , C_{Th} , and C_K are the activity concentrations in $Bq kg^{-1}$ of ^{226}Ra , ^{232}Th , and ^{40}K respectively.

2.4.4 Representative Level Index (I_γ)

To ascertain whether a particular sample conforms to the standard dose limit set for

building materials [28], the representative level index or gamma index (I_γ) is used to estimate the level of γ – radiation hazard associated with the natural radionuclides in specific investigated samples. Equation 7 was used to evaluate the gamma index for samples in this study.

$$I_\gamma = 0.0067C_U + 0.01C_{Th} + 0.00067 C_K \quad (7)$$

C_{Ra} , C_{Th} , and C_K are the activity concentrations in $Bq kg^{-1}$ of ^{226}Ra , ^{232}Th , and ^{40}K respectively.

Therefore, (I_γ) must be ≤ 1 which also corresponds to an annual effective dose of $\leq 1mSv$.

2.4.5 Annual Effective Dose Rate (AEDR)

This is the dose that an individual receives after being exposed to radiation outside especially through the air. The absorbed doses evaluated in $nGy h^{-1}$ were converted to annual effective dose in $\mu Sv y^{-1}$ by [7]. This was calculated using equation (8).

$$AEDR_{outdoor}(\mu Sv y^{-1}) = D_R(nGy h^{-1}) \times 8760 (h y^{-1}) \times 0.7 (Sv Gy^{-1}) \times 0.2 \times 10^{-3} \quad (8)$$

where D_R is the absorbed dose rate ($nGy h^{-1}$), 8760 is the time conversion factor ($h y^{-1}$), 0.7 is the dose conversion factor ($Sv Gy^{-1}$) and 0.2 is the outdoor occupancy factor.

3. RESULTS AND DISCUSSION

The daughter lines for ^{226}Ra and ^{232}Th used in this study were ^{214}Bi and ^{228}Ac respectively. The results of the radioactivity concentrations were presented in Table 3. It was revealed that all the three radionuclides ^{226}Ra , ^{232}Th , and ^{40}K investigated in the pegmatite rock samples were observed to have the highest activity concentrations in sample 1 and the lowest concentrations in samples 6. The mean activity concentrations of ^{226}Ra measured was $24068.11 \pm 368.03 BqKg^{-1}$ which is higher than the world average value of $35 Bq kg^{-1}$. The mean activity concentration of ^{232}Th estimated in this research was $387.72 \pm 18.50 Bq kg^{-1}$, and thus higher than the world average value of $45 Bq kg^{-1}$. The average activity concentration of ^{40}K was $9509.24 \pm 335.11 Bq kg^{-1}$ which is higher than the world average value of $420 Bq kg^{-1}$. Both the ^{226}Ra and ^{40}K contributed to the increase in the activity concentrations in the

pegmatite rock samples. The possible reasons for higher values of ²²⁶Ra could be as a result of geological differences in the formation of the rock, cosmogenic and terrestrial radionuclides. The increase in the activity concentration of ⁴⁰K is because of the presence of aluminum silicates of potassium, sodium, calcium, or barium.

Table 4 revealed the estimated absorbed dose rate (D_R) which ranged from 362.49 to 31888.00 nGy h⁻¹ with a mean concentration value of 11720.77 nGy h⁻¹. The mean value of the absorbed dose rate was found to be higher than the world average value of 59 nGy h⁻¹ [7]. This could be a result of large quantity of uranium and other radionuclides present in the rock samples. The Radium activity equivalent (Ra_{eq}) values obtained varied from 777.87 to 68878.11 Bq kg⁻¹ with a mean value of 25346.82 Bq kg⁻¹. The Ra_{eq} mean value exceeded the maximum limit of 370 Bq kg⁻¹ recommended by [29]. The evaluated values of the external hazard

index (H_{ex}) and internal hazard index for the pegmatite rock samples in this study ranged from 2.11 to 186.16 with a mean value of 68.52. The calculated values of (H_{in}) for the rock samples ranged from 4.04 to 364.02 with a mean value of 135.43 as shown in Table 4. Both external and internal indices mean values were higher than the world average value of 1 unity [7].

The representative level index I_γ was calculated using equation (7). For the radiation hazard to be negligible it must be less than unity. The evaluated values varied from 14.84 to 1340.49 with an average value of 357.95. The average value obtained is much greater than the world-recommended value of 1 unity. The annual effective dose rate (AEDR) ranged from 444.5 to 39107.4 μSv y⁻¹ with a mean concentration of 14374.36 μSv y⁻¹. The AEDR average value is thus higher than the recommended safety limit value of 70 μSv y⁻¹ for an individual [7].

Table 3. Specific activity of ²²⁶Ra, ²³²Th and ⁴⁰K in pegmatite rock samples in Bq kg⁻¹

Samples	²²⁶ Ra	²³² Th	⁴⁰ K
K1	716.42±11.56	7.98±0.39	650.70±31.83
K2	27755.55±424.28	418.58±19.96	10473.55±12.27
K3	30028.46±459.02	842.49±40.18	16726.56±818.12
K4	5825.98±89.17	82.80±3.96	1627.15±79.60
K5	6616.20±101.24	92.80±4.44	3354.09±164.06
K6	65809.11±1005.94	962.84±45.91	22015.65±1076.81
K7	1950.28±30.10	25.36±1.22	1313.38±64.24
K8	18036.72±275.82	200.65±9.58	6943.71±339.63
K9	38854.63±593.93	550.27±26.24	14765.44±722.20
K10	45087.76±689.23	693.45±33.07	17222.13±842.36
Min.	716.42±11.56-65	7.98±0.39- 962.84	650.70±31.83
Max.	65809.11±101.24	962.84±45.91	22015.65±1076.81
Mean value	24068.11±368.03	387.72±18.50	9509.24±335.11

Table 4. The evaluated radium equivalent Ra_{eq}, dose rate, external hazard index (H_{ex}), internal hazard index (H_{in}) and gamma index of the samples under investigation

Sample code	Ra _{eq}	D	H _{ex}	H _{in}	I _γ	AEDR
K1	777.87	362.49	2.11	4.04	14.84	444.56
K2	29159.18	13277.76	78.82	153.83	570.45	16283.84
K3	32518.68	15067.80	87.89	169.05	620.15	18479.15
K4	6002.83	2808.32	16.41	32.15	118.43	3444.12
K5	7006.78	3250.25	18.93	55.38	135.49	3986.11
K6	68878.11	31888.00	186.16	364.02	1340.49	39107.44
K7	2087.54	970.20	5.63	10.90	40.14	1189.85
K8	18857.49	8738.84	50.96	99.70	367.37	10717.31
K9	40776.53	18888.58	110.20	215.22	792.44	23164.95
K10	47403.18	21955.50	128.12	249.98	920.17	26926.23
Min.	777.87	362.49	2.11	4.04	14.84	444.56
Max.	68878.11	31888.00	186.16	364.02	1340.49	39107.44
Mean value	25346.82	11720.77	68.52	135.43	357.95	14374.36

Table 5. Comparison of the mean activity concentration with some selected studies

Sample used	²³⁸ U	²³² Th	⁴⁰ K	Place	Country	References
Soil	3.00	33.30	122.10	Sango- Ota	Nigeria	[30]
Soil	67.40	507.19	8217.77	Abua/Odua	Nigeria	[31]
Soil	32.52	56.23	403.96	Nasarawa	Nigeria	[32]
Rock	11.51	15.42	4441.06	Asa	Nigeria	[12]
Soil	11.90	17.72	70.44	Maiganga	Nigeria	[33]
Soil	12.14	23.23	270.14	Ile-Ife	Nigeria	[34]
Soil	24.50	51.80	344.90	Anatolia	Turkey	[35]
Rock	12.25	45.17	639.24	Aravali	India	[36]
Rock	24068.11	387.72	9509.24	Wamba	Nigeria	Present study

4. CONCLUSION

The mean activity concentrations of ²²⁶Ra (24068.1±368.0), ²³²Th (387.7±18.5), ⁴⁰K (9509.2±335.1) Bq kg⁻¹ and absorbed dose rate of 11720.77 nGy h⁻¹ were higher when compared to the United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations Sources and Effects of Ionizing Radiation values of 30 Bq kg⁻¹, 35 Bq Kg⁻¹, 400 Bq kg⁻¹ and 60 nGy h⁻¹, respectively. This may pose a serious health risk to the miners and the general public. The result of this study can serve as baseline data for the distribution of radioactivity present in pegmatite rock in the area and also allow more research work to be carried out to investigate the radioactive background level of the area. This will provide necessary guidelines for fruitful exploration of pegmatite so that their economic potential can be of use to the country at large. The illegal mining currently going on could be curbed to the barest minimum so that the general populace will not be prone to radiation hazards.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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