Distribution of radioactive elements of some rocks in South-western Nigeria

Porazdelitev radioaktivnih prvin v nekaterih kamninah jugozahodne Nigerije

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Abstract
Rock samples obtained from two locations in parts of south-western Nigeria were studied to establish the distribution of U, Th and K, present in them. Field data were collected using a gamma ray scintillometer while laboratory studies on the samples included spectrometric analysis, NaI(Tl) spectrometry and geochemical analysis (ICP-MS method). Gamma ray scintillometric reading showed that the outcrops present in the study areas gave consistently higher readings than the overburden materials. The analysis of spectrometric data indicated the presence of other radio-nuclides which are different from the three principal radio-nuclides, 238U, 232Th, and 40K. Results of the geochemical data revealed the presence of radioactive elements like Pb, Ce, La, Ti and Fe2+. This study showed that besides the presence of U, Th and K, other elements like Pb, Ce, La and Ti also contributed to the bulk radioactivity measured in rock samples at the study areas. Zircon, xenotime and possibly thorite are the main radioactive minerals present in the studied rocks.

Key words: Radioactive elements, Radio-nuclides, Radioactive minerals

Izvleček
Vzorce kamnin z dveh lokacij na območju jugozahodne Nigerije smo preiskali, da bi v njih določili vsebnosti U, Th in K. Na terenu smo opravili merjenje z gama scintilometrom, v laboratoriju pa dodatno s spektrometrično analizo z NaI(Tl)-spektrometrijo in geokemično analizo po metodi ICP-MS. Gama scintilometrija je pokazala na izdankih kamnine raziskovanega območja dosledno višje vsebnosti kakor v preperini. Analiza spektrometričnih podatkov nakazuje tudi navzočnost dodatnih radioaktivnih nuklidov, drugačnih od glavnih: 238U, 232Th in 40K. Z geokemično analizo so bile ugotovljene kemične prvine Pb, Ce, La, Ti in Fe2+. Iz raziskave izhaja, da prispevajo k celotni radioaktivnosti kamnin, merjeni na raziskovanem ozemlju, razen U, Th in K, tudi prvine, kot so Pb, Ce, La in Ti. Poglavitini radioaktivni minerali v preučevanih kamninah so cirkon, ksenotim in najbrž torit.

Ključne besede: radioaktivne prvine, radioaktivni nuklidi, radioaktivni minerali
Introduction

Gamma-ray spectrometry survey is an important source of information for soil, regolith, rock and geophysical studies. It is essentially a radioactive geophysical technique and its data is a vital sub-component of the geophysical data required for environmental and geological mapping, soil surveying, and mineral exploration and regolith studies. Measurements of gamma radiation at the surface of rock provide information on the abundances of radioactive elements in the rock. Many naturally occurring elements have radioisotopes, but only K, U and Th decay series have radioisotopes that produce gammarays of sufficient energy and intensity to be measured by gamma-ray spectrometry because they are relatively abundant in the natural environment. K is a major element in igneous rocks but only one of its isotopes, $^{40}$K is naturally radioactive. Although Th is less ubiquitous than U, it is well distributed throughout a considerable pressure-temperature range, chiefly as monazite and thorite, with monazite persisting as the main radioactive mineral of placer deposits. Primary accessory minerals mainly responsible for the bulk of the radioactivity of granitic and intermediate rocks are zircon, sphene, apatite, allanite, xenotime and monazite. Others rarely present but in some cases strongly radioactive, are uranothorite, thorianite, euxenite, thorite, pyrochlore, chevkinite, fluorite, bastnaesite and even davidite. Other possible contributors to radioactivity are haematite, pyrite, columbite, ilmenite and rutile and in alkalic rocks are zirconium and cerium silicates. Although uraninite has been reported as an accessory mineral of intrusive igneous rocks, it has not been demonstrated to be of primary origin.

The intensities of nuclear radiation in any particular environment are in general related to the abundances of natural radioactive elements in rocks and soils of that locality. Gamma ray spectrometric methods have been widely used in measurements of radioactive minerals in soils and basement rocks. In many rocks, high radioactivity values are associated with clusters of mafic minerals; e.g. biotite in granites or riebeckite in alkali granites, largely because radioactive minerals such as zircon, apatite, allanite and sphene with the high calcium granites and an assemblage consisting of »monazite-like« phases, and xenotime are commonly associated with mafic components. Oshin showed that the various rock types constituting the Precambrian rocks (granite-gneisses, porphyritic granites and meta-sediments) of south-western Nigeria are characterized by different U and Th contents and that the granite-gneisses and the coarse porphyritic granites corresponding to the older granite series are characterized by high Th and U contents respectively, while the meta-sediments are the most depleted in these radioactive elements. He also suggested an igneous origin for the gneisses (migmatite gneiss and granite gneiss) based on their contents of radioactive elements. Jibiri et al. studied the radioactivity levels of granitic rock in Jos (North central), porphyritic granite in Abeokuta (Southwest), augen gneiss in Ibadan (Southwest), shale and coal in Enugu (Southeast) Nigeria which are situated within high natural background radioactive areas and the results were compared with in-situ gamma-ray measurements made at the localities where samples were collected. It was concluded that $^{232}$Th was the major contributor of gamma absorbed doses that are received by the population in all different environments in the study areas.

The detection of radioactive mineral deposits since the mid-1940s by geologists and untrained prospectors yielded the discovery of an important number of new deposits. Many were stumble upon by untrained prospectors using Geiger counters and whose knowledge of uranium geology was insufficient to prejudice them against these »unfavourable« areas and who operated on the basis of »uranium is where you find it«. Modern prospecting methods still follow a similar pattern but with a more systematic approach using equipments with a higher degree of sensitivity like the gamma-ray scintillometer and air-borne gamma-ray spectrometer.
Radioactive raw materials are of great importance because they can be used as supplement to the energy obtained from fossil fuel hence in the last few decades surveying for these minerals have been in practice\textsuperscript{[10–12]}. In Nigeria, the increasing demand of energy and the instability of petroleum supplies make atomic energy an alternative to the energy problems. Although criticisms might arise related to its safe use, the utilization in the area of electricity generation as a supplement to hydro-electricity supply is a possibility. This study therefore aims at determining the distribution of radio-elements such as uranium, thorium and potassium in the different rock types available within the study areas; and to determine the concentration of the major, minor and trace elements present within the samples in order to constrain the possible complimentary role of other radioactive elements to the total radioactivity of the samples analysed. An attempt to establishing the relationship between radioactivity levels and elemental concentrations of the various radioactive elements present within the samples and identifying the different minerals which possibly host these radio-elements will be examined.

**Site description and geological setting**

The study area falls within sheet 59 of the geological map of south-western Nigeria and comprise an area of approximately 11 655 km\(^2\) (4 500 mi\(^2\)). Location 1 is opposite Lagelu industrial estate in Ibadan, Oyo State about 700 m from the Ogunpa channelization and is within latitudes 7° 19.964 N–7° 20.01 N and longitudes 3° 52.741 E–3° 53.224 E. Location 2 is at Odeda, in Ogun State within latitudes 7°14.288 N–7°15.662 N and longitudes 3°32.076 E–3°45.116 E (Figures 1 and 2). As a result of the latitudinal location, Ibadan and Odeda have the characteristic of West African monsoon climate, marked by distinct seasonal shift in the wind pattern. Between March and October, the area is under the influence of the mist maritime southwest monsoon winds which blow inland from the Atlantic Ocean. The dry season occurs from November to February.
when the dry dust laden wind blows from the Sahara Desert. The mean annual rainfall for the areas is 1 229.39 mm. The mean maximum annual temperature of 28.8 °C occurs in February while the mean minimum temperature of 24.5 °C occurs in August.\[13\]

Geologically, the study area falls within the Precambrian basement complex of Nigeria which occupies about half of the total area of the country. It has been intruded by the Younger Granites of Jurassic age. Cretaceous to recent sediments occupy the other half of the area (Figure 3).

The basement complex of Nigeria has been described as consisting mainly of a variety of gneisses, schists, amphibolites and quartzites most of which have been migmatised\[14\]. Other major rock types include deformed and undeformed granitic and dioritic intrusives\[15–17\].

The rock types found in the basement complex rocks of Ibadan can be grouped into major and minor rock types. The major rock types are quartzites of the meta-sedimentary series and a migmatitic complex comprising banded gneisses, augen gneisses and migmatites. The minor rock types include pegmatites, quartz veins, aplites, diorites, amphibolites and xenoliths. Pegmatite is a major intrusion in the banded gneiss, cross-cutting the rock in an East-West direction. The pegmatites are typically coarse grained with quartz and sodic feldspars as dominant minerals. Tourmaline (black variety) is present in small amounts. Most of the outcrops are well exposed as a result of the constructed road (expressway). Many of the rocks have been covered by a veneer of overburden materials. Field investigations have shown that rock types in Odeda area comprise migmatites, coarse grained porphyritic granites, granodiorites, quartz and fine grained porphyritic granites\[18\].

**Methods of study**

The radiometric mapping in the profiling mode was carried out using a Gamma Ray Scintillometer (GRS) model GR101A. At Lagelu, two sets of profiles were run parallel to one another in the north-south direction while eleven smaller profiles were run perpendicular to these two sets of profiles in the east-west direction (Figure 2). All measurements were taken using the full-scale range of 0.3 k and an audio-signal of 50 % of the GRS. The calibration equation

\[ Y = 19 514 X \]

\[ \text{Y in count per second in terms of the dose rate X in } \mu \text{Sv h}^{-1}. \]

The equipment measures the in-situ gamma ray emitted from natural radioactive sources such as \(^{40}\)K, \(^{238}\)U, \(^{222}\)Th and their daughter products such as \(^{87}\)Rb as well as anthropogenic radio nuclides such as \(^{137}\)Cs, \(^{90}\)Sr produced mostly from atomic bomb testing and nuclear explosions or accidents. The radiometric mapping was done to isolate outcrops of high emission counts to make rock sampling easier.

Gamma ray spectrometry analyses of eleven samples were carried out using Canberra 7.6 cm × 7.6 cm NaI(Tl) (Model No 802-series) detector coupled to Canberra serial 10 plus Multichannel Analyzer (MCA) through a pre-amplifier base. The gamma ray transition line of 1 460 MeV was used for the measurement of \(^{40}\)K, 1 760 MeV of \(^{214}\)Bi and 2 615 MeV of \(^{208}\)Ti were respectively used for the measurements of \(^{238}\)U and \(^{232}\)Th.

For geochemical and mineralogical analyses, eleven samples were taken based on the number of rock outcrop encountered when using the gamma ray scintillator in order to give a better representation of the rocks in the mapped area. Major element oxides and trace element geochemistry was carried out using inductively coupled plasma (ICP) analytical procedures. The ICP method was chosen because of its proven sensitivity, precision and accuracy for many elements and its ability to determine the concentration of a large number of elements simultaneously, thereby reducing the cost and time of analysis. Slides of the different rock samples obtained were made to ascertain the type of heavy minerals plus accessory minerals present and also to identify the possible minerals that might host the radio-nuclides (U, Th, K) as well as other radioactive elements such as Pb and Ce.
Results and discussion

Graphical representations of two profile lines corresponding to the measurement of radioactivity level versus distance are shown in Figures 4 and 5. Generally at both locations (Odeda and Lagelu) maximum peaks were recorded at or near the outcrops. Minimum peaks were recorded across the basement complex rocks area covered by thin or thick overburden materials which are thought to be weathered products of the parent rock. The contributions of radioactivity by the different primary radionuclides $^{238}U$, $^{232}Th$ and $^{40}K$ are presented in Table 1. This shows the different inputs of these elements in the total gamma ray measured. The largest contribution in terms of concentration is from $^{40}K$ followed by $^{232}Th$ and then $^{238}U$. The largest contributor in any given locality in terms of total absorbed dose is $^{232}Th$. The high radioactivity concentration of radionuclide displayed by $^{40}K$ in all the samples does not imply high contribution of dose or dose exposure rather it is a function of the energy and number of gamma rays emitted by the radionuclide$^{[19]}$. The variations in radiation level of the radionuclides may be attributable to the local geology of the area under consideration$^{[20]}$. For the plots of $^{40}K$, $^{238}U$, and $^{232}Th$ against in-situ gamma ray readings, a weak negative correlation was obtained for $^{40}K$ versus gamma ray readings (−0.605) with correlation being significant at 0.049 levels (2-tailed) i.e. normal distributions curve. Weak positive and negative correlations (0.587 and −0.592) were established for $^{232}Th$ versus gamma ray readings and $^{238}U$ versus $^{40}K$ with correlation being almost significant at (0.058 and 0.055 levels) respectively for a two – tailed distribution. For the other radionuclide ($^{238}U$) no correlation was established with in-situ gamma ray readings and other radio-nuclides of $^{232}Th$ and $^{40}K$ (Table 2). Table 3 shows the average concentration of some elements in rocks from the study areas.

Figure 4: Plot of radioactive count against distance at Lagelu.

Figure 5: Plot of radioactive count against distance at Odeda area.
### Table 1: Concentrations w of $^{238}$U, $^{232}$Th and $^{40}$K of rocks within the study area in ($\times 10^{-6}$). (Conversion factor was introduced to change the values from Bq kg$^{-1}$/($\times 10^{-6}$) γ-ray reading

<table>
<thead>
<tr>
<th>Location</th>
<th>Sample name</th>
<th>Count</th>
<th>$w$ ($^{40}$K)/%</th>
<th>$w$ ($^{238}$U)/($\times 10^{-6}$)</th>
<th>$w$ ($^{232}$Th)/($\times 10^{-6}$)</th>
</tr>
</thead>
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<td>ODEDA</td>
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<td>2.400 7</td>
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<td></td>
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<td>4.127 9</td>
<td>3.875 2</td>
<td>94.54</td>
</tr>
<tr>
<td></td>
<td>L$_2$T$_2$</td>
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<td>1.735 9</td>
<td>77.59</td>
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<td>5.051 5</td>
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<td>62.67</td>
</tr>
<tr>
<td></td>
<td>L$_3$T$_4$</td>
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<td>4.965 4</td>
<td>3.104 0</td>
<td>29.34</td>
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<td>L$_4$T$_4$</td>
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<td>4.283 2</td>
<td>2.117 3</td>
<td>38.38</td>
</tr>
<tr>
<td></td>
<td>L$_4$T$_5$</td>
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<td>4.486 2</td>
<td>1.848 8</td>
<td>86.83</td>
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<td>4.015 0</td>
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<td>2.117 3</td>
<td>38.38</td>
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<td>L$_7$T$_7$</td>
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<td>4.486 2</td>
<td>1.848 8</td>
<td>86.83</td>
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<tr>
<td></td>
<td>L$_8$T$_8$</td>
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<td>4.043 2</td>
<td>62.67</td>
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<td>L$<em>10$T$</em>{10}$</td>
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<td>8.100 8</td>
<td>4.500 8</td>
<td>14.07</td>
</tr>
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</table>

*Correlation is significant at the 0.05 level 2-tailed i.e. at 95% confidence interval

### Table 2: Correlations between gamma ray readings: $^{40}$K, $^{238}$U, $^{232}$Th

<table>
<thead>
<tr>
<th>γ-ray reading</th>
<th>$^{40}$K</th>
<th>$^{238}$U</th>
<th>$^{232}$Th</th>
</tr>
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<tr>
<td>$^{40}$K</td>
<td>Pearson Correlation (r)</td>
<td>1.00</td>
<td>-0.605</td>
</tr>
<tr>
<td></td>
<td>Significant (2-tailed)</td>
<td>-</td>
<td>0.049</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>Pearson Correlation (r)</td>
<td>0.605</td>
<td>1.000</td>
</tr>
<tr>
<td></td>
<td>Significant (2-tailed)</td>
<td>0.049</td>
<td>-</td>
</tr>
<tr>
<td>$^{232}$Th</td>
<td>Pearson Correlation (r)</td>
<td>-0.203</td>
<td>0.466</td>
</tr>
<tr>
<td></td>
<td>Significant (2-tailed)</td>
<td>0.550</td>
<td>0.149</td>
</tr>
</tbody>
</table>

* Correlation is significant at the 0.05 level 2-tailed i.e. at 95% confidence interval

### Table 3: Concentration of major (w%), minor (w%) and trace w/($\times 10^{-6}$) elements in rock samples within the study area

<table>
<thead>
<tr>
<th>Location</th>
<th>Sample name</th>
<th>SiO$_2$ %</th>
<th>Al$_2$O$_3$ %</th>
<th>CaO %</th>
<th>MgO %</th>
<th>FeO %</th>
<th>NaO %</th>
<th>K$_2$O %</th>
<th>TiO$_2$ %</th>
<th>P$_2$O$_5$ %</th>
<th>U ($\times 10^{-6}$)</th>
<th>Th ($\times 10^{-6}$)</th>
<th>K ($\times 10^{6}$)</th>
<th>Pb ($\times 10^{-6}$)</th>
<th>Zr ($\times 10^{-6}$)</th>
<th>Ce ($\times 10^{-6}$)</th>
<th>La ($\times 10^{-6}$)</th>
<th>Nb ($\times 10^{-6}$)</th>
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<td>4.84</td>
<td>0.66</td>
<td>0.46</td>
<td>4.6</td>
<td>102.3</td>
<td>4.02</td>
<td>31.5</td>
<td>94.4</td>
<td>311</td>
<td>127.5</td>
<td>20.5</td>
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<td>13.99</td>
<td>64.79</td>
<td>2.02</td>
<td>2.95</td>
<td>5.72</td>
<td>2.17</td>
<td>4.89</td>
<td>1.48</td>
<td>0.66</td>
<td>4.5</td>
<td>118</td>
<td>4.06</td>
<td>32.4</td>
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<td>98.4</td>
<td>4.2</td>
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<td>9.0</td>
<td>9.0</td>
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</tr>
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</table>

*Samples from Odeda are porphyritic granite
*Samples from opposite Lagelu Industrial Estate are pegmatite
compared to the average crustal and granite concentrations. A plot of Na₂O + K₂O against SiO₂ (Figure 6) showed the granites from Odeda as subalkaline rocks and the Lagelu pegmatites as alkali feldspar granites. A plot of PbO against alumina indicated that all the samples are strongly metaluminous to weakly peraluminous, with the pegmatites plotting higher up due to higher PbO content and mineralogy of the samples (Figure 7). In order to establish a link between in-situ gamma ray readings and geochemically derived K, U and Th, a plot of these elements was made against the gamma ray readings obtained and matched with those of radiogenic ⁴⁰K, ²³⁸U, ²³⁵Th. A better correlation was obtained for ²³⁵Th and Th vs. gamma ray readings when compared to the matches obtained for uranium and potassium. This indicates that thorium is the main contributor to the gamma ray radiation/emission detected by the scintillometer (Figures 8a, b and c). Since it is clear that the principal radionuclide responsible for the bulk of the radiation is thorium, plots of the concentration of other potentially radioactive elements were plotted against thorium concentration to establish a link between their levels of activity.

- For the plot of Th versus PbO, values \( r^2 = 0.77 \), \( p < 0.001 \) were obtained indicating a very high degree of correlation (Figure 9).
- Plot of U versus thorium did not show any correlation (\( r^2 = 0.17 \), \( p < 0.70 \), Figure 10).
- Plot of Zr versus thorium did not show any correlation implying that Zircon though radioactive does not contribute significantly to thorium’s activity (\( r^2 = 0.20 \), \( p < 0.16 \), Figure 11).
- Plot of TiO₂ versus Th showed a very strong positive correlation with \( r \) value of 0.927 i.e. \( r^2 = 0.86 \), \( p < 0.001 \) thus indicating that TiO₂ contributes significantly to thorium’s activity (Figure 12). Both U and Th possess ionic radii and charges which prevent them from fitting comfortably into the lattices of any of the common major rock forming minerals of granite, but they fit into certain accessory minerals such as Zircon, sphene, allanite, rutile and apatite. In Zircon for example, U and Th in the (+4) states can substitute for (+4) Zirconium.
Figure 8c: Regression plot of geochemical/spectrometric observations of Th and $^{238}$Th against scintillometric readings (counts per second) in samples.

Figure 12: Plot of TiO$_2$ vs. Th.

Figure 9: Plot of PbO vs. Th.

Figure 13: Plot of Ce + La vs. Th.

Figure 10: Plot of U vs. Th.

Figure 14: Plot of FeO + MgO vs. Th.

Figure 11: Plot of Zr vs. Th.

Figure 15: Plot of PbO + Th vs. Gamma ray scintillometric reading.
A very strong positive correlation was established for the plot of Ce + La versus Th with r value of 0.97 and p value < 0.001. This implies that Ce + La contribute significantly to thorium activity. Ce and La are rare earth metals which are naturally radioactive and are found in monazite (Figure 13).

A strong positive correlation was established for the plot of w(FeO)/% + w(MgO)/% versus Th with r value of 0.905 and p value < 0.001 thus indicating that thorium concentration has a direct relationship with Mg0 and Fe0 (Figure 14). This implies that thorium concentration in rock will increase with increased Mg0 and Fe0 content of such rocks. This could be through cations substitution with elements like titanite, rutile, zircon due to similar ionic size.

Plot of PbO + Th versus gamma ray counts showed a strong correlation with a r = 0.76 and p < 0.0001 value. The strong positive correlation is due to high concentration of Pb in the rocks where the radioactive measurement was carried out. The Pb content of the pegmatite in the area is responsible for the radioactive level detected in it. (Figure 15).

The study of rock slides in thin sections under the petrographic microscope revealed the following petrographic features;

- For the different samples observed, zircon is the main radioactive mineral present in all the thin-sections. Another radioactive mineral such as xentomite though in small amounts is also present (Figure 16).
- Titanium rich phases such as rutile and ilmenite could be present which may be responsible for the high Ti content.
- For the pegmatites, the high Pb concentration is attributed to the presence of a lead mineral.
- The high radioactivity observed in some samples can be attributed to the high zircon content as well as the cluster of mafic minerals e.g. biotite largely because radioactive accessory minerals are commonly associated with mafic constituents.
- The major constituents of the rocks sampled are quartz, feldspar and minor amounts orthopyroxene and hornblende.

Conclusions and recommendations

This study has been able to infer that rocks of outcrops within the basement complex of south-western Nigeria exhibit high levels of radioactivity compared with the overlying materials except where radioactive materials have been added to the soil through other means. Spectrometric studies of some of the rock samples analysed revealed that the three primary radio-nuclides $^{238}$U, $^{232}$Th and $^{40}$K are not the only source of radioactive emissions. Plots of Ce + La + Th + Pb vs. gamma ray scintillometric reading as well as TiO$_2$ vs. Th gave nearly perfect correlations, thus implying that these elements are the main radioactive phases in the rock samples analysed. Only for the Lage-lu pegmatites the bulk radioactivity is attributed to its high lead concentration. Presence of zircon, xenotime, monazite, ilmenite, rutile and galena were observed in the thin sections and corroborated the geochemical evidences. Based on this study, it can be concluded that the distribution of primary radio-elements in areas where nuclear energy generation purposes are intended, particularly U and Th are concentrated in late stage magmatic fractions and in accessory minerals.

It is recommended that further studies such as isotope geochemical analyses as well as multi-step spectrometric analyses should be carried out to determine the actual concentration of the radiogenic nuclide to be able to distinguish between stable and unstable phases, determine the concentration of the daughter products and also to determine the presence of other radioactive elements present in the rock samples.
References


