A COMPARATIVE STUDY OF REE GEOCHEMISTRY IN PRECAMBRIAN PEGMATITES AND ASSOCIATED HOST ROCKS FROM WESTERN OBAN MASSIF, SE – NIGERIA

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ABSTRACT

The area of study (Igbofia-Akwa Ibami-Iwuru) is located between Latitudes 5°20′N and 5°27′N, and Longitudes 8°09′E and 8°18′E, south-eastern Nigeria. The pegmatites of this area have been studied by several authors for several decades till now. The present study evaluates the use of trace and rare-earth elements (REE) as finger prints in comparing the geochemistry of pegmatites and their associated host rocks, and to deduce if there is any co-genetic relationship between them. Sixteen representative samples of pegmatites and their host rocks (schist and granodiorites) were collected and analysed using the ICP-MS analytical method. The pegmatites were found to be richer than their host rocks in SiO$_2$, Al$_2$O$_3$, Na$_2$O+K$_2$O (alkali), Rb, Li, Cs, Be, Na, Zn, Nb, and Ga while they are poorer in Fe$_2$O$_3$, CaO, Sr, Ba, Zr, U, Cr, Th, and all the REEs. Only in the MgO contents are the rocks similar. Compared trace elements and REE data of schist and granodiorite (host rocks) against associated pegmatite samples in this work, show that the pegmatites are not necessarily products of their host rocks.

KEYWORDS: western Oban massif; rare-earth elements; geochemistry; Precambrian pegmatites; co-genetic;

1. INTRODUCTION

Pegmatites have generated much interest due to the important metals, which are of great industrial use to the Nigerian Basement and could be hosted by any of the crystalline basement rocks of gneisses, schists, and granodiorites.

The area of study (Igbofia-Akwa Ibami-Iwuru) is located between Latitudes 5°20′N and 5°27′N, and Longitudes 8°09′E and 8°18′E, in the western part of the Oban massif, south-eastern Nigeria (Fig. 1). The pegmatites of western Oban massif have been studied by several authors for over eight decades. One of the earliest workers Raeburn, (1927) observed that the pegmatites of Calabar area were characterized by the presence of tinstone and tourmaline. Ekwueme and Matheis, (1995) had earlier dated the rocks as Precambrian; Kingsley and Ekwueme, (2009); Oden et al., (2010) also evaluated the economic potentials of these pegmatites; Oden, (2010) investigated their structural pattern; while Igonor et al., (2010) described the geochemistry of the pegmatites and their host rocks. This study aims at evaluating and using trace and rare earth elements as “finger prints” and subsequently comparing the geochemistry of pegmatites and their associated host rocks, and to further deduce if there is any co-genetic relationship between the pegmatites and their hosts.

1.1 Sample and sampling

Representative pegmatite samples were collected within schist and granodiorite. Samples of the host rocks were also collected and analysed geochemically. Major, trace and rare-element geochemical analyses were carried out at the Activation Laboratory, Canada using the ICP-MS analytical method. Results of REE from geochemical analyses were chondrite normalised.

2. Geologic setting

The Oban massif has been affected by metamorphic events, tectonism, magmatism and metasomatism which also affected the geology of other areas of Nigeria crystalline basement. This is based on the observed rock types and their lithotectonic patterns (Rahman et.al., 1981, 1988; Ekwueme and Schlag, 1989; and Ekwueme, 1990d). Consequently, the Oban massif has been identified to be made up of three main lithologic units viz: (1) Migmatic and sheared gneissic rocks, schists, phyllites, meta-conglomerates and quartzites, amphibolites and meladiorites, deformed pegmatites and aplites, and pyroxenite – which forms the basement rocks as in other Basement Complex area of the country; (2) Older Granite intrusive series comprising meladiorites, granodiorites, adamellites to granitic rocks; pegmatites, aplite and quartz veins – which intrudes the first unit of rocks above; (3) Unmetamorphosed dolerite to microdioritic intrusives. Detailed systematic mapping of the study area revealed the occurrence of the following lithologic units (Fig.1):

- Metamorphic rock unit – which comprises the schists and gneisses forming the country rocks and also metamorphosed metadiorites;
- Igneous rock unit – which is subdivided into the

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major and minor intrusives. The major intrusives are the early stage granitoids made-up principally of the granodiorites, while the minor intrusives are the late stage dykes and veins made-up principally of the pegmatites and quartz veins, aplite and dolerite dykes.

3. Results and discussion

3.1 Petrology

The schists formed from progressive metamorphism of sediments or sedimentary rocks (Ekwueme and Onyeagocha, 1986), occur in the study area as medium grained, dark-brown to black rocks. These rocks are highly and strongly foliated. The outcrops are observable along stream channels and river beds around Iwuru, Ojor Nkonemba, and particularly Akwa-Ibami (Fig. 1). Xenoliths of the schist occur in the granodiorites. There are also layered relic and lensoid bodies. Generally the schists strike in the NE – SW direction and dip between 50° to 80° predominantly in the SW direction conforming to the dominant orientation of the Pan African basement rocks in Nigeria (Oden, 2010).

Petrographic analyses reveal the presence of micas (biotite and muscovite), almandine garnet with a relatively high amount of quartz (though variable, between 4% and 12%), hornblende and glaucophane. Other minerals present as accessories include cordierite, plagioclase, iron-ores and other opaque minerals. The granodiorites are non-foliated, and have a gradational contact relationship with the schist. Two varieties of the granodiorites were mapped - The porphyritic and non-porphyritic varieties. The porphyritic variety dominates the NE and SE portion of study area with phenocrysts of K-feldspar, while the non-porphyritic variety is found mainly on the western part. Petrographic analysis shows high percentage (about 23%) of euhedral to subhedral crystals of quartz intergrown with orthoclase and sometimes microcline. Large euhedral crystals of plagioclase (oligoclase - 25%) with lath-like shape, dominate the rock. Interlocking crystals of orthoclase occurs mainly as phenocrysts and is twinned according to the Carlsbad law. Other minerals present include equant grains of brown to greenish biotite, microcline showing cross-hatched twining, and prismatic crystals of hornblende. Zircon and iron-ore occur as accessory minerals in small amounts. Glaucophane, a typical metamorphic mineral occurs in the non-porphyritic variety. This is assumed to have been derived from the schist intruded during emplacement. In addition to the presence of xenoliths of schist in the granodiorite, the action of magmatic stoping, which has been suggested as means of emplacement for the granodiorite (Nganje and Ekwueme, 1996) is here supported.

Pegmatites occur ubiquitously in the area of study. They occur as either external or internal vein structures, which are sometimes unmappable and sometimes as mappable ridges. These pegmatites occur in high angle structures as over 94% have dip values greater or equal to 50° (Oden, 2010). These rocks are hosted by the two main rock units found in the study area, which are the metamorphic (schists and gneisses) and the igneous (granodiorite) rocks. Detailed description of these host rocks have been described by Ekwueme and Schlag, (1989); Nganje and Ekwueme,
have undergone a high degree of weathering. Ekwueme specimen. Those found within the metamorphic terrain have been given in Igonor, et al., (2010). Ekwueme and Matheis (1995) suggested that the pegmatites and Older Granites of southeast Nigeria are closely related, that in fact the pegmatites are derived from the cooling of the granodiorites and hence both rock types have a common origin (Ekwueme, 1990d).

3.2 Geochemistry

3.2.1 Major oxides

Details of the major oxide analyses of representative samples are given in Table 1. Table 2 shows the range and mean values of major oxides, trace and rare-earth elements in the samples. SiO$_2$ contents vary widely within the various rock types. It ranges from 50.95 to 76.64%wt in the schist; 54.22 to 60.50%wt in the granodiorites; and 53.05 to 73.01%wt in the pegmatites. The pegmatites have the highest average content SiO$_2$ (68.72%wt). Al$_2$O$_3$ ranges widely from 9.52 to 15.63%wt in the schists; marginally from 11.01 to 13.60%wt in the granodiorites; and from 12.00 to 16.89%wt in the pegmatites. Though all the rock types have similar average Al$_2$O$_3$ content (13.16%wt = schist; 12.75%wt = granodiorite; and 14.49%wt = pegmatites), the pegmatites have a slightly higher value than that of the schists and granodiorites. All the rocks have been shown to be strongly peraluminous (Oden et al., 2010). A wide range of values is observed in the Fe$_2$O$_3$ content of the rocks (schist = 3.89 to 17.60%wt; granodiorites = 8.04 to 19.70%wt; and pegmatites = 1.40 to 15.70%wt). The pegmatites have a mean Fe$_2$O$_3$ value of 4.66%wt, which is almost one third mean value of its host (schist = 12.35%wt and granodiorite = 12.44%wt), which both have similar values. A wide range of alkali (Na$_2$O + K$_2$O) concentration is observed in the pegmatites (0.79%wt to 11.19%wt), while the range is marginal in the host rocks (schist = 3.53 to 5.70%wt and granodiorites = 4.10 to 5.87%wt). The pegmatites have average alkali content (6.24%wt), which is slightly higher than the host rocks (schist = 4.94%wt and granodiorite = 5.41%wt). The richness of the rocks in alkali content is possibly due to high K-feldspar content. All the rocks have relatively low MgO content. Comparatively, the pegmatites have the lowest average MgO content of 1.24%wt while the schists have the highest average value of 1.95%wt. With regards to the CaO content, the pegmatites have the least average content (2.56%wt); while the granodiorites have the highest (10.70%wt – almost five times that of the pegmatites).

3.2.2 Trace elements

Details of the trace elements analyses of representative samples are given in Tables 2 and 3. A spidergram of all trace elements (Ba-Be from table 3) is plotted to show the differences and similarities in the trace elements of the host rocks (schists and granodiorites- Figs. 2a and 2b) and their associated pegmatite veins (Fig. 2c). Large Iron Lithophile Elements (LILE) like Rb, Ba, and Sr are abundant (Table 3) and registers a positive slope in the spidergram (Figs. 2a and 2b) of the schists and granodiorites respectively, but the opposite is noticed in fig 2c, where the pegmatites are depleted in the LILE. High Field Strength elements such as Nb and Zr are high in the pegmatites but low in the granodiorites and schists as shown by a positive slope for the pegmatites and negative slope for the schists and granodiorites (Figs. 2a, 2b and 2c). Be has a negative anomaly for the schists and granodiorites (Figs. 2a and 2b) while the pegmatites has a positive anomaly (Fig. 2c). Ni and V similarly has negative anomaly in the pegmatites but positive in the schists and granodiorites (Figs. 2(a, b and c)). Y has a conspicuously negative anomaly for all the rock types, while Zn has positive anomaly for all the rock types (Figs. 2a, 2b and 2c) same as Ga (though the rocks cannot be said to be enriched in these elements).

All the rock types are enriched in Li when compared to the average concentration values of granodiorites (25ppm) and granites (30ppm) (Taylor, 1965). The pegmatites have a much higher average Rb value (344.70ppm) than their host rocks. All the pegmatites have extremely low values of Sr (mean value is 27.50ppm), which is not comparable to any ‘standard’ analyses geological material. The granodiorites are poorer in Sr (mean value =331.75ppm) compared to the expected average in world granodiorites (450ppm - Taylor, 1965). All the rocks from study area are enriched in Cs (15-41ppm). Though these rocks have higher values of Cs than those of similar average rocks, they do not show values anywhere close to that found in the late-stage Goldolphin granite from Cornwall (260ppm – Bowler, 1959), or that of Yugoslav granites which had value as high as 115ppm (Deleon and Ahrens, 1957). The Sr values for pegmatites (3.40 - 38.70ppm) and schists (56.2 - 121.8ppm) vary widely, while the Zr values in the granodiorites (52.30 - 97.10ppm) range narrowly. The rocks all have very low average concentration value of U (2.76ppm for schist; 5.6ppm for granodiorite; and 5.38ppm for the pegmatites). Normal acid rocks are not expected to have above 20ppm concentration of Cr (Swaine, 1955) but the acid rocks from study area (granodiorites and pegmatites) have far greater average values of 165.25ppm and 39.25ppm respectively. The pegmatites have very low average Th value (2.55ppm), which is equal to that of ‘normal’ basalts (2.2ppm – Taylor, 1965).

Comparing the trace element data of the pegmatites of study area to their host, the pegmatites are richer than their host only in Rb, Cs, and Be while in other trace elements discussed, they are more deficient. And also the spidergrams (figs 2a, 2b and 2c) patterns
observed in the granodiorites and schists are almost similar but vary from that of the associated pegmatites. To a reasonable extent, these do not reflect any genetic relationship between the pegmatites and their host.

Fig. 2a: Spidergram illustrating how the trace elements vary in the different schist sample from study area. (see Table 3 for legend explanation).

Fig. 2b: Spidergram illustrating how the trace elements vary in the different granodiorite sample from study area. (see Table 3 for legend explanation).
Fig. 2c: Spidergram illustrating how the trace elements vary in all the pegmatite samples from study area. (See Table 3 for legend explanation).

### 3.2.3 Rare-earth elements (REE)

Details of the rare-earth elements analyses of representative samples are given in Table 3, while Table 4 shows the normalised REE and REE ratios.

The schists and granodiorites have more abundance of REE than the associated pegmatite veins (Table 3). The REE abundance decreases from granodiorite-schist-pegmatites. Light REE (LREE) (La, Ce) concentrations vary narrowly in the schists but widely in the granodiorites and pegmatites while Heavy REE (HREE) (Lu, Yb) varies widely in the schists and granodiorites but narrowly in the pegmatites. However, the schists and granodiorites are slightly more enriched in LREE than the pegmatites as average values are 19.95ppm, 14.54ppm, and 11.46ppm for the schist, granodiorite and pegmatites respectively. Eu anomaly values range narrowly (almost negligibly) in the schist (0.6-0.8) and granodiorite (0.7-0.8) but widely in the pegmatites (0.2-1.2). (La/Yb)\textsubscript{N} ratio varies widely for all rock types-it is generally flat in the pegmatites; while increasing in the schists (3-38) and granodiorites (2-30). (La/Sm)\textsubscript{N} varies marginally in the schist and only narrowly in the pegmatites and granodiorite with both rock types having similar range of values (Table 4). But in most cases, the value is greater than one (1). All the rock types exhibit well pronounced negative Eu anomaly and fractionated LREE pattern (Fig. 4).

### 4. Interpretations

#### 4.1 Major and trace elements:

A spidergram (Fig 3a) shows the relative abundance of some major and trace elements in the pegmatites and their host rocks. In Fig. 3b, a ‘depletion and enrichment’ bar is included into fig. 3a with the pegmatite as the relative reference point. These diagrams (Figs. 3a and 3b) both show that the pegmatites are more enriched than their associated rocks in Na, Zn, Rb, Nb, and also in Ga. In all other compared elements both the schist and granodiorites have higher average contents than the pegmatites. The pegmatites are the most depleted in Sc, V, Mg, Co, Ni, and Y. All the rocks are rich in Zn (average values are schist - 320ppm; granodiorites - 2082ppm; and pegmatites - 2613ppm), though not up to the required workable cut-off grade for granitic rocks (2%wt – Umeshwar, 2000). Some pegmatites in Iwuru and Akwaibami (T9, T13, and T15) with values of 17ppm; 36ppm; and 82ppm respectively, are more enriched in Sn than other rock types. These samples are most likely to be tin bearing as tin bearing pegmatites usually contain between 15 – 30ppm of Sn (Barsukov, 1957). The Ni value in sample T7 (151ppm) is above the limit for intermediate/acid rocks (100ppm – Taylor, 1965). This could have been caused most likely by crustal materials contaminated by meteorite impact or that the rock is of basic origin. Though extreme enrichment in Li is expected in pegmatites (Taylor, 1965), this is not the case with the pegmatites of the study area (average Li value is 40ppm). This is most probably due to lack of Li minerals like lepidolite, spodumene, and petalite, which are the major minerals of lithium. Extreme enrichment of Li in acid rocks (< 100ppm) is a useful indicator of extreme fractionation and also may imply that the rocks sampled are late stage high level products. Of all the rocks analysed, only samples (T13- pegmatite, T1 and T2- schist) have greater than 100ppm to suggest extreme fractionation. But for the schists, this may probably reflect that their progenitors are late stage level products which have undergone extreme fractionation.
A common anomaly found in pegmatites is low K/Rb ratio (less than 100ppm), which indicates Rb enrichment (Heier and Taylor, 1959b). But the pegmatite of the study area shows the reverse, that is, a very high K/Rb ratio, which invarably indicates Rb deficiencies and K enrichment. The following are the probable reasons for the depletion of Rb in the samples: (1) since Rb enters into the rocks only after Ba\(^{2+}\) and K\(^+\), the abundance of Ba in the samples and presence of K-feldspar especially in the granodiorite leads to depletion in Rb; (2) also the presence of biotite especially in the schists and pegmatites which are high K\(^+\) minerals; (3) similarly high K\(_2\)O content obtained in the rocks; and (4) accumulation of early feldspars themselves depleted in Rb all leads to depletion of Rb in a rock (Upton, 1960). The pegmatites hosted by granodiorites are far richer in Be concentration than those hosted by schists. Since micas and hornblende accept Be more readily, this high content of Be in these rocks is accounted for by the high content of micas and hornblende in the rocks (Igonor et al., 2010). Furthermore, the samples from Igbofia and Akwa-Ibami are most likely to host Be minerals in the Oban Massif. The schist’s average concentration of Zr is similarly lower than average concentration values in known and analysed geologic materials (Taylor, 1965) hence rendering comparism for
progenitor inference in-effective; though Taylor, (1965) posits that Zr content of several hundred ppm could indicate a sedimentary origin. With that in mind, it is possible to infer that the schist (T1 and T2) is of sedimentary origin. The above 100ppm value of Cr for schist sample (T4) is an indication of derivation of the schist from an igneous progenitor, while that of the granodiorite is likely to be controlled by a Cr-rich phase or Cr-rich orthopyroxene. And it also indicates derivation from or near mantle magma (Faust et al., 1956). The Th concentration does not pose any health risk in the granodiorites of study area as the values obtained are within the range found in ‘normal’ acid rocks and hence can be used as construction materials. It is probable that the anomalous concentration of Ce in the rock from study area is caused by weathering and or burial metamorphism (Freyer, 1977). The high Cu and Ni content may indicate the earlier presence of primary sulphides and existence of a Ni-rich phase (presumably olivine). The samples all show the chemical traits of magmas that have experienced a high degree of crystal fractionation of ferromagnesian minerals (Radhakrishna and Joseph, 1995).

4.2 Rare-earth elements (REE):

Spidergrams showing the variation of average chondrite-normalised REE in all the rock samples (Fig. 4a) and in various rocks sample (Figs. 4b, 4c, 4d and 4e) is plotted. These show an almost flat pattern and progressive decline both in the LREE and HREE, with a negligible negative Eu anomaly in the schists and granodiorites. But the Eu anomaly is more pronounced in the pegmatites. The shape arrangement of the REE spectra as shown by the pegmatite and host rocks of the study area could be interpreted as an indication of systematic and progressive depletion of REE. This is also indicated by the steady increase of the ratio LREE/HREE (La/Yb) in all the rocks. This ratio increase, which also means the depletion of HREE, is most likely connected with the increase of SiO₂ and decrease of Zr (Stable et al., 1987) as well exemplified by the granodiorite and pegmatites of the study area, but weakly defined in the schists (Fig. 5). According to Henderson, (1984), this depletion in HREE may be due to dissolution of zircon during metamorphism (metasomatism in the case of the granodiorite and pegmatites) since zircon is a very important phase for concentrating the HREE. The strong enrichment in LREE is interpreted as resulting from a combination of factors: the nature of the source; possible crustal contamination; and metamorphic re-distribution (Gill and Bridgewater, 1979). Minerals containing Sr, Ba, and K tend to concentrate the large ions of the “cerium group” (LREE) those containing Sc, Zr, etc concentrate the “yttrium group” (HREE) (Semenov, 1958). Enrichment in LREE in the samples indicates the absence of minerals like monazite and allanite from these rocks as their presence even in small amounts (1%) could greatly cause depletion in the LREE (Taylor, 1965).

![Figure 4a](image-url)  
Figure 4a: A spidergram illustrating the variation of Chondrite normalised average REE concentration plotted against atomic numbers for all rocks from western Oban Massif.
Figure 4b: A spidergram illustrating the variation of Chondrite normalised REE concentration plotted against atomic numbers for all pegmatite samples hosted by granodiorites in western Oban Massif. (See Table 3 for legend explanation)

Figure 4c: A spidergram illustrating the variation of Chondrite normalised REE values plotted against atomic numbers for all granodiorite samples from western Oban Massif. (See Table 3 for legend explanation)
Figure 4d: A spidergram illustrating the variation of Chondrite normalised REE concentration plotted against atomic numbers for all pegmatite samples hosted by schists in western Oban Massif. (See Table 3 for legend explanation)

Figure 4e: A spidergram illustrating the variation of Chondrite normalised REE values plotted against atomic numbers for all schist samples from western Oban Massif. (See Table 3 for legend explanation)

Figure 5: Plot of (La/Yb)$_N$ against SiO$_2$ showing the systematic increase of (La/Yb)$_N$ with increase in SiO$_2$

The plot (La/Yb)$_N$ vs Ce$_N$ (Fig. 6) and (La/Yb)$_N$ vs Yb$_N$ (Fig. 7) which is a measure of the degree of REE fractionation with changing REE content both shows that on the whole, the granodiorite have experienced the highest degree of REE fractionation followed by the pegmatites while the schist are the least fractionated.
The plot of (Gd/Yb)\textsubscript{N} vs Yb\textsubscript{N} (Fig. 8) is a measure of the degree of HREE fractionation. This plot indicates that the schist from study area have experienced a high degree of HREE fractionation while the degree of HREE fractionation is much lower in the pegmatites. The granodiorites have the lowest degree of HREE fractionation. The degree of LREE fractionation in the rock samples from the study area is as shown in the plot (La/Sm)\textsubscript{N} vs Sm\textsubscript{N} (Fig. 9). The plot indicates the enrichment of LREE relative to the MREE Sm\textsubscript{N} in all the samples analysed. All the rocks have Eu/Eu* ratio less than 1.0 except sample T13. This ratio is a measure of the europium anomaly and a value greater than 1.0 indicates a positive anomaly while a value less than 1.0 is a negative anomaly (Taylor, 1965). Thus except for pegmatite sample(T13) with a positive europium anomaly, all the other rock samples show a negative europium anomaly as they all have values less than 1.0. Europium anomalies are mainly controlled by feldspars, especially in felsic magmas. So that removal of feldspar from a felsic melt by the process of crystal fractionation or during the partial melting of a rock in which feldspar is retained in the source, will give rise to a negative Eu anomaly in the melt. Minerals like hornblende, sphene, clinopyroxene, orthopyroxene and garnet may also affect (to a lesser extent) the Eu anomaly but by giving rise to a positive anomaly. Possible fractional crystallization of orthopyroxene and clinopyroxene in place of olivine might have resulted in the ratio (La/Sm)\textsubscript{N} been greater than one for the rocks of study area (Jahn and Sun, 1979). It seems metamorphism and fractionation has had an effect on the mobility of elements considering the wide variation in La/Yb and Ce/Sm value in the schist and granodiorite.

Figure 6: Plot of (La/Yb)\textsubscript{N} against Ce\textsubscript{(N)} measuring the degree of REE fractionation with changing REE content.

Figure 7: Plot of (La/Yb)\textsubscript{N} against Yb\textsubscript{(N)} measuring the degree of REE fractionation with changing REE content.
Summary and Conclusions

In summary, the pegmatites are on the average richer than their host rocks with respect to SiO$_2$, Al$_2$O$_3$ and alkalis, while they are poorer in Fe$_2$O$_3$ and CaO. Only in MgO content are the rocks similar. They are also more enriched than their host in Rb, Li, Cs, Be, Na, Zn, Nb, and Ga but deficient in Sr, Ba, Zr, U, Cr, Th, and all the REE considered.

The occurrence of pegmatites within schist and granodiorites in the Oban Massif has led to suggestions that the pegmatites and associated host rocks have some petrogenetic relationships (Ekwueme, 1990d, Ekwueme and Matheis, 1995). Since REE are believed to be rarely affected in their distribution in rocks by the processes of either metamorphism or fractionation, they are essential in determining petrogenesis of rocks. So that if the pegmatites of the study area are products of their host rocks in any way, they ought to bear similar chemical attributes (especially trace and REE) linking them together. However the trace and rare-earth element analyses from this work do not show any chemical similarities. Even though this work is preliminary, the results reveal that the pegmatites are not necessarily products of their host rocks. For further studies, any genetic model for the pegmatites needs to answer if there are any chemical attributes of the pegmatites consistent with fractional crystallization and or partial melting of the granodiorites and schist respectively.

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