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Rare Earth Element Geochemistry and Abundances in Syenites and Charnockitic Rocks of Selected Locations within Southwestern Nigeria

Geokemija prvin redkih zemelj in njihova vsebnost v sienitih in čarnokitnih kamninah z izbranih lokacij v jugozahodni Nigeriji

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Abstract in English

Rare earth elements (REE) are critical metals widely used in modern technology. There is an increased demand for these elements which has necessitated additional exploration. The REE geochemistry of syenites and charnockitic rocks from selected locations within southwestern Nigeria were studied to determine their concentrations, distribution patterns, and mineralisation potential to deduce if they can be economically exploited. Mineralogical studies were carried out using scanning electron microscopy (SEM). Ten rock samples including five syenites and five charnockites were analysed for their elemental composition using inductively coupled plasma – mass spectrometry (ICP-MS). The REE-bearing minerals identified from the SEM analysis include apatite and monazite. The fractionation ratio $(La/Yb)_N$ ranged from 29.36 to 48.75 for the syenites and from 24.43 to 48.29 for the charnockites, indicating magmatic differentiation. Total REE (∑REE) in the syenites (342–675 ppm) and charnockites (220–802 ppm) suggests that they are enriched with respect to REE but the REE concentration in the rocks of the study area had very low ∑REE compared to the REE contents of the rocks where they have been mined and may therefore not be economically viable.

Keywords: rare earth elements, syenites, charnockites, abundances, fractionation

Povzetek

Prvine redkih zemelj (REE) so kritične kovine, široko uporabljane v moderni tehnologiji. Povečano povpraševanje po teh prvinah vodi v dodatna raziskovanja. V izbranih lokacijah jugozahodne Nigerije smo v sienitih in čarnokitnih kamninah določili vsebnosti REE, porazdelitvene vzorce in mineralizacijski potencial, da bi ugotovili upravičenost ekonomskega izkoriščanja. Mineraloške raziskave so bile opravljene z vrstičnim elektronskim mikroskopom (SEM). Elementna sestava desetih vzorcev, od tega pet sienitov in pet čarnokitov, je bila določena z induktivno sklopljeno plazemsko masno spektrometrijo (ICP-MS). S SEM smo kot nosilca REE prepoznali apatit in monacit. Frakcionacijsko razmerje $(La/Yb)_N$ ima za sienite razpon od 29,36 do 48,75, za čarnokite 24,43–48,29, kar kaže na magmatsko diferenciacijo. Skupna vsebnost REE (∑REE) v sienitih (342–675 ppm) in čarnokitih (220–802 ppm) nakazuje na obogatitev REE, so pa vsebnosti REE v raziskanih vzorcih nižje od tistih, ki jih izkazujejo kamnine iz trenutno rentabilnih rudnikov, zato ekonomsko izkoriščanje preiskanih sienitov in čarnokitov ni upravičeno.

Ključne besede: elementi redkih zemelj, sieniti, čarnokiti, vsebnost, frakcionacija

Introduction

Rare earth elements (REE) have attracted much attention in recent years due to their high significance to the entire human population. Their importance in the production of modern technological applications cannot be underestimated, and their global reserve, which is gradually diminishing, has led to an increased demand for these elements. As the global demand for REE increases, the exploration of new REE deposits is essential [1].

Economically, the REE mineralisation and deposits include two separate types: light rare earth element (LREE) deposits and heavy rare earth element (HREE) deposits. Most LREE are produced from carbonatite deposits. Ion-adsorption clay deposits in southern China (referred to as "south China clays") are currently the world's main source of HREE, they have low REE content, but they are economical because the REE can be easily extracted from them. The second significant HREE sources are alkaline rock-hosted deposits containing HREE and Y as their primary product or co-product. The deposits containing HREE generally tend to be lower grade than the LREE deposits. However, the HREE, based on unit value, can be more valuable and their low-grade deposits may still be economically exploitable [2]. LREE are generally more abundant, and less valuable than the HREE.

Many studies have been carried out in various areas to check for possible REE potential and recent studies have shown possible potential for REE mineralisation in areas that might have been initially overlooked. Because of the need for more exploration and exploitation of these REEs, there is an awakening call for more studies to be carried out in other areas and in rocks that have not been explored [3].

The focus of this study is on determining the abundances of REEs in syenites and charnockitic rocks of selected areas in southwestern Nigeria, in order to determine their mineralisation potential. The study area is underlain by the basement complex of Nigeria. Rahaman [4] reviewed the geology of southwestern Nigeria

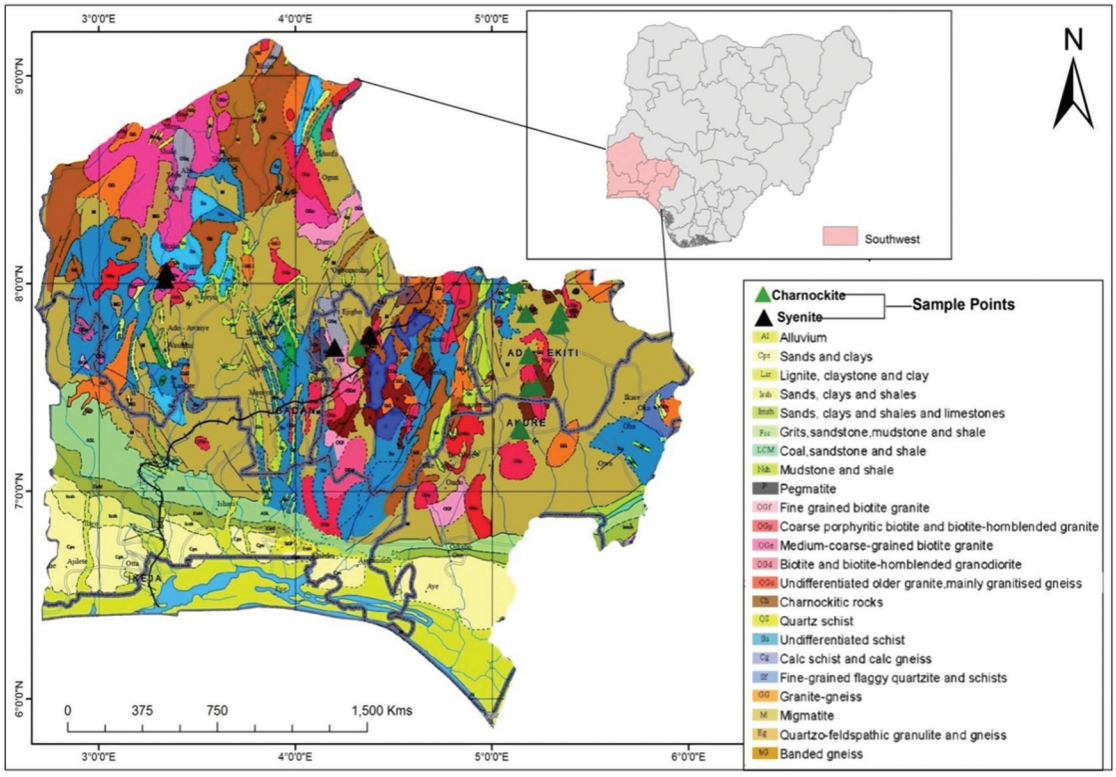

Figure 1: *Geological map of southwestern Nigeria showing the sampling points. (Modified after the Nigerian Geological Survey Agency [5]).*

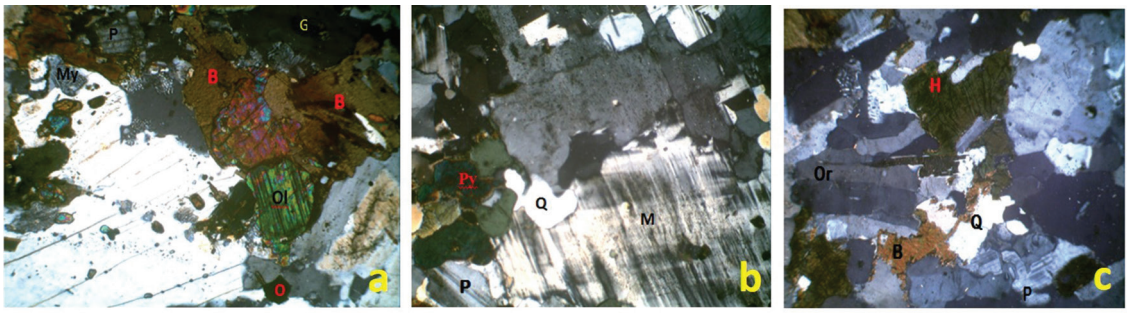

Figure 2: *Photomicrograph of syenites from: (a) Okeho showing B = biotite, P = plagioclase, Ol = Oolivine, My = mymerkite, O = opaque, G = garnet (b) Iwo showing Q = qQuartz, M = microcline, P = plagioclase, Py = pyroxene (c) Kajola showing P = plagioclase, B = biotite, Or = orthoclase, H = hornblende.*

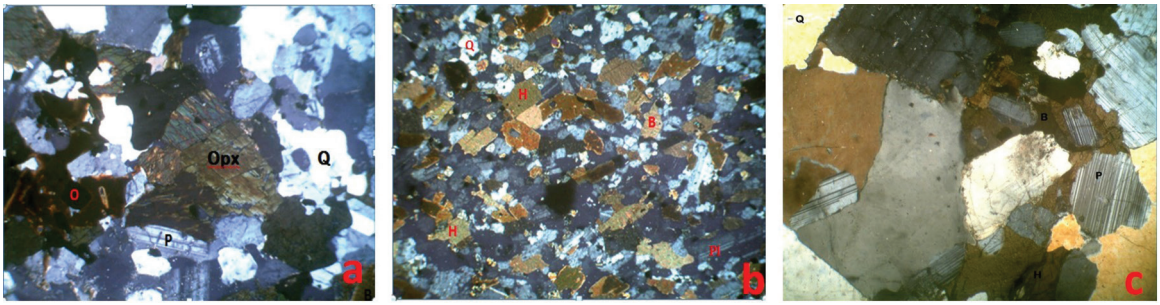

Figure 3: *Photomicrographs of charnockite from a) Akure showing Q = quartz, B = biotite, Opx = orthopyroxene, P = plagioclase, O = opaque; b) Ikere showing Q = quartz, B = biotite, Pl = plagioclase, H = Hornblendeh; and c) Ado showing Q = quartz, B = biotite, P = plagioclase, H = hornblende.*

and recognised six major lithological units, which include (1) the migmatite-gneiss-quartzite complex; (2) the paraschists and metaigneous rocks that consist of pelitic schist, amphibolite, and talcose rocks; (3) metaconglomerates, marbles, and calc-silicate rocks; (4) the charnockitic rocks, (5) the older granites varying in composition from granodiorite to true granites; and (6) potassic syenites and the dolerites. Fifteen different locations around southwestern Nigeria were chosen for the study (Figure 1). The mapped lithological units for these studies are syenites and charnockitic rocks.

Materials and methods

Fresh samples of syenites and charnockitic rocks were obtained during systematic field mapping. Petrographic analysis was carried out at the University of Ibadan. Scanning Electron Microscopy-Energy Dispersive Spectroscopy (SEM-EDS) was used to study possible REE-bearing minerals at the Department of Geology, University of the Free State, South Africa. Ten representative rock samples inclusive of five syenites and five charnockites were analysed for their elemental concentration at the Bureau Veritas Mineral Services, Vancouver, Canada using an inductively coupled plasma-mass spectrometer (ICP-MS).

Results and discussion

Field occurrences and petrographic descriptions

Syenites: The syenites studied were found in the following areas: Awo, Iwo, Okeho, Ayetoro-Oke, and Iwere-Oke. The syenite of Iwo is found as a low-lying and very extensive outcrop; quartzo-feldspathic and pegmatitic veins were observed in the outcrop. It is grey in colour with fine- to medium-grained texture. The Okeho, Ayetoro Oke, and Iwere Oke syenite outcrops were found as low-lying and huge

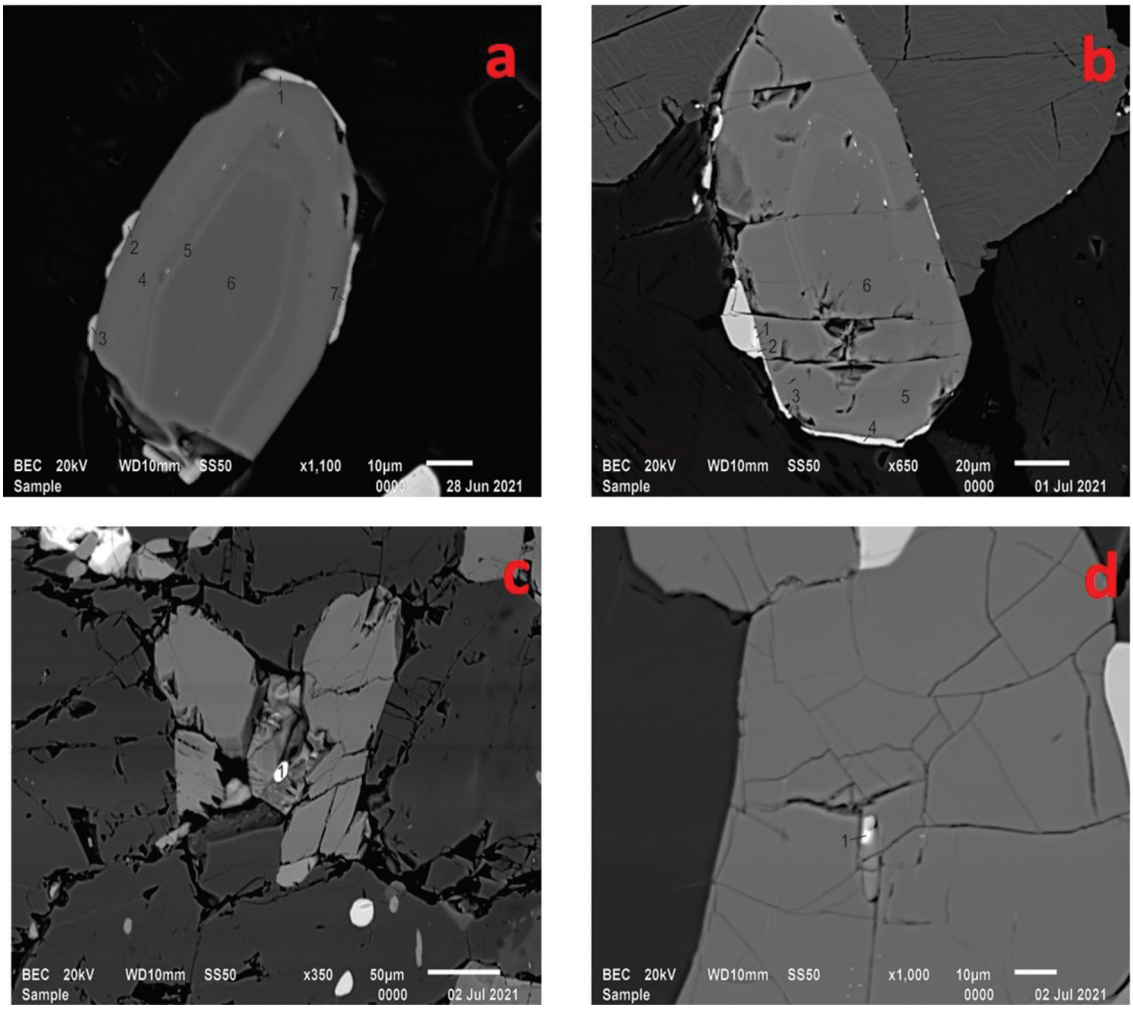

Figure 4: *(a and b): Back-scattered electron (BSE) image of REE-bearing minerals in the rims of zoned apatite from charnockites; (c and d): BSE image of REE-bearing minerals in syenites.*

continuous boulders. They are dark green to dark grey in colour with medium- to coarse-grained texture. The petrographic analysis for syenites reveals mineral assemblages of quartz, plagioclase, biotite, olivine, pyroxenes, hornblende, microcline, augites, and opaque minerals (Figure 2a–c).

Charnockites: The charnockite samples were collected from: Oye, Otun, Ido Osi, Ado, Ikere Ekiti, Akure, and Osuntedo. The charnockitic rocks of Oye occur as low-lying outcrops, boulders, and as road cut in some places. The charnockites in Ado-Ekiti, Ikere, and Akure occur mostly as hills and as large, elongated, and rounded boulders in close association with the porphyritic Older Granite. They are coarse grained with a colour variety ranging from dark green to greenish grey and whitish grey as observed in the sample from Ado-Ekiti. The petrographic analysis revealed mineral assemblages of quartz, plagioclase, orthoclase, biotite, pyroxenes, hornblende, microcline, and opaque minerals (Figure 3a–c).

REE minerals

REE-bearing accessory minerals were studied using their chemical compositions as observed via Scanning Electron Microscopy. The observed REE in the charnockite occurred along the rims of zoned apatite (Ca_5 $(PO_4)_3$ (F,

Cl, OH) grain (Figure 4a and b). The zoning might be due to a change in temperature or composition of the fluid from which the crystal crystalised.

Apatite was observed to be associated with a mineral suspected to be monazite, which is an REE-concentrating mineral more enriched in LREE. REE minerals in syenites are small and scarce (Figure 4 c and d). Their composition is also very different to the REE minerals in the other samples. The REE-bearing mineral identified in the BSE image of the syenite is suspected to be monazite, on the basis based of its chemical compositions (Table 1).

REE geochemistry

The total REE (∑REE) ranges from 342.54 to 675.15 ppm. The syenites are more enriched in LREE, with values ranging from 325.66 to 648.24 ppm, and Ce, being the most prevalent REE, HREE ranged 19.03-26.91 ppm. The Iwo quartz potassic syenite is characterized by higher ∑REE than the syenites from other areas, with a value of 675.15 ppm. The Kajola and Iwere–Oke syenites have comparable values of 469.17 ppm and 462.69 ppm, respectively, for their ∑REE, while the syenites from Ayetoro–Oke have the lowest ∑REE value of 325.66 ppm (Table 2).

The ∑REE of the charnockitic rocks varies with location, ranging from 224.26 ppm in the charnockite of Otun to 801.03 ppm in Akure charnockite. The charnockites are more enriched in LREE with values ranging from 211.28 ppm to 754.03 ppm and relatively depleted in HREE with values ranging from 8.64 ppm to 47.7 ppm. The Akure charnockite is characterised by anomalously high REE contents of 801.03 ppm compared to other charnockites. The syenites and charnockites are characterised by high concentration of LREE and relatively low contents of HREEs with significant LREE/HREE fractionation.

The rocks having more LREE than HREE may be a result of newly formed P-bearing minerals such as apatite or monazite being important carriers of LREE [6, 7]. That the syenites and charnockitic rocks are characterised by high REE concentrations, which is due to the presence of high values of LREE, suggests REE-concentrating minerals (especially monazite, which contains LREE), which indicates that their original magma had crustal input [8].

Table 1: *Chemical composition of the REE minerals in charnockites and syenites.*

	1	2	3	4	5	6	7	8	9
F				2.41	2.57	2.41			
Na_2O			1.88				3.6		
MgO	0.4	0.52	0.61				0.37		0.86
Al_2O_3	13.46	13.17	13.3				15.74	1.7	0.49
SiO_2	28.39	27.91	29.48	0.75	0.63	0.16	38.14	3.78	4.94
P_2O_5	5.25	5.85	7	39.4	39.7	40.57	1.87	32.45	25.65
Cl				0.12	0.17	0.12			
CaO	18.06	20.12	19.04	57.32	56.94	56.73	12.86	10.9	8.92
Fe_2O_3	14.22	13.76	12.72				11.85	0.32	12.6
La_2O_3	5.88	4.91	4.83				4.36	16.99	13.36
Ce_2O_3	11.01	10.54	8.52				8.66	28.13	25.92
Nd_2O_3	3.33	3.21	2.62				2.55	5.38	7.25
Total	100	99.99	100	100	100.01	99.99	100		
Mineral	Monazite	Monazite	Monazite	Apatite	Apatite	Apatite	Monazite		

1–7, REE minerals in charnockites, 8–9 REE in syenites.

Table 2: *Trace and rare elemental concentration in syenites and charnockites in selected locations in southwestern Nigeria (all values in ppm)*

	1	2	3	4	5	6	7	8	9	10
Ba	2495	1565	1493	1754	1429	1942	1915	1467	1144	2018
Ga	20.3	19.8	20.1	18.4	20	18.7	19	19.8	19	22.6
Hf	24.5	15.7	11.6	10.2	11.6	5.6	5.6	7.4	12.1	20.6
Nb	31.3	30.4	27.3	22.9	27.9	13	12.9	12.2	24.7	39.3
Rb	384	366.9	263.5	315.7	296	93.4	86	69.8	78.6	105.4
Sr	842	438.1	463.9	455.1	464.1	1062	1028	535.7	465.6	399.3
Th	79.6	60.7	30.8	36.1	45.7	7.6	6	16.6	9	16.4
U	5.9	4.8	4.9	3.9	3.4	1.5	1.1	2.2	1.6	0.8
V	90	60	76	51	64	88	85	59	88	116
W	0.7	1.8	2.2	1.7	1.1	0.9	0.7	<0.5	<0.5	0.8
Zr	1063	597	461	3867	441.6	223.6	220	301.5	531.3	895
Y	28	24.9	26.8	22	32.2	17	15.9	11	26.1	64.1
La	141	111.4	93.6	81.7	106.7	51.4	50.8	70.2	72.1	195.6
Ce	290	212.4	178.5	150.2	200.7	96.1	93.3	109.6	137.1	338.4
Pr	39	24.35	23.08	18.32	25.87	11.97	11.66	11.81	17.45	41.96
Nd	149	81.1	83.6	63.5	93.4	42.9	42.7	37.7	62.6	150.7
Sm	24	12.04	13.54	9.75	15.15	6.77	6.64	4.81	10	23.06
Eu	4.58	2.37	2.57	2.19	2.67	2.12	2.11	2.49	2.86	4.31
Gd	14	7.73	8.96	6.85	10.29	5.1	4.82	3.46	7.94	18.72
Tb	1.44	0.97	1.11	0.83	1.24	0.64	0.6	0.42	1.03	2.39
Dy	6.02	4.72	5.31	4.2	6.02	3.2	3.19	2.05	5.07	12.51
Ho	0.88	0.83	0.94	0.73	1.06	0.6	0.56	0.37	0.93	2.23
Er	2.21	2.11	2.4	1.89	2.85	1.63	1.54	1.08	2.49	5.73
Tm	0.32	0.32	0.33	0.28	0.41	0.22	0.22	0.14	0.33	0.77
Yb	1.95	2.03	2.12	1.82	2.45	1.4	1.35	0.98	1.99	4.64
Lu	0.28	0.32	0.31	0.28	0.36	0.21	0.2	0.14	0.31	0.71
Y	28	24.9	26.8	22	32.2	17	15.9	11	26.1	64.1
ΣLREE	648	443.66	394.89	325.66	444.49	211	207	236	302.11	754.03
ΣHREE	26	19.03	21.48	16.88	24.68	13	12.48	8.64	20.09	47.7
ΣREE	675	462.69	416.37	342.54	469.17	224	220	245	322	802
ΣLREE/ ΣHREE	24	23.31	18.38	19.29	18.01	16.26	16.6	27.39	15.1	18.81
ΣREE+Y	703	487.59	443.17	364.54	501.37	241.26	235.59	256.25	348.3	865.83
Eu/Eu*	0.77	0.75	0.71	0.82	0.65	1.1	1.14	1.87	0.98	0.63
Ce/Ce*	0.94	0.98	0.92	0.93	0.92	0.93	0.92	0.92	0.93	0.9
$(La/Yb)_N$	48	37	29.77	30.26	29.36	24.75	25.37	48.29	24.43	28.42
$(La/Sm)_N$	3.66	5.82	4.35	5.27	4.43	4.78	4.81	9.18	4.54	5.34
$(Ce/Yb)_N$	38	27.06	21.78	21.35	21.19	17.76	17.88	28.93	17.82	18.86

Syenites: 1-1wo, 2-Iwere-Ile, 3 - Okeho, 4 - Ayetoro Oke, 5 - Kajola.

Charnockites: 6 - Otun. 7, Ado, 8 – Afao, 9 - Kajola, 10-Akure

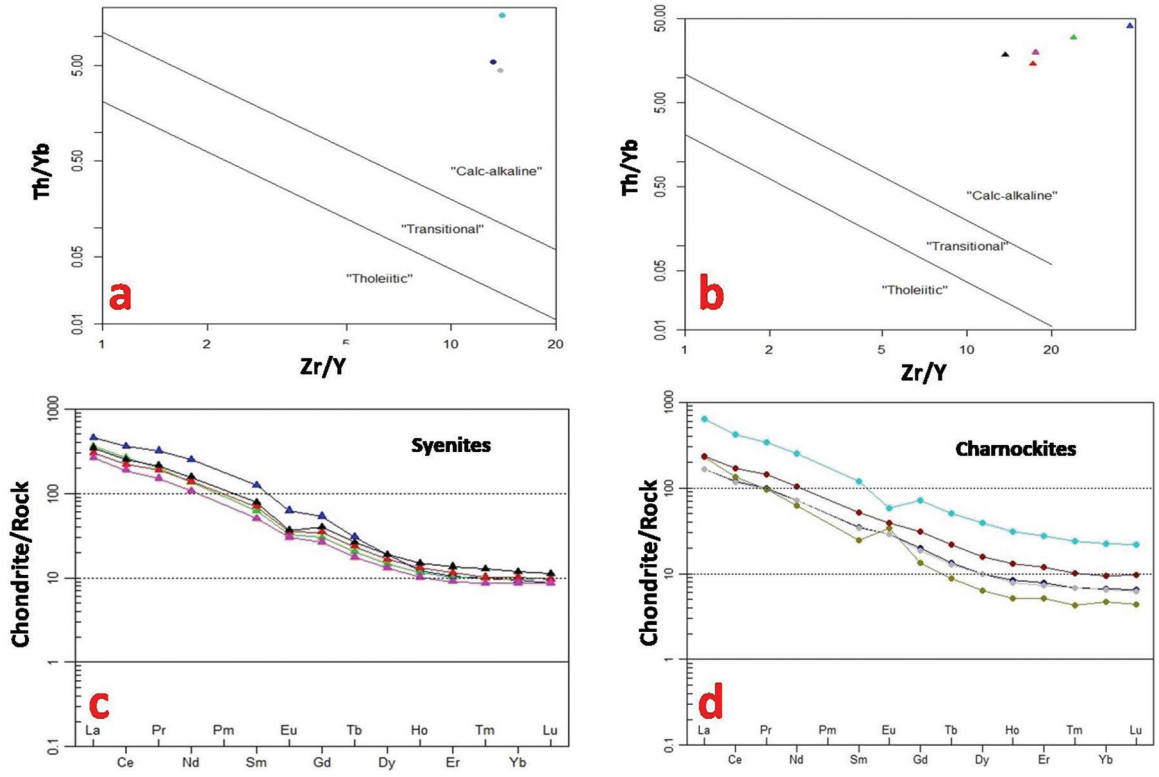

Figure 5: *a) Plots of Zr/Y vs Th/Yb for charnockites of the study areas showing three sampling points (after Ross and Bédard [14]; b) Plots of Zr/Y vs Th/Yb for syenites of the study area (after Ross and Bédard [14]); c) Chondrite normalised REE distribution pattern for the syenites (after Boynton [13]); d) Chondrite normalised REE distribution pattern for the charnockites (after Boynton [13]).*

The $(La/Yb)_N$ values ranged from 29.36 to 48.75 for the syenites and from 24.43 to 48.29 for the charnockites; the $(La/Sm)_N$ values in syenites is 4.35 : 5.82; and 4.54 : 9.18 in the charnockites. The $(Ce/Yb)_N$ ranges from 21.19 to 38.49 for the syenites, and from 17.76 to 28.93 for the charnockites. The high values of the normalised ratios of La to Yb, La to Sm, and Ce to Yb in the different rock types are evidence of a high degree of fractionation, which shows that the REE patterns are LREE enriched.

According to Ukaegbu and Ekwueme [9], the La/Yb, Ce/Yb, and La/Sm values are normally higher in residual products than in primary melts. Hence, $(La/Yb)_N$ values higher than 5 are an indication of magmatic differentiation [10]. This implies that even though the magma might have originated from a mixed source, differentiation was important in the formation of these rocks [8].

The syenites showed negative Eu anomalies of 0.65–0.82, suggesting that a large amount of plagioclase was removed from felsic magma during fractional crystallisation. It could also be due to the presence of pyroxenes in them, as pyroxene-rich cumulates acquire a negative Eu anomaly [11]. The charnockitic rocks showed both a negative and a positive Eu anomaly of 0.63–1.87, indicating accumulation of plagioclase during the fractionation of magma for the later (Figure 5). According to Weill and Drake [12], magma crystallising stable plagioclase has most of the Eu being incorporated into the plagioclase mineral leading to a higher concentration of Eu in the mineral relative to other rare earth elements in the magma.

The Ce anomaly ranges from 0.92 to 0.98 in the syenites, and from 0.90 to 0.93 in the charnockitic rocks. Ce anomalies reflect the oxygen fugacity in the environment of formation, [13]. According to Boynton [13], when there is no large negative Ce anomaly, it indicates formation under reducing conditions; a large negative Ce anomaly indicates formation under oxidising conditions.

Table 3: *Comparison of the REE concentration in the syenites of the study area with other related rock types (all values in ppm).*

	1	2	3	4	5	6
REE	RANGE	AVERAGE	AVERAGE	Mean	RANGE	AVERAGE
La	81.72–111.4	257	93.36	42.07	5082–94810	449.5
Ce	150.2–290.2	450	214.36	60.57	7021–90630	798
Pr	18.32–39.02	N/A	N/A	5.01	510–5835	75.5
Nd	63.5–149.2	304	105.51	13.04	1592–16418	43.5
Sm	9.75–24.25	66.5	16.72	1.18	110–1147	265.5
Eu	2.19–2.67	7.1	3.69	0.36	22.8–236.5	39
Gd	7.73–10.29	54.8	9.43	1.9	53–513	1.9
Tb	0.83–1.44	6	N/A	0.28	26–52	26
Dy	4.2–6.02	N/A	5.2	1.77	29–338	2.9
Ho	0.73–1.06	N/A	0.98	0.47	23–40	12.5
Er	1.89–2.85	N/A	2.31	1.56	36–395	2.35
Tm	0.28–0.41	N/A	N/A	0.15	6–7	5.95
Yb	1.82–2.45	14.4	1.51	1.61	3–52	0.85
Lu	0.28–0.36	2.55	0.21	0.29	0.5–10	0.8
ΣLREE	325.66–648.24	1084.6	433.64	122.23	N/A	1655.4
ΣHREE	16.88–26.91	77.75	19.64	8.03	N/A	29.89
REE	342.54–674.85	1162.35	453.28	130.26	14458–199154	1685.3
ΣLREE/ΣHREE	18.01–24.11	13.95	22.07	15.22	N/A	55.38

1. Syenites of the study areas
2. Syenites of Nigeria Mesozoic granites [15]
3. Syenites of Minias Grais, Brazil [16]
4. Nepheline syenites of Gboko area, Nigeria [17]
5. Carbonatites from Wu dyke, Bayan Obo, North China [18]
6. Quartz syenites of Prakasam alkaline province, India [19]

REE mineralisation potential in the syenites and charnockites

In order to determine the mineralisation potential of the syenites and charnockites, the REE-bearing minerals in the rocks were searched for, the mineral apatite and monazite were observed, and a comparative study was carried out comparing between the syenites of this present study with related rocks, which includes the following: the nepheline syenites of the Gboko area, with ∑REE = 130.26 ppm; quartz syenites of the Prakasam alkaline province of India ∑REE = 1685.3 ppm; the quartz syenites from the Nigerian younger granites province ∑REE =1084.6 ppm; and the syenites of Capituva Massif, Brazil ∑REE = 453.28 ppm in order to determine the abundance of ∑REE in the study area syenites relative to those of other areas, (Table 3).

Charnockitic rocks of the study area were compared with related rock types to determine their ∑REE abundance relative to those of the related rock types. The comparison was done with Charnockites/enderbites of the Obudu Plateau; Garnet-charnockitic gneiss of Congo; charnockites from Kogi, Nigeria; charnockite gneiss from Norway; and charnockites from Pallavaram, Madras city, India (Table 4). The abundance of REE in the syenites and charnockites, when compared to concentrations from regions where REE has been studied and shown to be substantially enriched, suggests that the syenites are fairly enriched, while the charnockites are moderately enriched.

Further comparison was done for both the syenites and the charnockitic rocks with carbonatites from Wu dyke, Bayan Obo, North China, an area where REE is mined. The REE

Table 4: *Comparison of the REE concentration in the charnockites of the study area with other related rock types (all values in ppm).*

	1	2	3	4	5	6
REE	RANGE	RANGE	AVERAGE	AVERAGE	AVERAGE	RANGE
La	50.8–195.6	40.9–147	280.38	136	80.5	20.7–81.7
Ce	93.3–338.4	85.3–287	26.24	301	182.5	28.0–159.0
Pr	11.66–41.96	10.4–31.9	85.5	35.3	N/A	N/A
Nd	37.7–150.7	46.9–123	13.62	146	95.5	6.9–66.0
Sm	4.81–23.06	9.7–20.3	4.33	26	20	0.76–15.9
Eu	2.11–4.31	2.35–3.85	12.55	5.8	2.5	1.23–3.17
Gd	3.46–18.72	8.2–15.1	1.95	17.7	18.5	6.0–19.4
Tb	0.42–2.39	1.2–1.8	12.28	2.4	2.95	0.08–0.78
Dy	2.05–12.51	5.9–9.2	2.41	12.6	N/A	N/A
Ho	0.37–2.23	1.1–1.6	6.65	2.2	N/A	N/A
Er	1.08–5.73	3.1–4.2	0.97	5.7	N/A	N/A
Tm	0.22–0.77	0.43–0.54	6.47	0.75	1.445	0.03–2.25
Yb	0.98–4.64	2.6–3.2	0.95	4.2	8.2	0.27–13.8
Lu	0.14–0.71	0.37–0.51	30	0.63	1.13	0.21–1.91
ΣLREE	211.26–754.03	196.7–611	422	650.1	381	57.59–325.77
ΣHREE	8.64–47.7	25.9–36.3	61.7	46.18	32.23	6.59–38.14
ΣREE	219.69–801.73	220.02–647.96	484	696.28	413.23	64.18–363.91

N/A - Not Available

1. Charnockite of the study area
2. Charnockites of Obudu Plateau [20]
3. Garnet-charnockitic gneiss of Congo, South Cameroon [21]
4. Charnockites from Kogi area, Nigeria [22]
5. Charnockites gneiss from South Norway [23]
6. Charnockites from Pallavaram, Madras City, India [24]

concentration in the rocks of the study areas had very low ∑REE relative to the very high REE contents of this rock and are therefore not be economically viable.

Conclusion

The SEM studies of discrete phases of the charnockites revealed the possible presence of REE-bearing minerals, apatite, and monazite, while studies of syenites revealed a possible presence of monazite. The abundance of REEs in the syenites and charnockites, when compared to concentrations from regions where REEs have been studied and shown to be substantially enriched, suggests that the syenites are fairly enriched, while the charnockites are moderately enriched.

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