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THE ROLE OF EXTREME FRACTIONATION IN THE FORMATION OF RARE METALS: A CASE STUDY FROM GABAL DARA GRANITE AND CO-GENETIC PEGMATITES, NORTH EASTERN DESERT, EGYPT

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ABSTRACT

The granite of Gabal Dara, North Eastern Desert of Egypt, grades to the pegmatites along its boundaries, which are ranging from small pockets to large bodies with lenticular and irregular shapes in the concerned granite. Among them, three main pegmatite bodies display different degrees of enrichment in Zr, Nb, Th, U and REEs. The rare-metal mineralization is essentially composed of zircon and ishikawaite, while uranothorite and allanite-Ce are recorded as inclusions in them. A geochemical comparison among the Gabal Dara granite, barren and mineralized pegmatites indicated that they are exhibiting similar geochemical signatures; however, the mineralized pegmatites are more fractionated than the investigated granite. The Gabal Dara granite and pegmatites are highly fractionated rocks with pronounced alkaline affinity and extensive fractional crystallization. Extreme fractionation from granite to pegmatites is leading to the accumulation of the rare metals in some parts of the highly evolved pegmatites.

Key words: Fractionation, Rare Elements, Gabal Dara Granite, Pegmatites, Alkaline Affinity.

INTRODUCTION

Pegmatites of the rare-element and, in part, miarolitic classes display extreme fractionation and accumulation of numerous lithophile elements beyond the limits observed in other igneous and post-magmatic assemblages (Černý et al., 1985). Numerous studies have been carried out on pegmatites in Egypt (e.g. Abu Steet et al., 2018; El Sundoly, 2021; Mahdy, 2021). Particularly, the pegmatites in the northern domain of the Eastern Desert are recognized for their elevated contents of radioelements (Th and U). In this domain, the pegmatites appear usually as intrusive bodies encountered in the granitic rocks, which representing a complete in situ sequence from granites to highly evolved pegmatites.

Three mineralized pegmatite bodies are well-known in the Gabal Dara region, which located in the North Eastern Desert. The area located approximately 70 km southwest of Ras Gharib city (Fig. 1a), and has been the focus of substantial geological exploration (e.g. Bishady et al., 2001; Mahdy et al., 2007; Daas, 2009; El-Desoky and Hafez, 2018) and extensive ground radiometric surveying, where the radioactive pegmatites were discovered (e.g. Ali, 2007; Shalaby, 1985; El Sundoly, 2021). In this paper, mineralogical studies of the rare metal mineralization have been performed. Subsequently, a detailed geochemical comparison among the Gabal Dara granite, barren and mineralized pegmatites, is presented, to clarify the role of fractionation in the formation of rare metals.

GEOLOGICAL SETTING

The Gabal Dara area is mainly covered by Late Proterozoic younger granitoids, intruded within a volcanosedimentary successions and older granitoids as well as metavolcanics (Fig. 1b). The younger granites are represented by monzogranites, alkali feldspar granites and alkali granites. The pegmatites and aplites are abundantly encountered at the marginal parts of the alkali feldspar granites at different topographic levels. In terms of rare metal mineralization, three pegmatite bodies are recognized for their elevated radioactivity, although many pegmatite bodies are barren (Fig. 1c).

The alkali feldspar granites occur as an elongated, NNW trending block, displaying, in most cases, apparent field relation with their country rocks on the surface. Whole rock K-Ar dating indicates age of 591-602±11 Ma for this granite (Daas, 2009). The contacts of the alkali feldspar granites with the surrounding older rocks are usually of sharp intrusive nature. The effect of granitization of these older rocks is gradual and well demonstrated in their composition (Ali, 2007).

The alkali feldspar granites form an oval shaped pluton of about 12km in length and 4km width, having a rough topographic nature (Fig. 2a). They are pink to pinkish red, medium to coarse-grained rocks, showing variable weathering types and jointing systems. The alkali feldspar granites are dissected by numerous faults, mainly striking NNW-SSE (Daas, 2009). Jointing pattern is well developed as a result of the compositional stress (El Sundoly 2021). They are cut by mafic, intermediate, and felsic dykes and dyke swarms having different trends (Fig. 2b).

The pegmatites of Gabal Dara area are occurred as a massive enclave with gradational contacts, indicating that pegmatites are the in situ-differentiation product of alkali feldspar granites. They occur as unzoned and zoned bodies. Unzoned pegmatites are essentially formed of quartz, K-feldspars and flaky micas. Zoned pegmatites are composed of extremely coarse-grained milky quartz core, surrounded by reddish buff K-feldspars in the outer zones (Fig. 2c). The sizes of the pegmatites are ranging from small pockets to large bodies, exhibiting in most cases irregular shapes in the Gabal Dara granite. The dimensions of the large bodies reach to 2.5x12.5 meter across. Hematite as a product of surficial weathering is observed as stains on the surface of the K-feldspars, especially in the mineralized pegmatites (Fig. 2d).

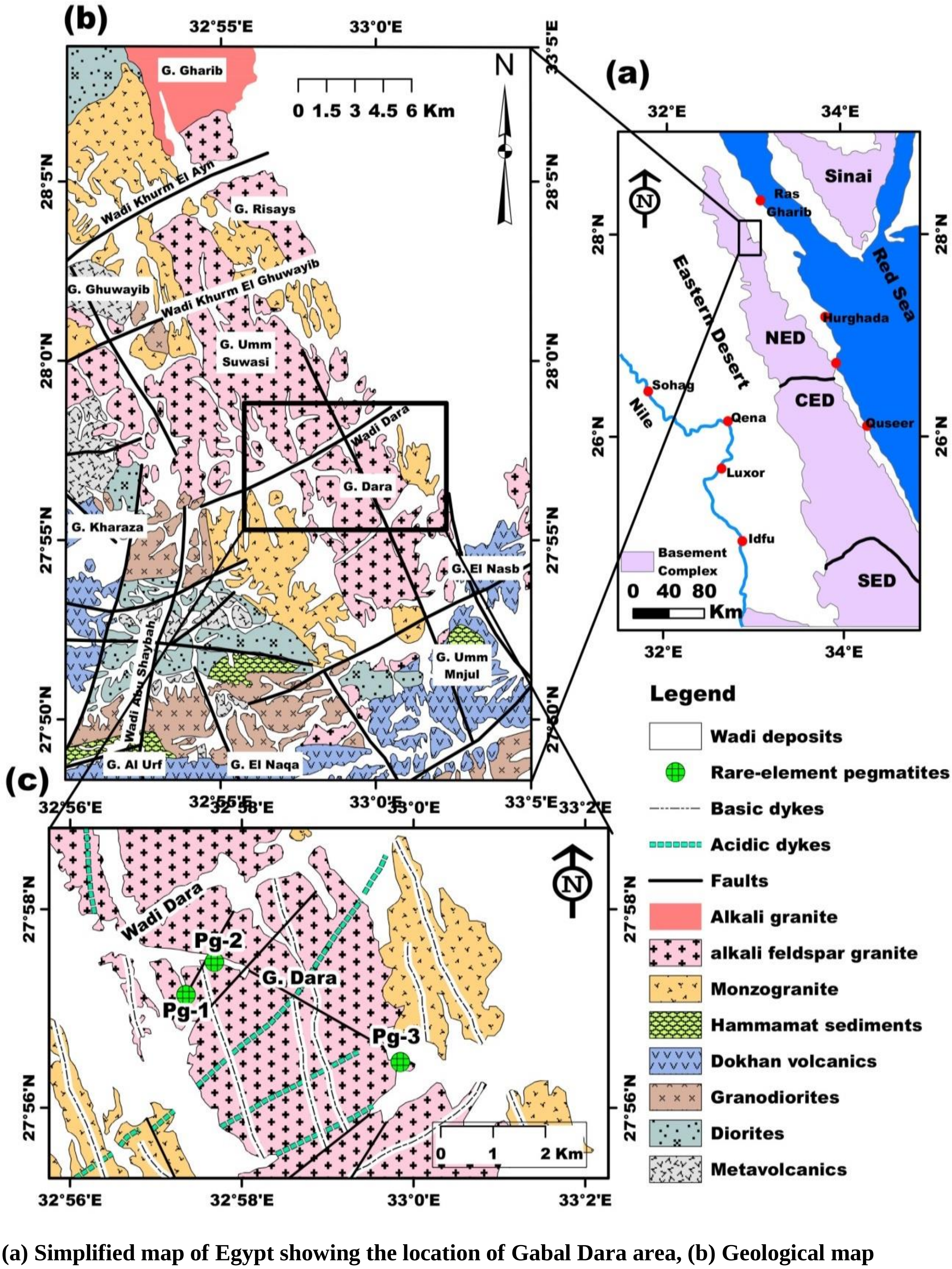

Fig. 1: (a) Simplified map of Egypt showing the location of Gabal Dara area, (b) Geological map showing the distribution of the different rock units in Gabal Dara area (modified after Daas, 2009), and (c) Locations of mineralized pegmatites.

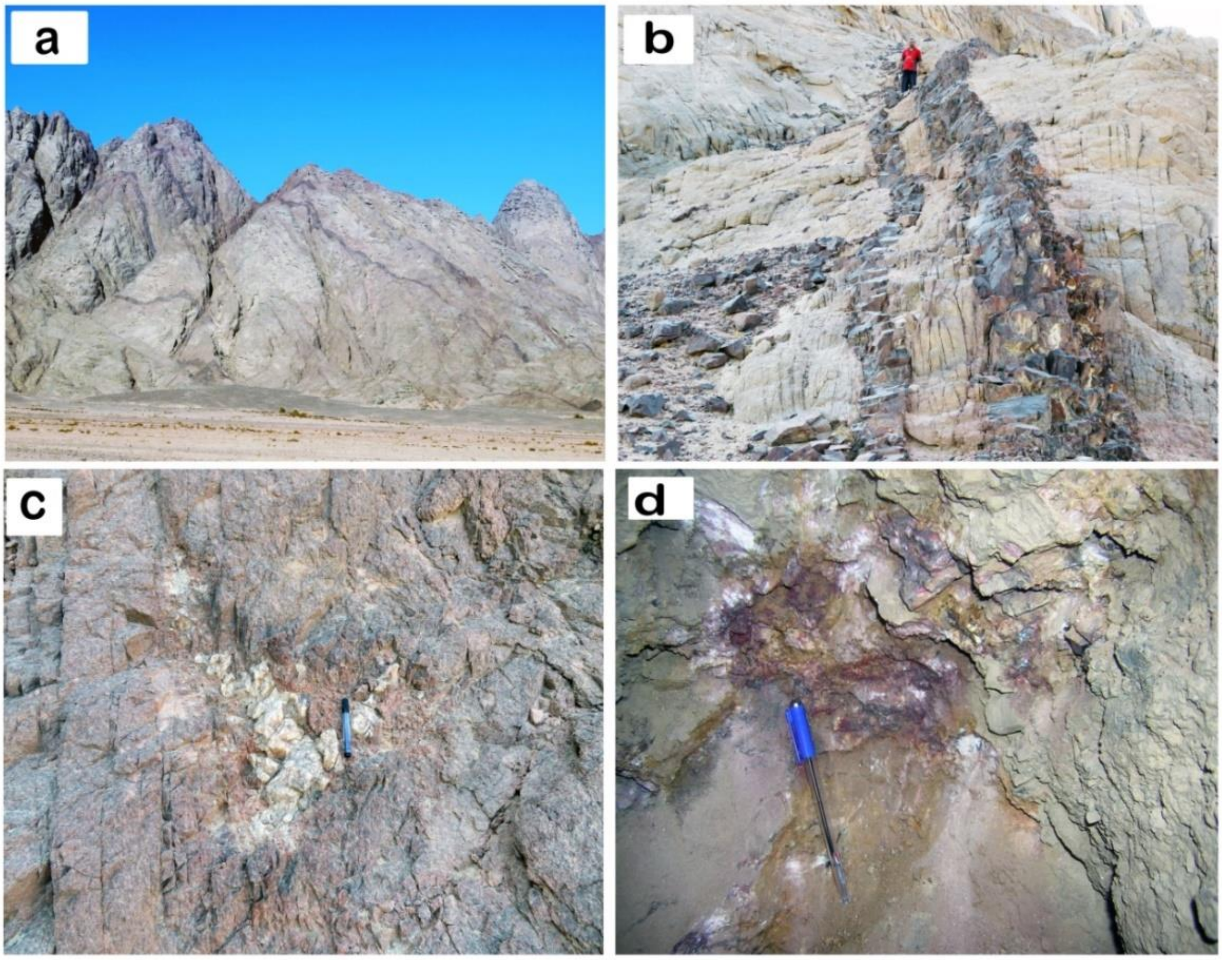

Fig. 2: Field photographs of pegmatites and the alkali feldspar granites from Gabal Dara region, (a) The alkali feldspar granites forming high relief with abundant fractures, (b) acidic dykes cut through the alkali feldspar granites, (c) Zoned pegmatite has irregular outline with quartz in the core, and (d) Mineralized portion of the pegmatite stained by red hematite.

ANALYTICAL METHODS

1- Whole-Rock Compositional Analysis

Fresh whole-rock samples from the Gabal Dara granite (6 samples), barren pegmatites (5 samples) and mineralized pegmatites (3 samples) were collected from the study area. They are crushed in a steel jawcrusher and then powdered to <200 mesh using an agate mill. Whole-rock major and trace elements were determined at the Acme laboratory, Vancouver, Canada.

2- Mineral Compositions Analysis

The rare metals are separated from the mineralized pegmatite samples by the standard heavy liquid method using bromoform (SG 2.89). Back scattered electron (BSE) imaging and energy dispersive spectroscopy (EDS) analyses were conducted over the picked mineral grains with a Phillips XL 30 Scanning Electron microscope at the laboratory of the Nuclear Materials Authority of Egypt.

RARE-METAL MINERALIZATION

Rare-metal mineralization in the Gabal Dara pegmatites is mainly represented by the Zr-, Nb-, Th-, U- and REEs-bearing accessory minerals. The rare-metal mineralization identified in the present study is described below.

Zircon ($ZrSiO_4$) grains from the mineralized pegmatites are subhedral to euhedral crystals. It forms short prismatitic grains or dipyramidal crystals (Fig. 3a). They are transparent and typically a few millimeters in size. Their colors are principally brown although dark brown and yellow are also encountered. The studied zircon grains are metamicted due to radiation. Its chemistry is more complex, as it shows large variations of U, Th, Ca, Hf, REEs and other elements (Fig. 3a). The crystals are porous and host abundant inclusions, identified by EDS as allanite-Ce (Fig. 3b).

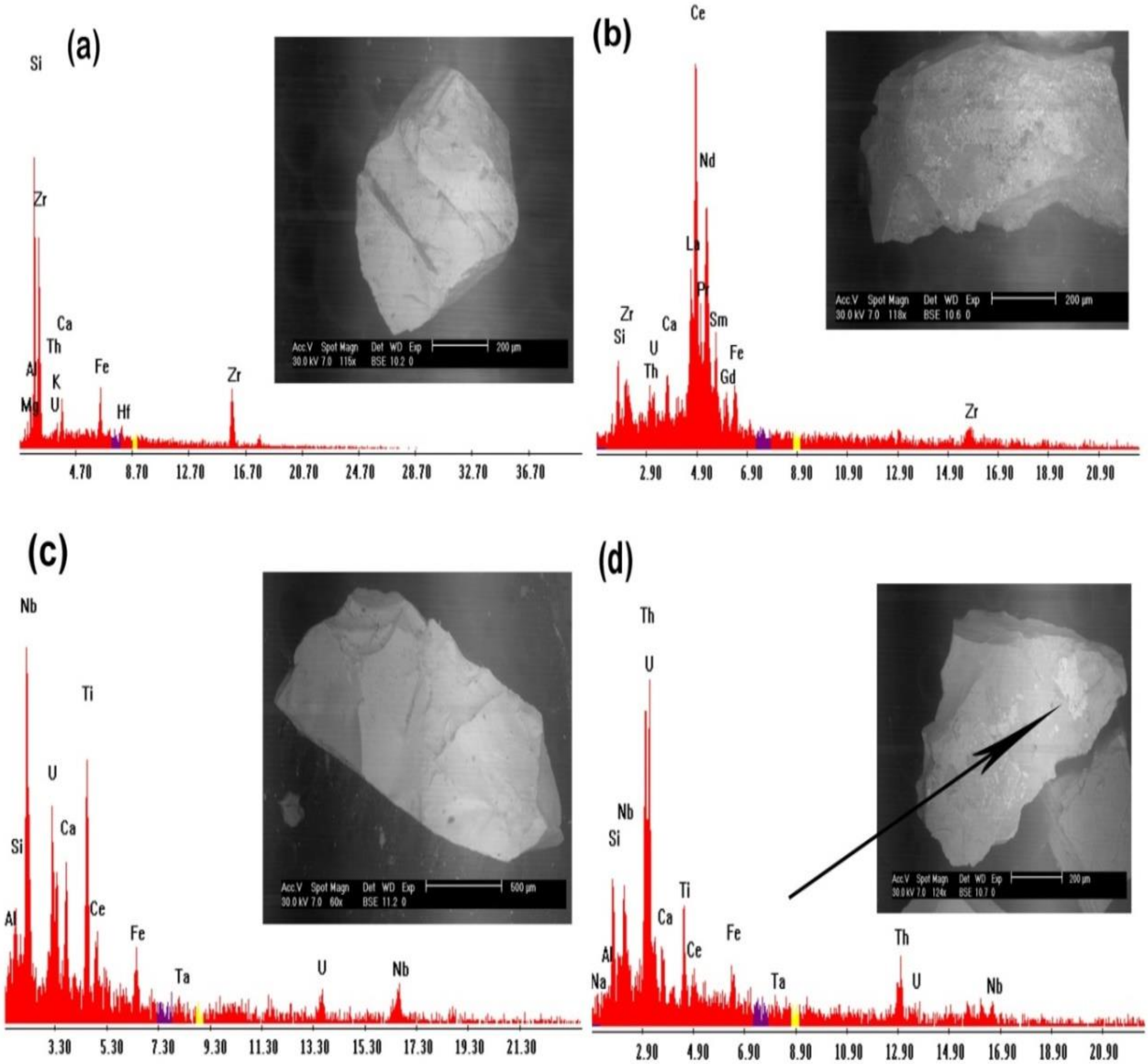

Fig. 3: Back-scatter electron images and EDS analyses of rare-metal assemblages from the Gabal Dara mineralized pegmatites, (a) Dipyramidal crystal of zircon, (b) Inclusions of allanite-Ce in zircon, (c) Ishikawaite, and (d) Inclusions of uranothroite in Ishikawaite.

Ishikawaite [(U, Fe, Y, Ca) (Nb, Ta) O_4] occurs as black to dark brown anhedral grains with a black to brown streak. Ishikawaite is opaque with vitreous luster. Cleavage and parting are absent because it is completely metamict. The destruction of a mineral crystal lattice in a metamict mineral is reflected in its physical properties. Usually, this means a decrease in the density, the hardness, the birefringence and the refraction indices for metamict minerals. The obtained EDX analysis of ishikawaite indicates that Nb, U, and Ti are the essential components (Fig. 3c). Other elements present in small to minor amounts include Ta, Ce, Fe, Ca and Si. Inclusions of uranothorite [(Th,U)SiO_4] are detected through the studied ishikawaite grains (Fig. 3d).

RESULTS

WHOLE-ROCK GEOCHEMISTRY

1- Gabal Dara Granite

Whole-rock major- and trace-element results for the Gabal Dara granite, barren and mineralized pegmatites are listed in Tables (1&2). The Gabal Dara granite is characterized by high and restricted SiO_2 range of 76.12–77.28 wt% with high Na_2O+K_2O contents (8.65–9.27 wt%). Generally, this granite exhibits lower Al_2O_3 (11.06– 11.85 wt%), TiO_2 (0.06–0.12 wt%), FeOt (0.09–0.28 wt%), CaO (0.37–0.51 wt%), MgO (0.08–0.17 wt%), and P_2O_5 (0.02–0.04 wt%).

The studied granite can be classified using the cationic classification R_1-R_2 of De la Roche et al., (1980), whereas the studied samples are entirely located in the alkali granite field (Fig. 4). On major elements discrimination diagram of Sylvester, (1989), the studied granitic samples plot in the alkaline field (Fig. 5). Moreover, on the A/NK-A/CNK diagram (Fig. 6), the studied samples plot in the peralkaline field (A/CNK<1). The studied granite has low Zr and Nb, contents, distinguishing them from A-type granites (Whalen et al., 1987). On the (Na_2O+K_2O)/CaO vs. Zr + Nb + Ce + Y diagram, the studied samples fall within the field of fractionated I-type granites (Fig. 7).

2- Pegmatites

The barren and mineralized granitic pegmatites have lower SiO_2 contents (averages of 72.16 and 65.68 wt %, respectively), compared to the Gabal Dara granite (Table 1). The values of TiO_2, MgO and CaO < 1 wt %, and Na_2O + K_2O values are high in contrast to the studied granite confirming albitization in the pegmatites. The A/NK of the pegmatites is >1 such as the Gabal Dara granite.

Table (1): Major oxides (wt. %) and trace elements (ppm) analyses with some parameters and ratios of the Gabal Dara granite, barren pegmatites and mineralized pegmatites.

Elements	Gabal Dara highly fractionated granite						Feldspar-rich barren pegmatites					Feldspar-rich mineralized pegmatites		
	HF G-1	HF G-2	HF G-3	HF G-4	HF G-5	HF G-6	BP-1	BP-2	BP-3	BP-4	BP-5	MP-1	MP-2	MP-3
Major elements (Wt.%)														
SiO_2	76.23	76.35	76.12	76.84	77.12	77.28	73.15	72.85	69.5	71.4	73.9	65.79	63.9	67.36
TiO_2	0.12	0.08	0.08	0.1	0.06	0.06	0.12	0.19	0.1	0.25	0.28	0.52	0.71	0.45
Al_2O_3	11.85	11.56	11.54	11.36	11.15	11.06	13.15	13.38	15.3	14.8	12.75	15.37	16.52	14.21
Fe_2O_3	1.12	1.18	1.06	1.08	1.24	0.88	0.93	1.41	1.51	1.24	0.89	1.88	3.96	2.96
FeO	0.17	0.21	0.28	0.15	0.18	0.09	0.08	0.07	0.04	0.08	0.07	0.02	0.04	0.05
MnO	0.06	0.01	0.08	0.04	0.04	0.03	0.01	0.02	0.03	0.02	0.03	0.05	0.08	0.04
MgO	0.12	0.17	0.16	0.1	0.08	0.1	0.09	0.06	0.05	0.08	0.1	0.24	0.27	0.12
CaO	0.48	0.51	0.42	0.37	0.41	0.39	0.51	0.48	0.35	0.5	0.64	0.95	0.64	0.53
Na_2O	4.62	4.88	4.65	4.47	4.69	4.71	5.14	6.32	6.18	5.82	5.48	5.54	5.29	6.4
K_2O	4.23	4.39	4.32	4.18	4.21	4.45	5.36	4.13	5.74	4.76	5.06	7.21	6.24	5.13
P_2O_5	0.02	0.03	0.04	0.02	0.03	0.02	0.04	0.06	0.08	0.06	0.04	0.16	0.08	0.06
L.O.I	0.95	0.63	1.25	1.22	0.79	0.88	1.42	1.02	1.12	0.98	0.76	2.24	2.27	2.64
Total	99.97	100	100	99.93	100	99.95	100	99.99	100	99.99	100	99.97	100	99.95
Trace elements (ppm)														
Ba	325	124	491	489	265	185	35	46	32	52	29	55	48	36
Rb	145	167	118	123	112	131	310	215	340	236	325	149	286	215
Sr	43	24	33	38	25	21	28	41	65	32	31	57	31	39
Cs	2.1	1.8	1.3	2.5	1.6	2.2	3.2	4.2	5.6	3.5	3.8	4.9	4.1	2.2
Ga	19.5	22.4	18.5	18.1	18.8	17.9	48	55	75	62	31	63	85	51
Ta	1.47	1.36	1.58	1.16	1.12	1.36	6	7	8	6	7	126	41	25
Nb	13.5	13.8	19.2	14.5	13.1	17.2	34	35	41	31	40	1297	256	90
Hf	5.46	4.24	4.83	4.79	4.51	4.73	7	6	5	6	4	112	46	21
Zr	161	102	135	125	108	131	98	56	67	51	54	937	423	108
Y	30	27	37	29	28	30	47	41	38	45	35	508	128	93
Ni	4	3	4	3	3	2.5	3	3	4	4	3	1.6	3.1	2.5
Cr	9.4	10.5	5.2	6.3	5.3	4.2	5	6	4	3	2	12	8	8
Co	18	20	18	25	31	21	0.3	0.4	0.1	0.8	0.7	0.8	0.6	0.3
V	10	9	12	18	12	9	5	5	4	3	5	12	9	4
Cu	8	8	7	4	9	15	4	5	7	8	12	4	6	15
Pb	89	80	60	55	68	97	36	55	61	38	40	68	41	87
Zn	192	221	280	193	145	292	85	92	105	98	76	139	286	114
Th	22.7	25.7	23.2	30.8	27.6	21.2	28	31	25	34	42	640	315	148
U	4.5	5.8	5.5	8.3	6.7	5.4	12	14	8	15	19	321	159	77
Nb/Ta	9.18	10.15	12.15	12.5	11.7	12.65	5.67	5	5.13	5.17	5.71	10.29	6.24	3.6
Zr/Hf	29.49	24.06	27.95	26.1	23.95	27.7	14	9.33	13.4	8.5	13.5	8.37	9.2	5.14
Th/U	5.04	4.43	4.22	3.71	4.12	3.93	2.33	2.21	3.13	2.27	2.21	1.99	1.98	1.92

Table (2): Rare earth element concentrations (ppm) of the Gabal Dara granite, barren pegmatites and mineralized pegmatites.

Elements	Gabal Dara highly fractionated granite						Feldspar-rich barren pegmatites					Feldspar-rich mineralized pegmatites		
	HFG -1	HFG -2	HFG -3	HFG -4	HFG -5	HFG -6	BP-1	BP-2	BP-3	BP-4	BP-5	MP-1	MP-2	MP-3
La	35.2	26.5	33.9	32.1	25.9	40.8	14.4	3.8	11.9	3.9	4.9	16.2	9.8	6.4
Ce	80.9	48	69.2	64	60.2	81.5	32.4	63.7	26.7	65.7	16.1	97.2	32.2	107.2
Pr	9.6	6.2	6.5	6.3	5	8.1	4.8	1.9	4.0	2.0	4.2	18.6	8.4	3.2
Nd	31.8	20.2	16.2	12.9	13.7	16.5	19.2	6.7	15.8	6.9	18.2	118.8	36.4	11.2
Sm	6.1	4.8	6.2	3.3	1.7	4.2	7.2	28.5	5.9	29.4	10.5	64.2	21	48
Eu	0.5	0.3	0.4	0.3	0.1	0.3	0.12	0.10	0.10	0.10	0.07	0.12	0.14	0.16
Gd	4.1	4.1	2.9	5.4	1.6	6.9	8.4	3.8	6.9	3.9	14.7	80.4	29.4	6.4
Tb	0.8	1.1	0.7	1.7	0.3	2.2	1.6	1.0	1.28	0.98	3.5	19.2	7.0	1.6
Dy	5.5	9	7.3	5.8	1.6	7.4	9.7	9.5	8.0	9.8	27.3	147.6	54.6	16
Ho	1.1	2.4	1.6	2.1	1.1	2.7	2.3	2.7	1.9	2.7	6.3	33	12.6	4.48
Er	4	9.1	5.9	7.1	1.7	9.1	7.0	11.4	5.7	11.8	22.4	115.8	44.8	19.2
Tm	0.9	1.8	1.6	1.7	1.1	2.2	1.4	2.5	1.2	2.5	4.2	20.4	8.4	4.16
Yb	7	12.9	7.4	8.2	3.4	10.4	12.0	21.9	9.9	22.5	30.8	145.2	61.6	36.8
Lu	1.1	2	1.6	1.1	0.5	1.3	1.9	3.8	1.6	3.9	5.0	21	10.1	6.4
ΣREE	188.6	148.4	161.4	152	117.9	193.6	122.4	161.0	101.0	166.1	168.2	897.7	336.4	271.2

Rb contents in the barren pegmatites (av.= 285ppm) and mineralized pegmatites (av.=216ppm) are higher compared to the Gabal Dara granite (av.=132ppm). In contrast, the values of Ba (av.=39, 46ppm) are very low compared to the studied granite (av.=313ppm), while Sr contents (av.=39, 42ppm) are slightly higher than the investigated granite (av.=30ppm). However, the concentrations of these elements are below average crustal abundances of Wedepohl, (1995). Nb and Ta values are higher in the barren pegmatites (av.=36.2, 6.8ppm) with reference to the Gabal Dara granite, and progressively highly increased in the mineralized pegmatites to 548 and 64ppm, respectively (Table 1).

The pegmatites in the study area show more variable trace element and REEs patterns and are strongly fractionated especially the mineralized pegmatites. On the primitive mantle-normalized spider diagrams, most of the pegmatite samples show positive anomalies of Rb, Th, K, Nb, Ta, U and Zr, and negative anomalies of Ba, Pb, and Ti similar to the Gabal Dara granite (Fig. 8). The major difference between the pegmatites and the average contents of Gabal Dara granite are the enrichment of U, Th, Zr, and Nb in the pegmatites, which can be attributed to the fractionation of pegmatites.

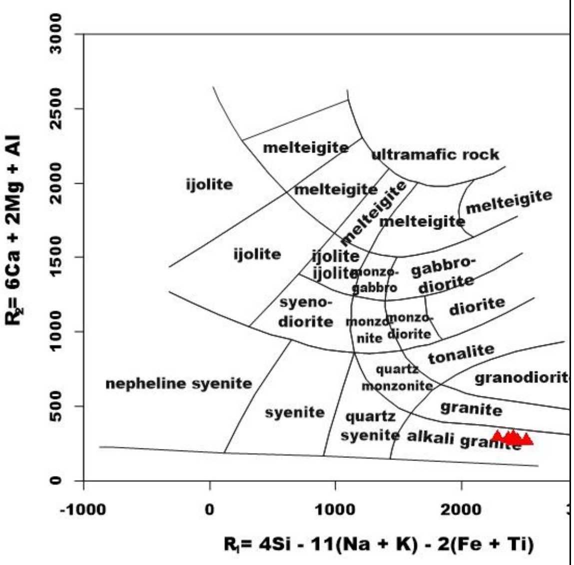

Fig. 4: Chemical classification diagram R1-R2 of De La Roche et al., (1980) for the studied granite.

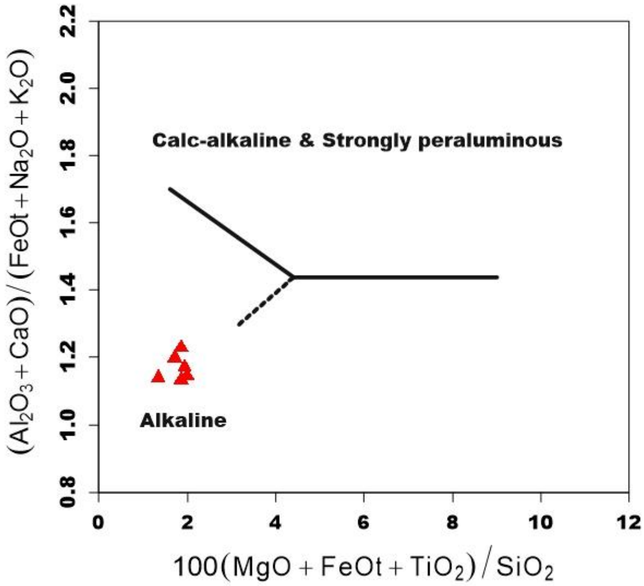

Fig. 5: Discrimination diagram of Sylvester, (1989) for the investigated granite.

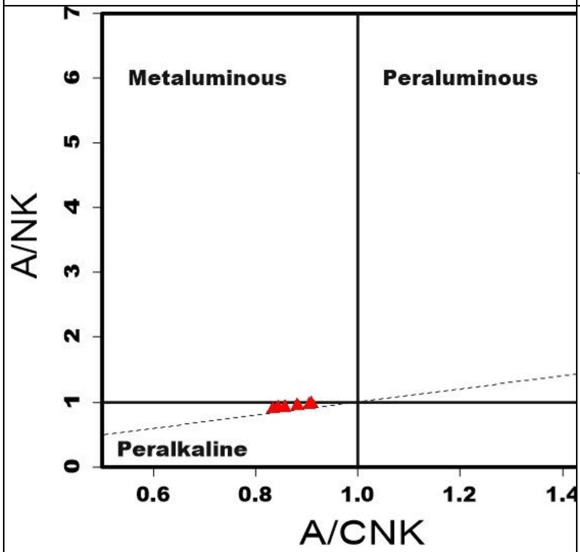

Fig. 6: A/CNK versus A/NK classification diagram after Maniar and Piccoli, (1989) for the studied granite.

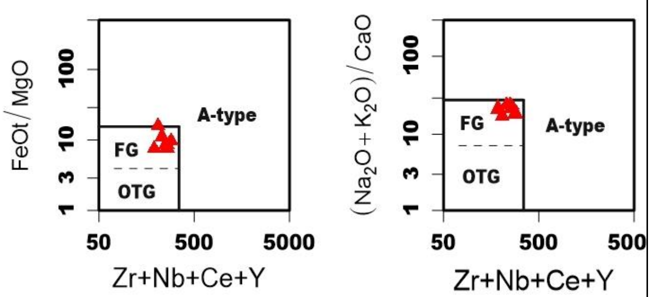

Fig. 7: A-type discrimination diagrams of Whalen et al., (1987) for the studied granite. OTG: unfractionated I-, S- and M-type granites, FG: fractionated I-type granites.

The barren pegmatites generally show lower concentrations of REEs (av.=143ppm) compared to the Gabal Dara granite (160ppm), while the mineralized pegmatites display high concentrations of REEs (av.=501ppm; Table 2). The studied granite and associated pegmatites display nearly similar patterns on the chondrite-normalized REE diagrams, which are characterized by seagull-like normalized REE pattern, with significant negative Eu anomalies (Fig. 9). The Eu concentrations in the pegmatites are very low compared to the studied granite. The Eu anomalies can be explained by fractional crystallization of feldspars and to melt-fluid interaction in the last stage of magma solidification (Abdelfadil et al., 2016; Chukwu and Obiora, 2014).

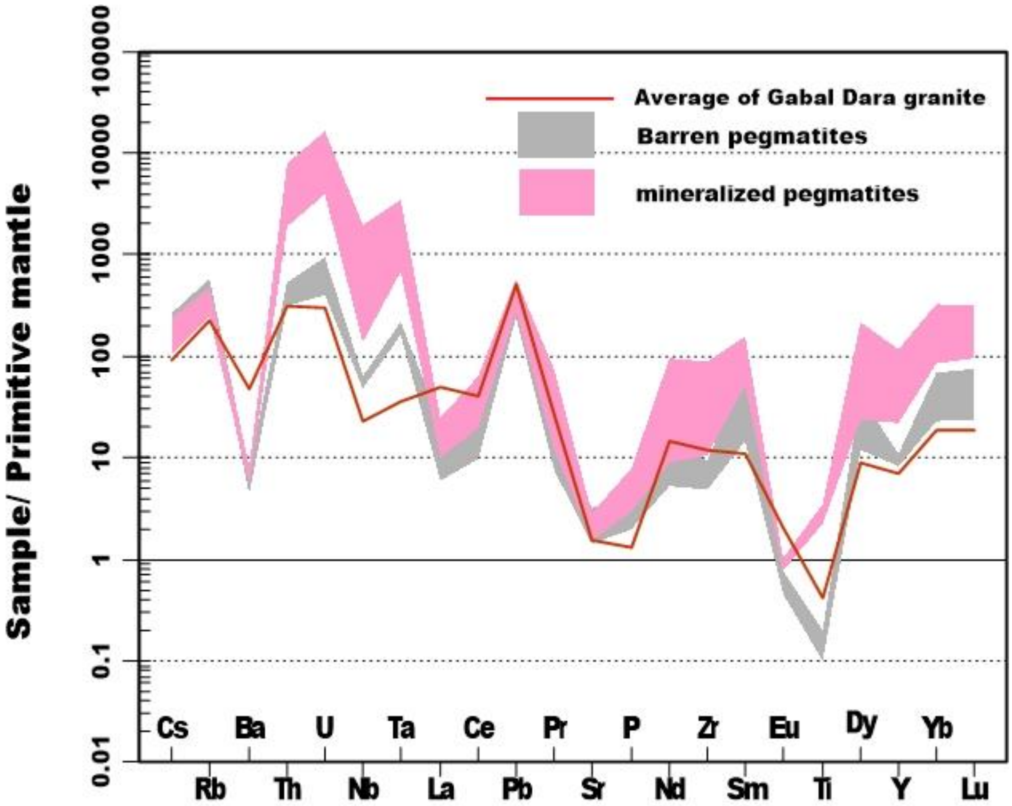

Fig. 8: Primitive-mantle normalized multi-element diagram (McDonough and Sun, 1995) of the studied pegmatites.

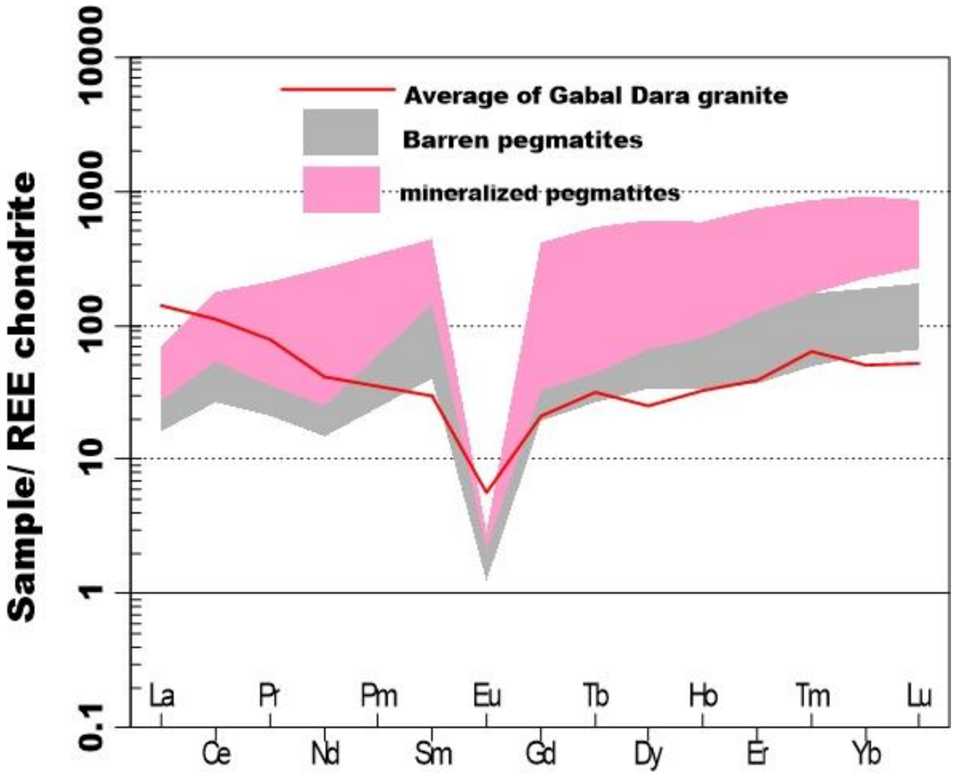

Fig. 9: Chondrite-normalized REE patterns (Anders and Grevesse, 1989) of the investigated pegmatites.

DISCUSSION

1- Fractionation processes

The alkali feldspar granites at Gabal Dara area is characterized by several geochemical features indicating of extensive crystal fractionation, such as strong depletions in Sr, Ba, Ti, P, and Eu relative to Zr, Nb, Rb and REEs (except Eu). Such geochemical characteristics are often observed in peralkaline igneous rocks thought to represent the most evolved melts produced via fractional crystallization of mafic parental magmas (e.g. Zozulya et al., 2009; Dostal and Shellnut, 2016; Jeffery and Gertisser, 2018).

The Zr/Hf, Rb/Sr or Nb/Ta ratios are used generally as an indicator of pegmatite evolution (Halliday et al., 1991; Linnen and Keppler, 2002; Bau, 1996). The trends of the plots of La versus Zr/Hf ratios (Fig. 10a) could be interpreted in terms of zircon fractional crystallization from alkali feldspar granites to the pegmatites. The plots of La versus Rb/Sr ratios (Fig. 10b) show evolution trends that could be interpreted in terms of feldspar and mica (biotite and/or muscovite) and fractional crystallization. The trends of the plots of La versus Nb/Ta ratios could be interpreted in terms of mica (biotite and/or muscovite) fractional crystallization (Fig. 10c). The overall geochemical evolution is consistent with fractional crystallization from alkali feldspar granites to pegmatites.

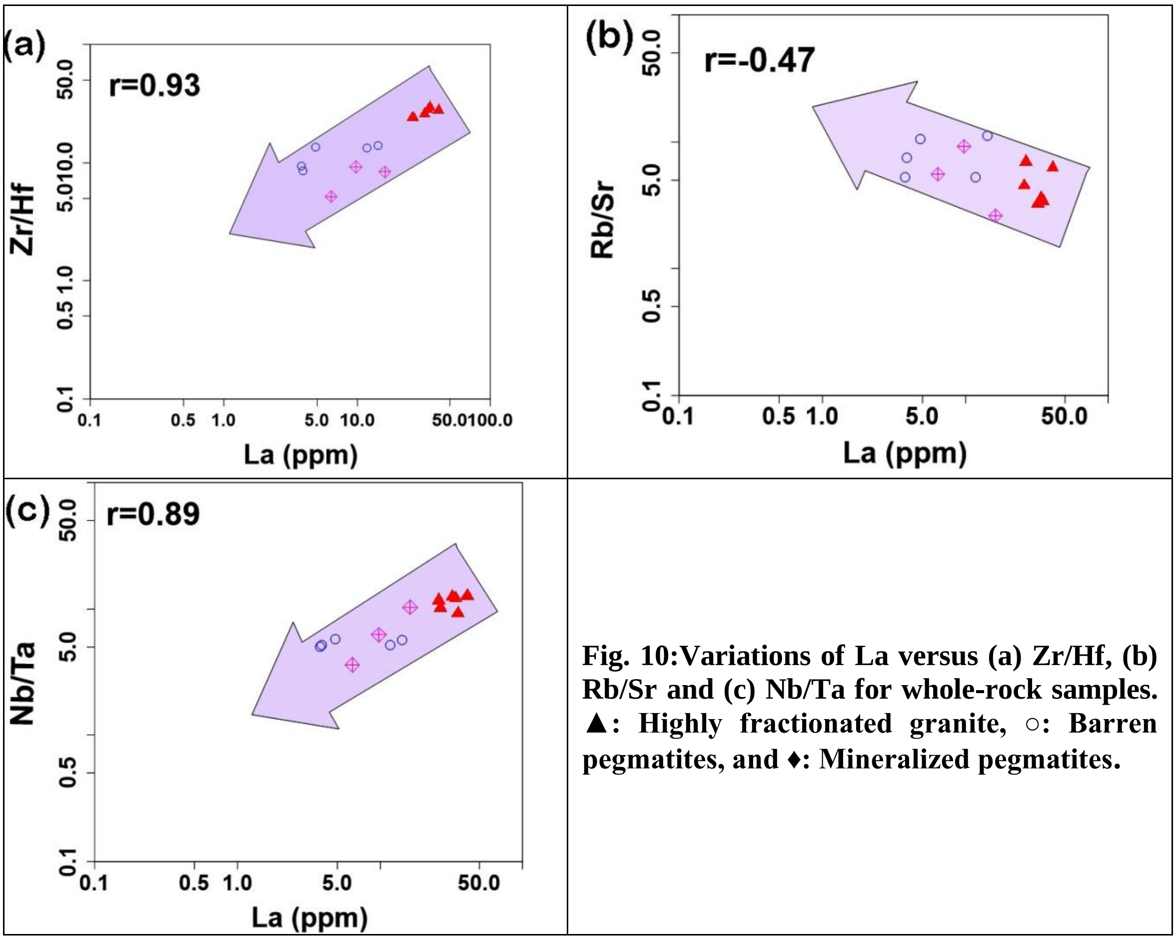

Fig. 10:Variations of La versus (a) Zr/Hf, (b) Rb/Sr and (c) Nb/Ta for whole-rock samples. ▲: Highly fractionated granite, ○: Barren pegmatites, and ♦: Mineralized pegmatites.

2- Rare metal enrichment

Most fertile crustal lithologies contain sufficient amounts of REEs, Y, Zr, and P to supply an anatectic melt in these components and allow early precipitation of accessory phases (Watt and Harley, 1993). Such an enrichment of Zr-Nb-Th-U and REEs in pegmatites is due to progressive fractionation leading to gradual enrichment of these elements in successively evolved melts, with maximum concentration being in geochemically most evolved pegmatites. It, thus, also suggests that the parent granites formed from the evolved melts, and the co-existing pegmatites had formed from still more evolved melts. As that case, with the investigated felsic rocks, such evolved granite-pegmatites are also known for hosting rare metals and rare earths from elsewhere (Christiansen et al., 1988; Černý and Meintzer, 1985; Breaks et al., 2005; Cuney and Kyser, 2009).

Tantalum and, to a lesser degree also titanium, are relatively immobile in most granite-related hydrothermal systems which is why the whole-rock abundances of these two elements can be used as an approximative indicator of magmatic fractionation in a given granite sample (Lehmann, 1993). Distribution patterns of U+Th, Zr, Nb and total REEs as a function of Ta are shown in Fig. (11). These patterns reflect both a magmatic rare-element enrichment trend of variable degree, which give good linear correlation trends in log-log space, suggestive of element control dominantly by magmatic processes, i.e. fractional crystallization.

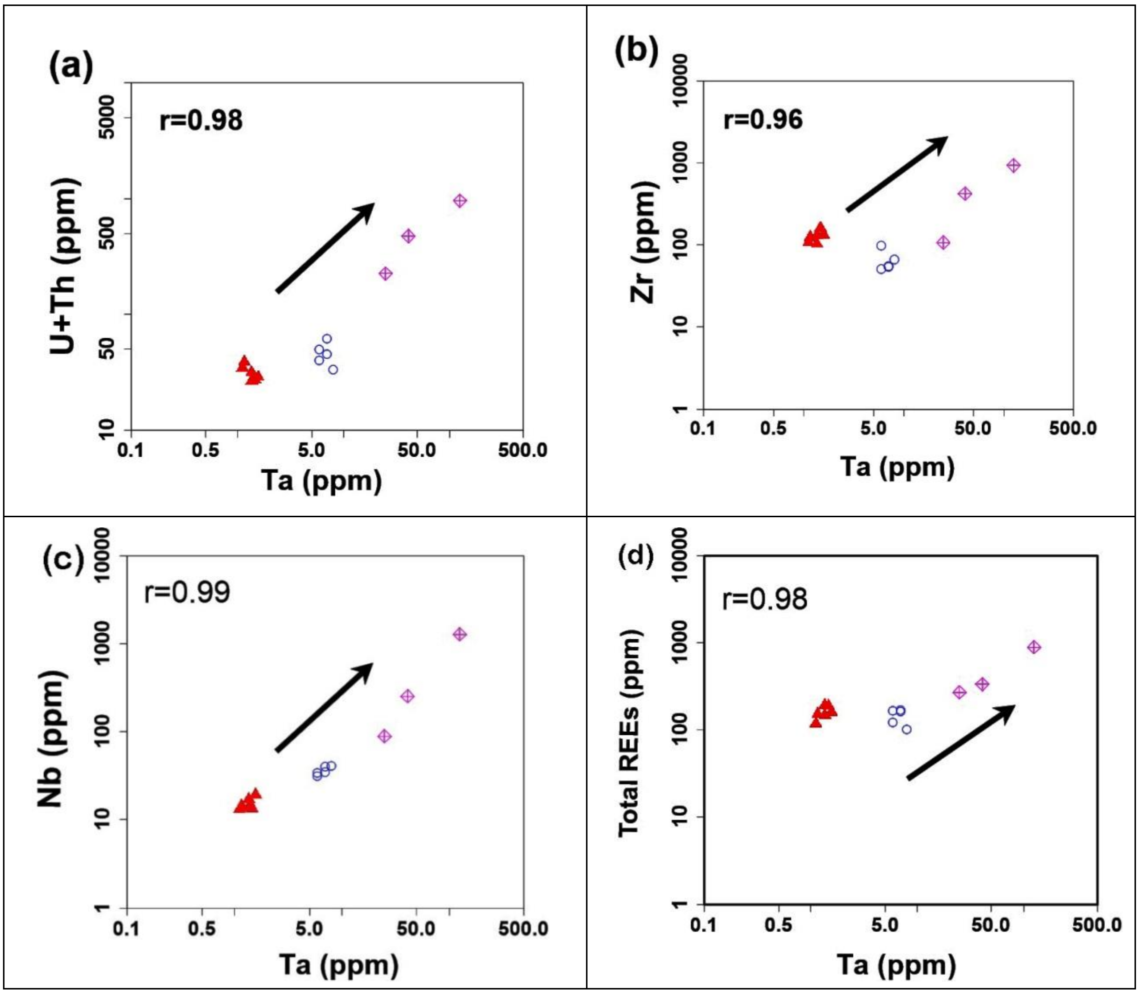

Fig. 11: Ta (ppm) vs. (a) U + Th (ppm), (b) Zr (ppm), (c) Nb (ppm), and (d) Total REEs (ppm) variations plots, with arrows indicating the direction of progressive fractionation, leading to enrichment in Zr, Nb, Th, U and REEs in the mineralized pegmatites. Symbols as in Fig. 10.

CONCLUSIONS

The alkali feldspar granites of Gabal Dara area contain numerous pegmatites in its peripheral parts. Three main pegmatite bodies are well-known in such area to be radiometrically anomalous. In the present study, the alkali feldspar granites, barren and mineralized pegmatites were sampled and geochemically studied to clarify the role of fractionation in the formation of rare metals in the pegmatites. The alkali feldspar granites show highly fractionated characteristics with pronounced alkaline affinity. The chemical composition of the alkali feldspar granites and their associated pegmatites from the Gabal Dara region, suggests that the pegmatites differed from granites by their high Zr, Nb, Th, U and REEs contents. Such an enrichment of these elements is due to progressive fractionation, leading to gradual enrichment of these elements in most evolved pegmatites. The overall geochemical evolution is consistent with fractional crystallization trend from alkali feldspar granites to pegmatites.

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