



HYDROTHERMAL ZIRCON, NB-TA AND REE MINERALIZATION IN RADIOACTIVE PEGMATITE-APLITE DYKES, NORTH RAS MOHAMMED AREA, SOUTH SINAI, EGYPT

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Abstract: The alkaline pegmatite-aplite dyke system hosted in alkaline granite was identified in the north Ras Mohammed area, south Sinai, Egypt. It consists of brown-red dykes up to several kilometers long and one meter wide. The system is generally trending NE-SW, E-W, N-S and NW-SE directions. The dykes are fine to medium-grained rocks with granular texture and are mainly composed of K-feldspar as microperthite, quartz, riebeckite and plagioclase. The dykes have anomalous radioactivity up to 305 ppm U. They are characterized by the presence of hydrothermal zircon (enriched with U, Th and trace elements) which recorded for the first time in Sinai as far as the author is aware, in addition to Th (53 to 735.6 ppm), Nb (91.7 to 5121.9 ppm), Ta (8.5 to 693.5 ppm), Y (81.9 to 1915.8 ppm), Zr (623 to 16950.4 ppm), La (9.8 to 275.6 ppm), Ce (39.3 to 923.7 ppm) and Σ REE (143.36 to 2271.49 ppm). Based on scanning electron microscope (SEM) technique studies, some of the previously mentioned elements form its own minerals, such as hydrothermal zircon, columbite mineral group and Tornebohmite-(Ce) minerals.

Introduction

In general, alkaline granitoid rocks contain rare metal deposits (Zr-Hf-Nb-Ta-REE etc.) Clarke (1992). Jackson *et al.* (1985) and Kinnaird *et al.* (1985) mentioned that the mineralized plutons are late in the magmatic cycle, that high levels of fluorine may indicate that it was responsible for complexing the REEs and other high field-strength (HFS) elements, and that the mineralization occurs as disseminations both in the host pluton and in the matrices of breccia pipes, and as concentrated zones in aplite-pegmatite-dyke system. The influence of F may go all the way back to the depleted source region at the time of partial melting. The alkaline granites and few pegmatites are of particular interest in the exploration for Zr, Nb, Ta, Be, Y, and rare earth element (REE) mineral resources (e.g. Currie, 1976; Sinclair *et al.*, 1992; Sørensen, 1992; Richardson and Birkett, 1995).

The present aplite-pegmatite dykes system is hosted by alkaline granites at Wadi Lathi, south Sinai, Egypt has high radiometric anomalies and rare earth elements REE mineralization (Moselhi 2006). These dykes also contained a hydrothermal zircon which incorporate considerable amount of uranium, thorium

and REE. The term "hydrothermal zircon" is an imprecise term that may in a general way be used to refer to zircon crystallized from or altered by an aqueous fluid.

In this paper a geological map of the pegmatite-aplite dykes, exposed in north Ras Mohammed area involving petrography, major- and trace elements and REE geochemistry has been given. Hydrothermal zircon, U, Th and REEs-bearing minerals will be studied using SEM technique.

Field description and petrography

The aplite-pegmatite dykes are previously mentioned by Mosalhi, (2006) during his studies on the granitoid rocks of Ras Mohammed area, south Sinai (Fig.1). They occur as intersected dykes with different trends and sizes. These dykes are hosted by the alkaline granites without clear sharp contact. The diffusive nature of these contacts may suggest the late stage alkaline magma emplacement of these dykes. The dykes have different widths ranging from 40 cm to one meter (Fig. 2). They are medium to very coarse-grained with brownish red colour. They also subjected to severe alteration due to the effects of hydrothermal solutions manifested by both hematitization and kaolinitization, especially at the contact with the hosted rocks, in addition to the presence of hydrothermal zircon.

The aplite-pegmatite dykes are composed mainly of quartz, k-feldspar, riebeckite and minor plagioclase. Quartz occurs in subhedral grains filling the interspaces between the other constituents. It shows some cracking and undulose extinction due to deformation effects. Potash feldspars are mostly represented by fine string microperthite occasionally corroded by quartz. Plagioclase is less in abundance and shows well developed albite twinning and have prismatic nature and relatively fresh. Riebeckite occurs as subhedral crystals with blue colour. The presence of riebeckite suggests the alkaline affinities of these dykes (Fig. 3).

Sampling and analytical Techniques

The separation of heavy minerals was carried by using heavy liquids separation technique in the laboratories of the Nuclear Materials Authority of Egypt. Major oxides, trace elements and REE were analyzed in ACME Analytical Laboratories, LTD., Vancouver, Canada. Mineral chemistry was determined using SEM attached with EDAX unit, Nuclear Materials Authority, Egypt.

Geochemistry

Eight samples were collected from the aplite-pegmatite dykes for chemical analyses; five samples were chosen for major oxides analyses while all the eight samples were analyzed for trace and rare earth elements.

The data obtained of the major oxides and CIPW norms are given in Table 1.

Figure 4 represents the R1-R2 discrimination diagram of De La Roche et al. (1980), where $R1 = (4Si - 11(Na + K)) - 2(Fe + Ti)$ and $R2 = 6Ca + 2Mg + Al$ (millications), the samples of aplite-pegmatite rocks fall within alkali granite field (Fig. 4). The presence of acmite in most samples suggests the alkaline nature of the studied aplite-pegmatite dykes.

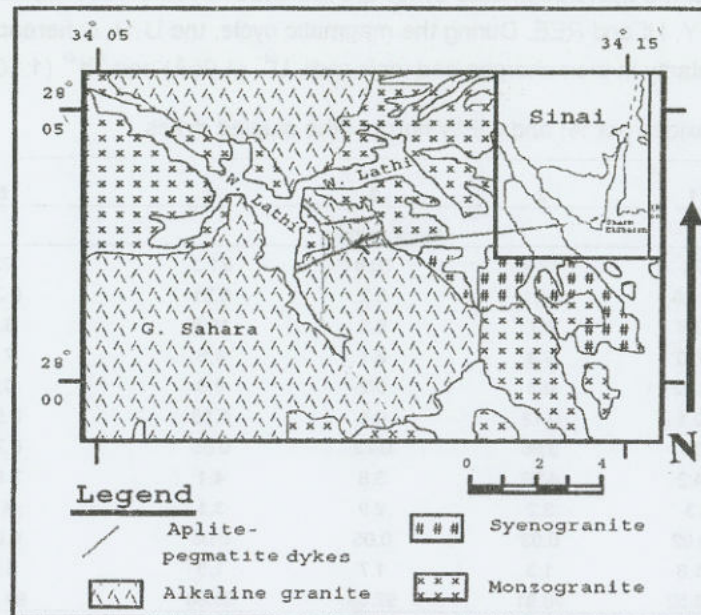


Fig. (1): Geologic map of aplite-pegmatite dykes, north Ras Mohamed, Sinai, Egypt.(Modified after Mosehi, 2006)

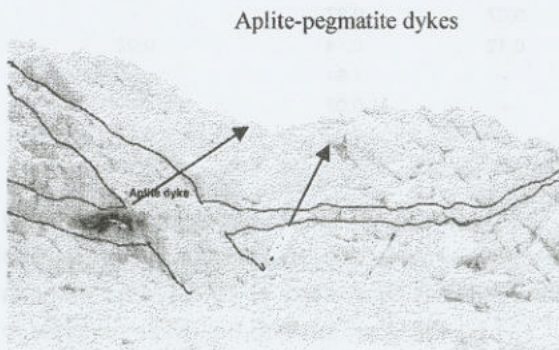


Fig. (2): Aplite-pegmatite dykes hosted by alkaline granites



Fig. (3): Microphotograph shows sodic amphibole (S) in aplite dykes (PPL., x25).

Shand's index diagram is originally used to distinguish between metaluminous, peraluminous and alkaline rock suites. The data of the studied samples plot within the peralkaline field, except one sample filled in metaluminous field (Fig. 5).

Batchelor and Bowden (1985) used the R1 and R2 multi-cationic parameters of De La Roche *et al.* (1980) to discriminate between the different tectonic settings of granitoid rocks, (Fig. 6). The studied aplite-pegmatite samples fall near the Post-Orogenic field.

Trace and rare earth elements contents are listed in Tables (2 and 3) respectively. The data show that the aplite-pegmatite dykes are enriched in some trace elements such as U, Th, Nb, Ta, Zr, Y, Hf and REE. During the magmatic cycle, the U-Th coherence attributed to the marked similarity in their charges and ionic radii, U^{4+} (1.05Å) and Th^{4+} (1.10Å).

Table 1. Major oxides (wt %) and CIPW norm of the studied dykes.

Sample No.	1	2	3	4	5
Major oxides					
SiO ₂	71	72.6	70.8	71.5	72
TiO ₂	0.28	0.21	0.2	0.21	0.23
Al ₂ O ₃	9.6	8.7	9.3	10.8	8.7
Fe ₂ O ₃	7.6	6.8	8.1	6.5	7.9
MnO	0.22	0.3	0.2	0.3	0.2
MgO	0.1	0.12	0.1	0.16	0.51
CaO	0.7	0.66	0.73	0.65	0.74
Na ₂ O	4.2	4.99	3.8	4.1	3.99
K ₂ O	3	3.2	2.9	3.1	3.1
P ₂ O ₅	0.02	0.03	0.05	0.06	0.01
L.O.I	1.8	1.3	1.7	1.3	1.6
Total	98.52	98.91	97.88	98.68	98.98
CIPW norm					
Qz	34.87	33.99	36.84	34.52	36.82
Or	17.73	18.91	17.14	18.32	18.32
Ab	32.68	26.94	31.69	34.69	27.49
Ac	2.52	13.46	0.4	-	5.52
Di	0.54	0.64	0.54	0.72	2.74
Wo	1.06	0.94	1.09	-	0.03
Hm	6.73	0.89	7.91	6.24	5.99
Il	0.47	0.4	0.38	0.4	0.43
Mt	-	0.37	0.07	0.37	-
Ap	-	0.07	0.12	0.14	0.02
An	-	-	-	1.91	-
Hy	-	-	-	0.07	-
Tn	-	-	-	-	0.01

The estimated averages of U and Th contents in the granitoid rocks are 4 ppm and 18 ppm respectively (Rogers and Adams, 1969). The U and Th contents in the studied aplite-pegmatite dykes range from 19 to 305 ppm with an average of 103 ppm for uranium and from 53 to 735 ppm with an average of 274 ppm for thorium. Also the dykes enriched in Nb (91.7 – 5121.9 ppm) with an average of 1485.3 ppm, Ta (8.5 – 693.5 ppm) with an average 208.1 ppm, Zr (623 – 16950.4 ppm) with an average 3967 ppm, Y (81.9 – 1915.8 ppm) with an average 569.5 ppm and Σ REE (143.36 – 2271.49 ppm) with an average 1143.2 ppm.

High field strength cations include the lanthanides Sc and Y, and Th, U, Pb, Zr, Hf, Ti, Nb and Ta. The element pairs Th and U, and, Nb and Ta, and, La and Y, and, Zr and Hf are very similar in size and charge and show very similar geochemical behaviour as shown in figure 7A-D. The low field strength large ion lithophile cations include Cs and Rb, Ba and Rb (Fig. 7E- F). The plot samples show a well developed positive correlation between those element pairs.

On the other hand, the geochemical behaviour of REE is intrinsically interesting but use of REE abundances in determining evolutionary histories of suites of rocks is even more interesting and important. The REE are used to illustrate the petrogenesis of a rock after being normalized to a common reference standard, which most commonly comprises the values for chondritic meteorites. The rare earth elements of the studied dykes are normalized to chondrite values cited in Nakamura (1974).

The chondrite-normalized REE patterns are shown in figure (8). It shows moderate to large negative Eu anomalies and a wide variation in Σ REE in addition to the tetrad effect. Highly fractionated magma is suggested for the studied dykes since the pattern has moderate to large negative Eu (Henderson 1984). It is conformable with the field relation since the aplite-pegmatite dykes are late stage magmatism.

The most of recent discussion about the tetrad effect focus on highly evolved igneous rocks (Bau, 1996, 1997; Pan 1997) which are often interpreted as transitional between the end – members of magmatic and high-temperature hydrothermal systems.

To determine the hypothetical tetrad effect-free REE pattern according to Irbër (1999), the corner points of the single tetrads La-Nd (and Gd-Ho) serve as a respective reference. A virtual line is drawn in between these corner points, and the mean deviation of Ce and Pr (and Tb, Dy) from this line expresses the contribution of the respective tetrad (equations 1 and 2).

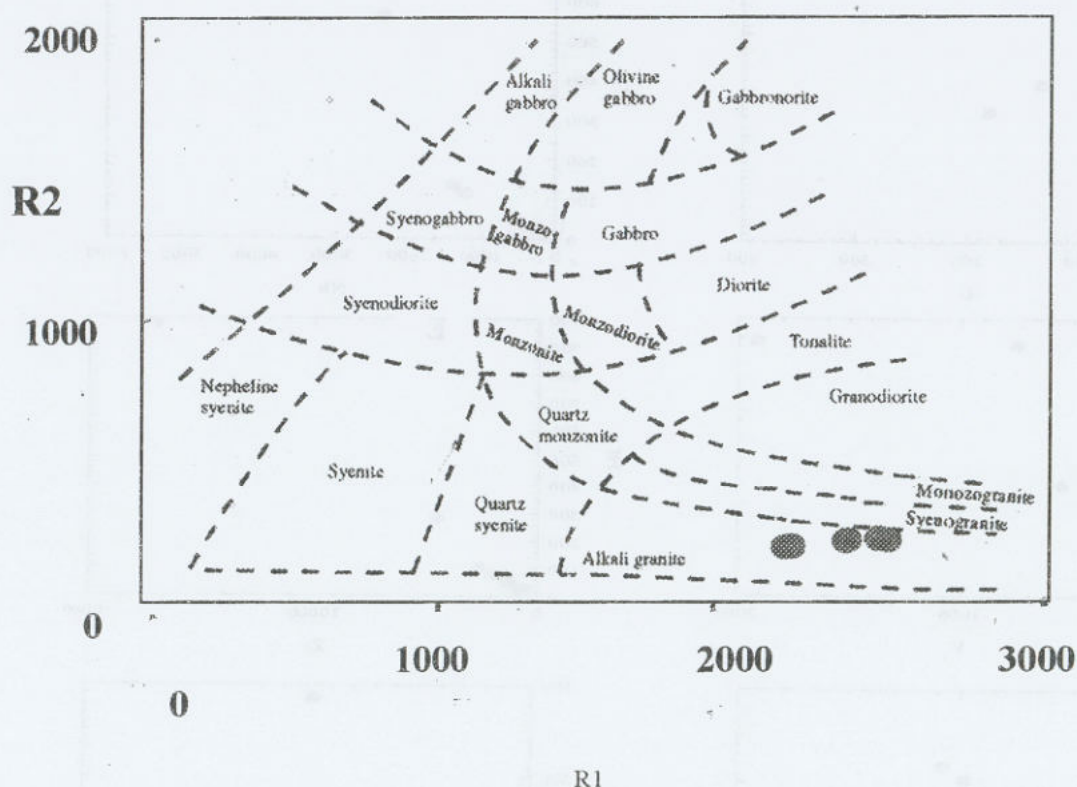


Fig. (4): Chemical classification of the studied aplite-pegmatite dykes, using R1- R2 Millicationic proportions (after De La Roche *et al.*, 1980)

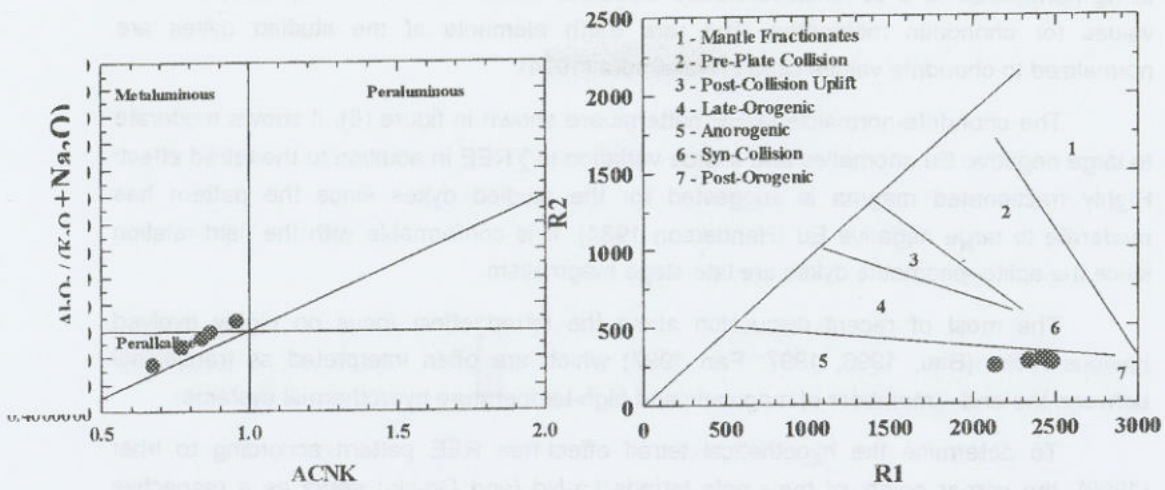


Fig.(5): Shand's index for the studied dykes

Fig.(6): Timing of emplacement of the studied dykes

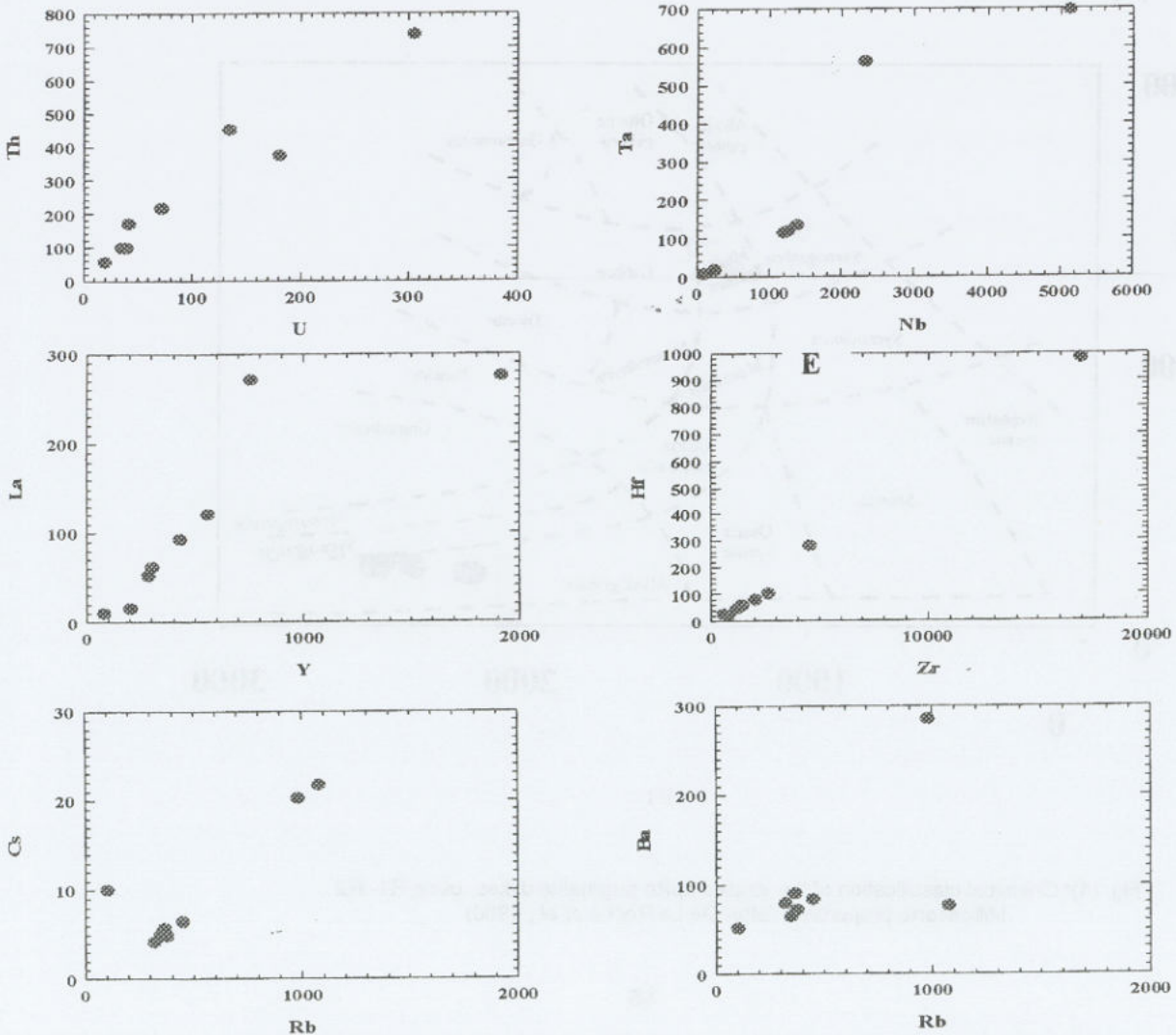


Fig. 7A – F. Variation diagrams between pairs of similar geochemical behaviour for some trace elements

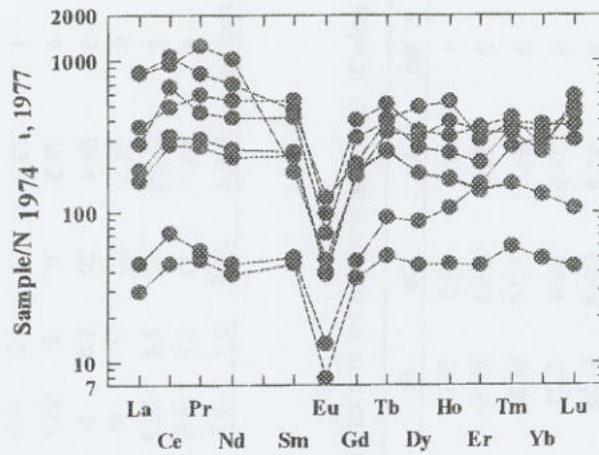


Fig. (8): Normalized REE distribution pattern for the studied dykes.

The geometric mean of both values for the first (t_1) and the third tetrad (t_3) yields the overall value of the tetrad effect (equation 3: $TE_{1,3}$).

$$t_1 = (Ce/Ce^* \times Pr/Pr^*)^{0.5} \quad (1)$$

$$t_3 = (Tb/Tb^* \times Dy/Dy^*)^{0.5} \quad (2)$$

where

$$Ce/Ce^* = Ce_{cn} / (La_{cn}^{2/3} \times Nd_{cn}^{1/3})$$

$$Pr/Pr^* = Pr_{cn} / (La_{cn}^{1/3} \times Nd_{cn}^{2/3})$$

$$Tb/Tb^* = Tb_{cn} / (Gd_{cn}^{2/3} \times Ho_{cn}^{2/3})$$

$$Dy/Dy^* = Dy_{cn} / (Gd_{cn}^{1/3} \times Ho_{cn}^{2/3})$$

$$\text{Degree of the tetrad effect} = TE_{1,3} = t_1 \times t_3^{0.5} \quad (3)$$

The calculated values of the tetrad effect ($TE_{1,3}$) range from 1.0 for a REE pattern without tetrad effect and only samples with values of $TE_{1,3} > 1.1$ are considered to show the tetrad effect.

The data of tetrad effect values for the studied aplite-pegmatite dykes are shown in Table 4. The values of tetrad effect ($TE_{1,3}$) for all samples exceed 1.1 where the values range from 1.14 to 1.31 which means that all samples have well visible tetrad effect.

Table 2: Chemical analyses for trace elements (ppm) of the studied rocks

S.No.	Ba	Be	Co	Cs	Ga	Hf	Nb	Rb	Sr	Ta	Th	U	V	W	Zr	Y	
1	90.1	18	0.6	5.7	28.1	27.2	91.7	369.8	10	14.8	8.5	53.7	19.4	<5	0.8	623	81.9
2	72.6	32	1.5	4.9	31.3	48.9	244.3	378.3	38	37	17.7	96.9	36	8	1.5	1239.1	291.9
3	79.7	31	1.2	4.2	29.3	56.7	252.7	325.2	40	26.9	18.1	98.7	39.9	8	1.7	1468.7	306.8
4	76.9	6	2	21.7	44	279.3	1384.3	1076.3	124	14.5	133.4	371.8	180.6	<5	3.3	4581.2	205
5	64.4	30	1.5	5	34.3	78.8	1203.4	348.6	50	27.6	113.9	216	72.6	10	1.7	2076.3	433.3
6	51.5	50	1	10	40	23.64	2320.34	104.5	20	52.4	561.6	169.6	42.6	<5	1.8	830.5	1915.8
7	284.5	22	7.4	20.2	36.8	983.4	5121.9	982.3	30	53.6	693.5	735.6	305.2	11	2.2	16950.4	758.5
8	83.7	39	1.9	6.5	36.5	102.4	1264.4	453.8	65	35.8	118.6	450.9	135.3	13	1.2	2699.9	563.2
Average	100.4	28.5	2.1	9.7	35	200	1485.3	504.8	47.1	32.8	208.1	274.1	103.9	10	1.7	3967	569.5

Table 3: Chemical analyses for rare earth elements (ppm) of the studied rocks

S.No.	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	ΣREE
1	9.8	39.3	5.37	24.1	9.22	0.6	10.05	2.38	15.23	3.18	10.08	1.77	10.79	1.49	143.36
2	52.2	235.5	30.32	143.5	47.4	2.95	50.8	11.75	61.54	11.64	31.4	4.55	28.28	3.57	715.4
3	61.8	273.6	34.23	162.2	50.25	3.08	51.87	11.89	61.65	11.37	31.02	4.59	28.69	3.58	789.82
4	14.9	62.4	6.21	28.6	10.1	1.01	12.89	4.3	29.41	7.45	34.4	8	66.16	10.18	296
5	92.8	576.6	51.38	265.1	85.3	5.58	85.48	18.45	89.81	16.93	47.57	10.7	76.34	15.25	1437.29
6	275.6	923.7	91.6	438.6	96.5	9.69	56.68	19.35	169.43	37.24	66.59	11.7	84.2	12.62	1961.22
7	271	811.6	139.9	646.4	37.3	3.73	47.69	15.91	108.87	27.56	77.28	9.8	54.79	19.66	2271.49
8	120.6	424.9	66.79	344.6	110.8	7.54	111.4	23.98	116.75	22	81.84	12.7	59.44	16.8	1401.2
Average	89	418.4	53.2	256.6	55.8	3.8	43.1	13.5	81.5	17.1	47.5	7.9	51	10.3	1143.2

Table 4. Theoretical values of tetrad effect for the studied rocks

Sample No.	Eu/Eu*	Ce/Ce*	Pr/Pr*	Tb/Tb*	Dy/Dy*	t ₁	t ₃	TE _{1,3}
1	0.19	1.36	1.32	1.2	1.01	1.34	1.1	1.21
2	0.18	1.47	1.3	1.31	1	1.38	1.14	1.26
3	0.18	1.45	1.27	1.32	1	1.37	1.16	1.26
4	0.27	1.54	1.18	1.39	1	1.35	1.19	1.26
5	0.2	2	1.2	1.28	0.96	1.55	1.11	1.31
6	0.4	1.31	1.07	1.36	1.2	1.18	1.29	1.23
7	0.27	1.02	1.27	1.4	1.02	1.14	1.19	1.16
8	0.2	1.13	1.2	1.3	0.96	1.17	1.11	1.14

Mineralogy

Methodology

Five samples were selected for mineralogical studies. These samples were crushed and sieved to the fraction between (-500 μ m and 0.063 mm grain size). The obtained fractions were separated by heavy liquids technique and iso-dynamic magnetic separator at 0.5A°, 0.7A°, 1.0A°, 1.2A° and 1.5A°. The obtained heavy fractions were hand picked with binocular microscope and then analyzed by SEM attached with EDAX unit. The analyses were carried out at the Central Laboratories of Nuclear Materials Authority, Cairo, Egypt.

SEM technique analyses reveal the presence of some minerals such as hydrothermal zircon which recorded for the first time in Sinai as far as the author is aware, columbite minerals group and Tornebohmite-(Ce) minerals.

Hydrothermal zircon

The term "hydrothermal zircon" is an imprecise term that may in a general way be used to refer to zircon crystallized from or altered by an aqueous fluid. In detail, however, there are three distinct mechanisms forming hydrothermal zircon although the zircon they produce may share trace-element characteristics:

(i) Dissolution-reprecipitation, documented for high-pressure/low-temperature meta-igneous rocks (Tomaschek *et al.*, 2003).

(ii) Ion-exchange between low-temperature (120 to 200°C) aqueous fluid and amorphized (metamict) zircon and structural recovery as described experimentally (Geisler *et al.*, 2003a) and for zircon from the Eastern Desert, Egypt (Geisler *et al.*, 2003b).

(iii) Direct crystallization by a (locally?) zircon-saturated aqueous fluid, as described by Hoskin (2005).

The studied hydrothermal zircon is formed by direct crystallization from zircon-saturated aqueous fluid. It often has a "spongy," pitted appearance. Backscatter-electron imaging shows that the hydrothermal zircon (both aggregates and single crystals) to be unzoned with a slightly brighter response (i.e., black in gray-scale imaging). The hydrothermal zircon occurs usually as aggregates and has heterogeneous composition. Three grains are studied as

profile from the bright colour to dark colour to reveal the difference in composition in one grain. The EDAX results are shown in figs. 9 - 11. The EDAX results show general enrichment of zircon with trace elements such as U, Th and Hf, and rare earth elements (REE). It is noted that the Zr decreases with increasing of Th and U in the same grain (Figs.9-11). The hydrothermal zircon is recorded for the first time in Sinai, Egypt as far as the author is aware.

Columbite group

The columbite group of minerals has the general formula AB_2O_6 , in which, most typically, A represents Fe^{+2} , Mn, and Mg, and B represents Nb and Ta. These minerals are associated with rare-earth pegmatites and are also known from placer deposits. Most of the compounds probably formed initially with U^{4+} , most probably as a coupled substitution $Ca^{2+} + U^{4+}$ for a trivalent ion. Those minerals which contain radioactive elements are usually metamict and require heating to develop crystallinity (Smith, 1984).

Samarskite (Y, Ce, U,Ca, Fe)(Nb, Ta, Ti)₂O₆

Samarskite is common enough and locally abundant enough that it has potential to be a valuable resource for its rare earth metals. It is usually associated with quartz, feldspars, tantalate and rare earth minerals. It is dark black to dark brown colour and has subconchoidal fracture. The EDAX analyses (Fig. 12)

Ferrocolumbite mineral

It has formula $(Fe^{2+}Nb_2O_6)$. The EDAX analyses reveal the presence of ferrocolumbite mineral (Fig. 13). Ferrocolumbite minerals are characterized by dark pitchy to dark brown colour with conchoidal fracture.

Tornebohmite-(Ce) minerals

It has formula $(Ce,La)_2Al(SiO_4)_2(OH)$. It is important resource for REE (Fig. 14).

Conclusion

The detailed field studies reveal the presence of NE-SW, E-W, N-S and NW-SE trending alkaline pegmatite-aplite dyke system hosted by alkaline granite at north Ras Mohammed area, south Sinai, Egypt. It consists of brown-red dykes up to several kilometers long and 0.4 to 1m wide. The dykes are fine to medium-grained rocks with granular texture and are mainly composed of K-feldspar as microperthite, quartz, riebeckite and plagioclase. The dykes have anomalous radioactivity up to 305 ppm U due to the presence of the uranium mineral samarskite and hydrothermal zircon. The dykes are characterized by the presence of hydrothermal zircon (enriched with U, Th and trace elements) in addition to U (19 to 305 ppm), Th (53 to 735 ppm), Nb (91.7 to 5121.9 ppm), Ta (8.5 to 693.5 ppm), Y (81.9 to 1915.8 ppm), Zr (623 to 16950.4 ppm), La (9.8 to 275.6 ppm), Ce (39.3 to 923.7 ppm) and Σ REE (143.36 to 2271.49 ppm). The distribution of the trace elements in the aplite-pegmatite dykes can be understood on basis of the behaviour of these elements during the course of magma evolution. When the earth's mantle is melted, trace elements display a preference either for the melt phase or the solid mineral phase.

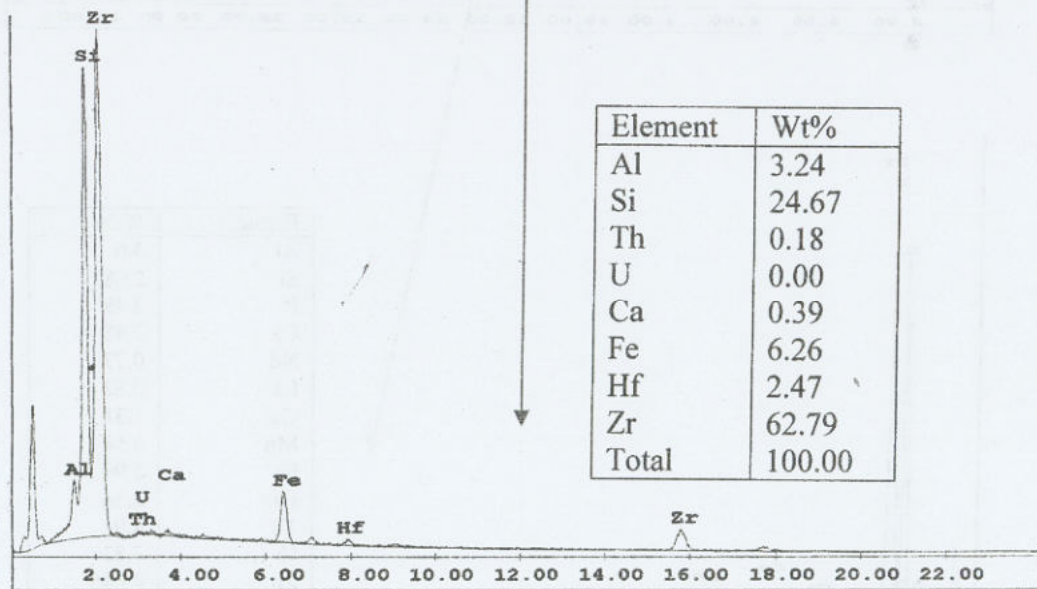
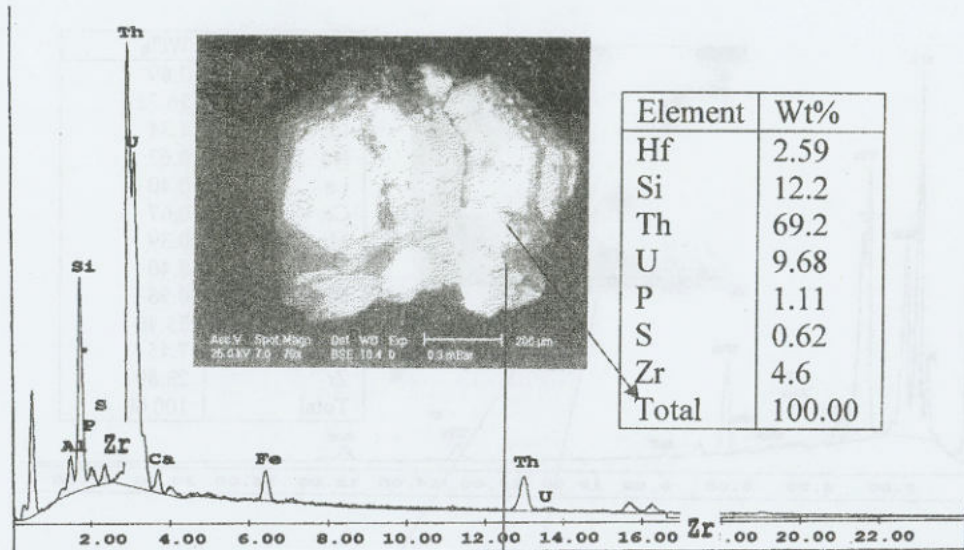


Fig.(9): SEM-EDAX semiquantitative compositional data for hydrothermal zircon

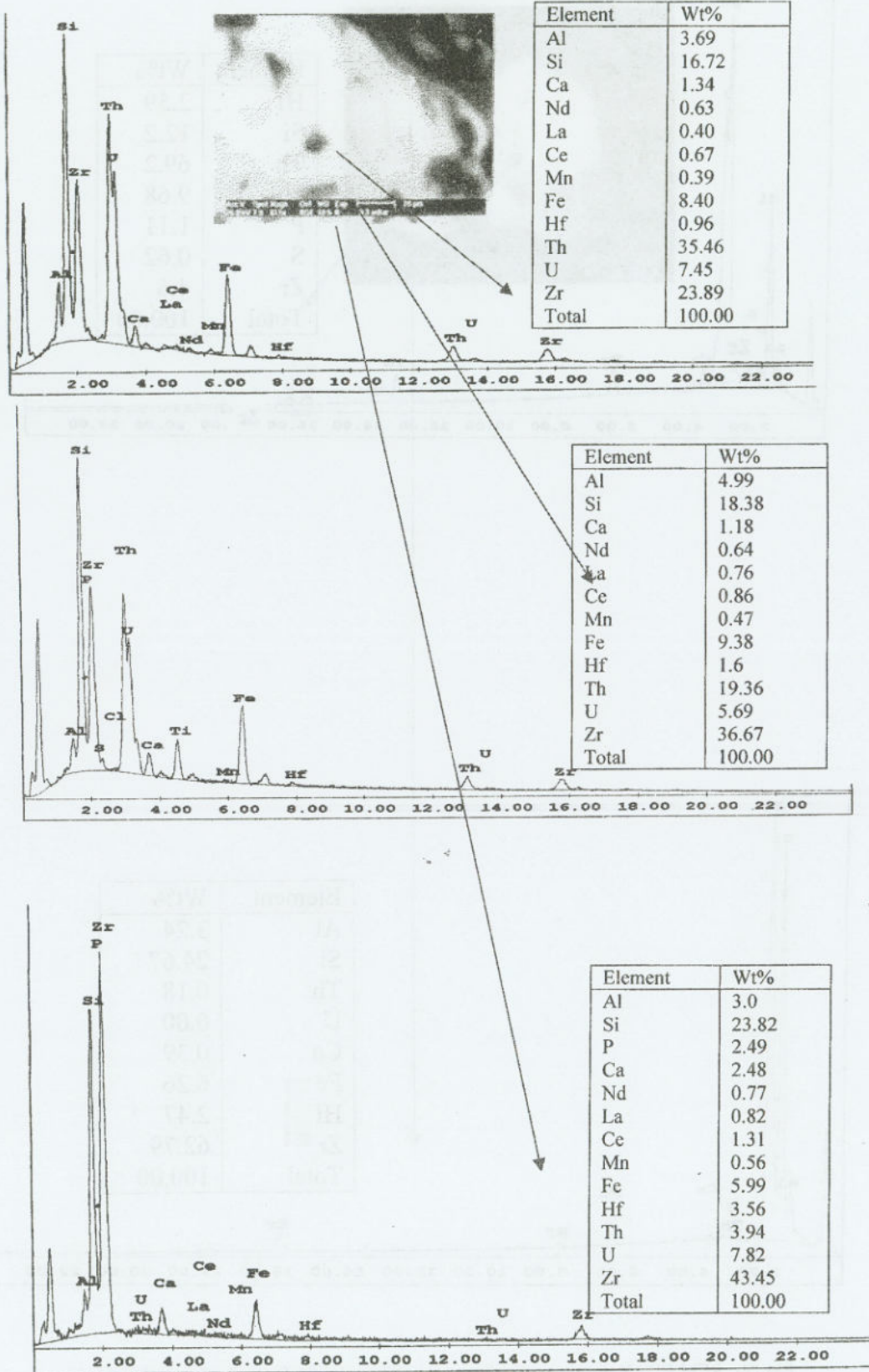


Fig. (10): SEM-EDAX semiquantitative compositional data for hydrothermal zircon mineral

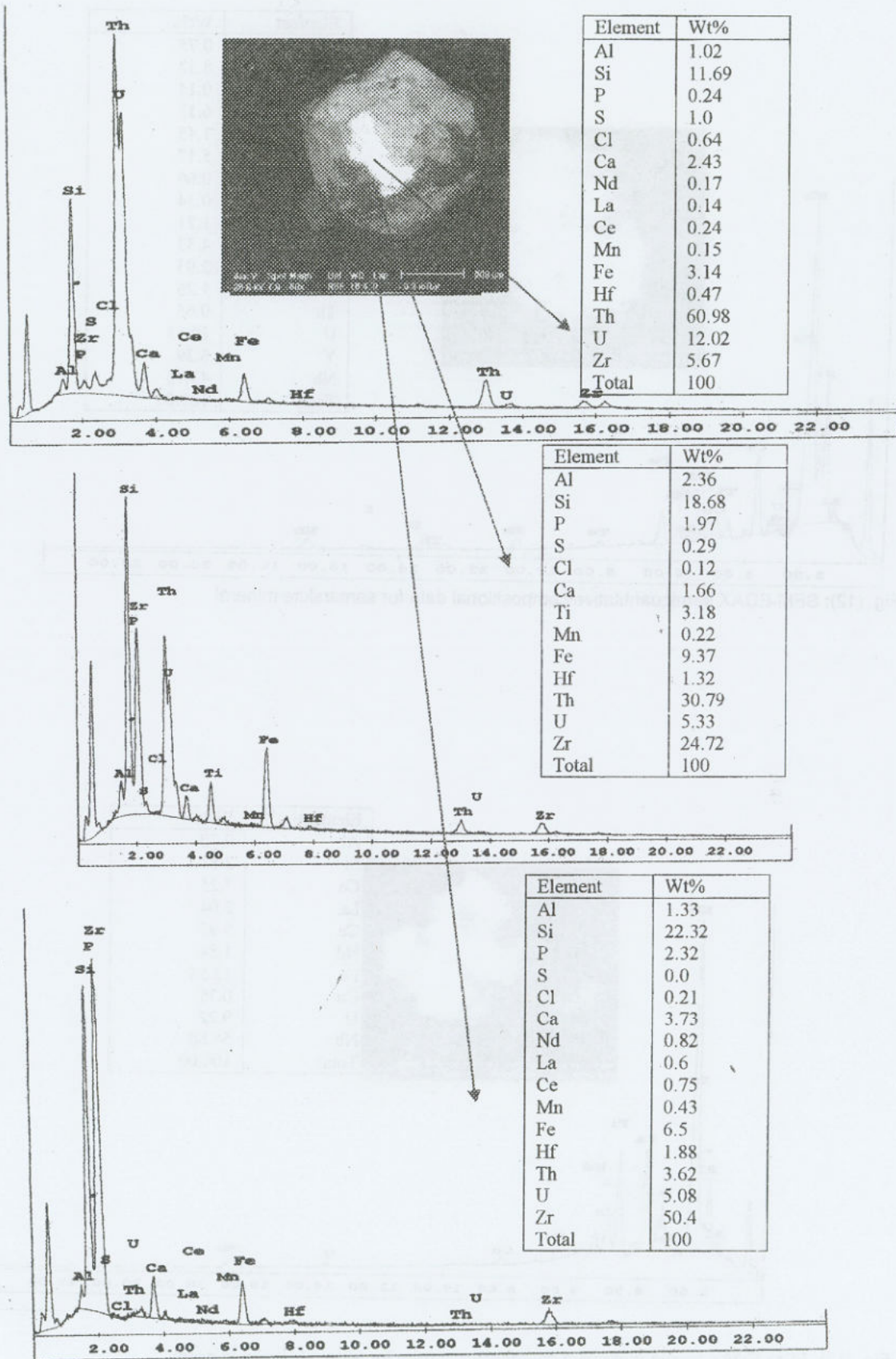


Fig. (11): SEM-EDAX semiquantitative compositional data for hydrothermal zircon

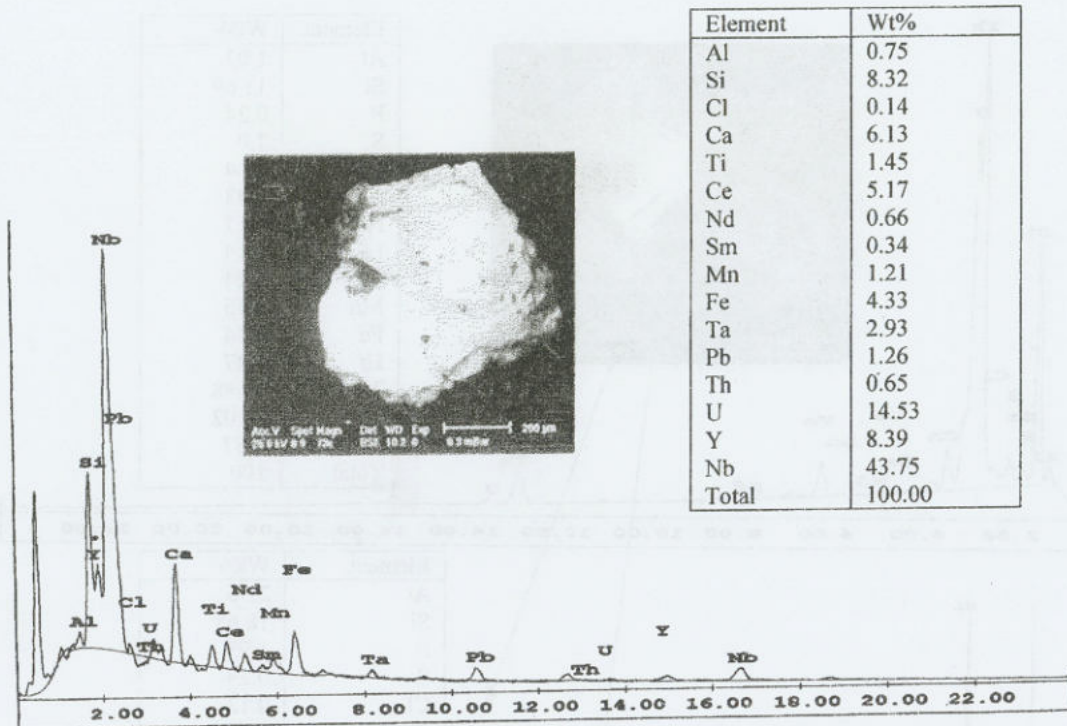


Fig. (12): SEM-EDAX semiquantitative compositional data for samarskite mineral

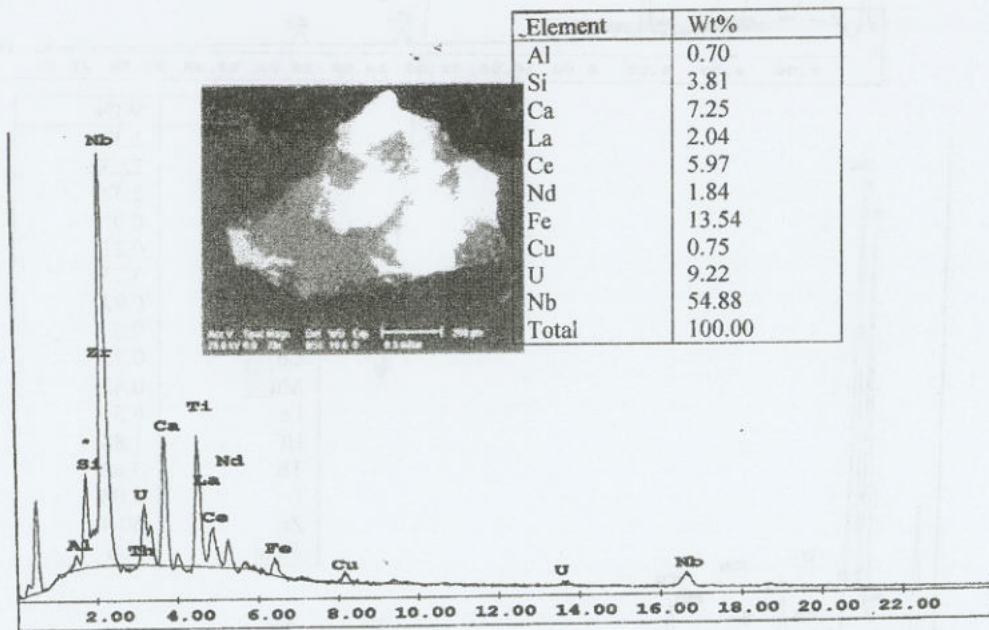


Fig. (13): SEM-EDAX semiquantitative compositional data for ferrocolumbite mineral

Trace elements whose preference is the mineral phase are described as compatible, whereas elements whose preference is the melt are described as incompatible-i.e. they are incompatible in the mineral structure and will leave at the first available opportunity. Incompatible elements have also been called hydromagmatophile, a term first introduced by Treuil and Varet (1973).

The scan electron microscope studies reveal the presence of hydrothermal zircon, columbite mineral group and Tornebohmite-(Ce) minerals.

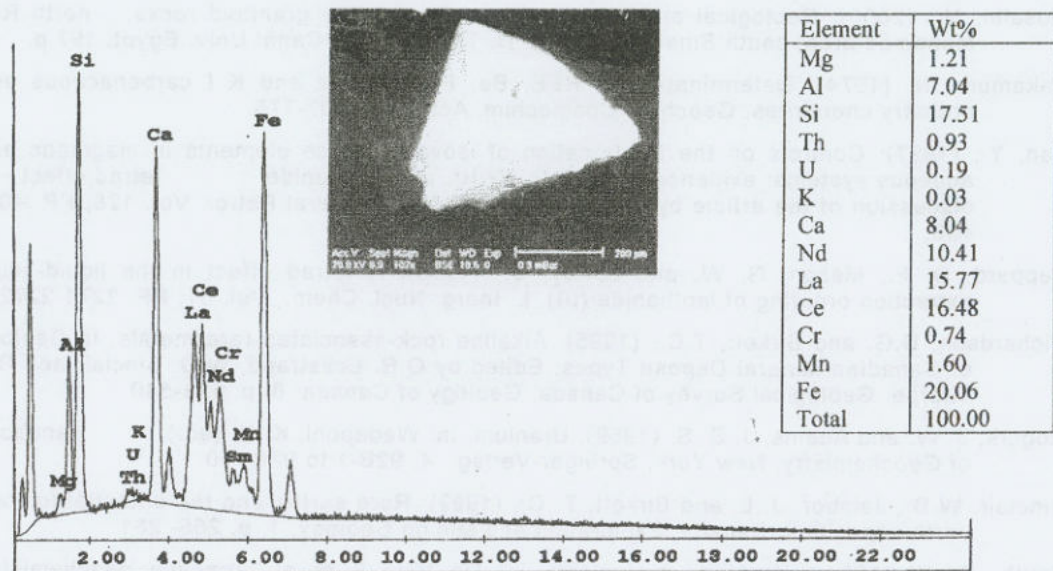


Fig. (14): SEM-EDAX semiquantitative compositional data for Tornebohmite-(Ce)

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تمعدنات الزيركون الحرمانى و العناصر الارضية النادرة فى سدود الابليت و البجمانيت بمنطقة رأس محمد، جنوب سيناء، مصر

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لقد كشفت الدراسات الحقلية التفصيلية لمنطقة شمال رأس محمد جنوب سيناء النقاب عن وجود مجموعة فواطع من نوع الأبلت-بجمانيت ذات الاتجاه شمال شرق- جنوب غرب و شرق-غرب و شمال-جنوب والتي تتميز بلونها البنى المشرب بالحمر وطولها الذى يبلغ عدة كيلومترات وعرضها الذى يتراوح ما بين ٤٠ سم الى مترا واحدا. وقد وجد أن هذه الفواطع تحتوى على نسبة من اليورانيوم تقدر بحوالى ٢٠٥ جزء فى المليون ويحتوى أيضا على نسبة من العناصر الشحيحة والعناصر الأرضية النادرة. عندما أجريت الدراسات بواسطة الميكروسكوب المساح الألكترونى على بعض العينات وجد أنها تحتوى على معدن الزيركون ذات الأصل الحرمانى، الذى تم اكتشافه لأول مرة فى سيناء بالإضافة الى المعادن الحاملة لليورانيوم والعناصر الأرضية النادرة مثل معادن السمارسكيت والكولمبيت والتورنيوهيميت.