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TRACE ELEMENTS MOBILITY AND PARTITIONING IN ZIRCON OF GABAL BAB EL-MEKHANIQ GRANITES, NORTH EASTERN DESERT, EGYPT

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ABSTRACT

More attention has been paid to trace elements in zircons, because they carry important information such as trace elements mobility and partitioning. The studied zircons were separated from both older and younger granite of G. Bab El-Mekhaniq, south of Wadi El Atrash, Northeastern Desert, Egypt.

Electron microprobe analyses show that zircons from younger granites contain higher contents of Al_2O_3, Fe_2O_3, ThO_2, UO_2, HfO_2 and CaO and lower values of ZrO_2 compared to zircons from older granitoids.

Two geochemical populations of zircons of the younger granites are recognized. Zircons of population (1) are higher in HfO_2 content than zircons of population (2), which are higher in UO_2 + ThO_2 + REE contents. The replacement of Zr by tetravalent cations (Th, U and Hf) is interpreted regarding the trace elements mobility and partitioning in zircons of the most evolved granites. The enrichment of LREEs may be due to subsets of analyses of population 2 zircons that appear to result from incorporation of mineral inclusions in the analyzed volume.

INTRODUCTION

Zircon is characterized by its high resistance to chemical weathering, metasomatic alteration, and resistance to high-temperature diffusive re-equilibration (Watson, 1996). According to these unique properties, zircon is used as a crude thermometer for crystallization conditions and as petrogenetic indicator (Pupin, 1980; Vavra, 1994; Hanchar and Westrenen,2007; Kempe et al., 2004; Watson et al., 2005). Moreover, zircon is widely used in geochronological studies (Parrish and Noble, 2003, Jackson et al.,2004; Soba et al., 2007; and Simon and Nigel, 2007).

Many studies have been devoted to zircon composition in a wide variety of rocks (Pupin 2000; Caironi et al., 2000; Rubatto et al., 2001; Hoskin and Schaltegger 2003; Fedo et al. 2003; and Belousova et al., 2002 and 2006). The composition of zircon varies widely owing to the substitution of Zr by Hf, Th and U, however it often incorporates wide range of minor and trace amounts of lithophile elements within its crystal structure such as Sc, Y, REE, Ti, Nb, Ta, V, and P (Hanshar and Westrenen, 2007). Trace elements in zircons, extracted from different rock types demonstrate that zircons may record geochemical fingerprints from their parent magma and are often the only remaining witnesses of ancient magmatic events (Belousova et al., 2002; Finch and Hanchar, (2003); Watson and Harrison, (2005); Cavosie, et al., (2004). Recently Hancher and Westrenen, (2007) indicated that natural zircon crystals incorporate REE into their structure at concentrations determined by the pressure, temperature and composition of their growth environment. Changes in zircon composition, is recently been used as a function of changes in degree of fractional crystallization of magma. This function is the goal of the present work by studying the trace elements mobility and partitioning in zircon grains from granites with variable degrees of fractionation in G. Bab El-Mekhaniq South of Wadi El Atrash. These granites are located at Gabal (G.) Bab El-Mekhaniq area, Northeastern desert which lies between Latitudes 26^0 50'- 27^0 00' N and longitudes 33^0 00'- 33^0 7'E (Fig.1).

The rock units exposed in the area are metavolcanics (oldest), older granitoids, and younger granites (youngest). Study of granites was included in many previous studies Sabet et al., (1972), El –Gaby et al., (1988), Hassan and Hashad; (1990), and Asran et al, (2008).

The older granitoids north of Bab El-Mekheniq cover about 15 % of the total mapped area they occur as scattered masses at the northeastern and northwestern parts of the area. They intruded the metavolcanics, On the other hand they are intruded by the younger granites. Generally they are of pinkish grey color, medium to coarse-grained and massive.

The younger granites occupy the south western part of the mapped area are exposed around G.Bab El-Mekhineq. these granitic rocks are intruded . The older granites of the norhtern part of the mapped area (Bab El-Mekhineq granites) form massive masses with moderate relief, pink color, and medium to coarse grained. According to Asran et al,

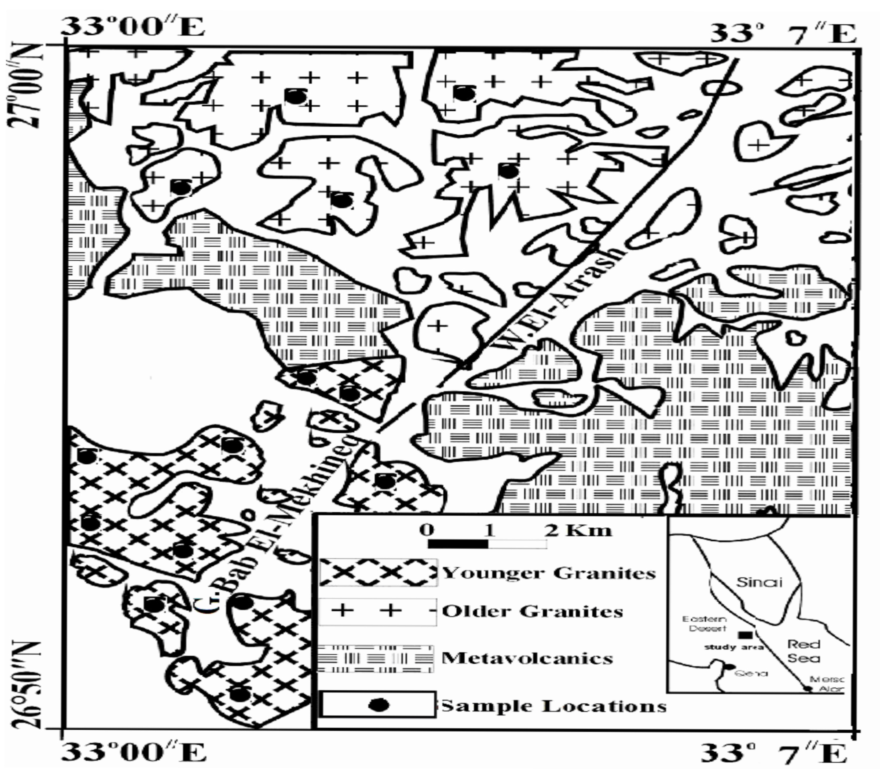

Fig.1:A part of the geologic map of Wadi El Atrash (Modified after Asran et al., 2008)

(2008), they range in composition from monzo to syeno-granite.

SAMPLING AND TECHNIQUES

Ten representative samples were collected to separate zircon grains from older granitoids and younger granites , Zircons were separated from both older and younger granites outcropping in the studied area. Each sample weighing 0.5 kg. was crushed to 60 – 150 mesh, the crushed samples were sieved using two standard screens with aperture diameter of 0.1 - 0.5 mm, each separated grain size fraction was washed by distilled water and dried by using an oven at about 90 °C, The heavy concentrates (containing zircon) were separated by bromoform (sp. gr. 2.85 g/cm^3) and methylene iodide (sp. gr. 3.3 g/cm^3). Then heavy fractions containing zircon were washed by acetone and distilled water and dried. Magnetite was separated from heavy fraction using hand magnet covered by a thin plastic film.

Zircon crystals were examined by optical microscopy and scanning electron microscope (SEM), then all zircons were prepared as round epoxy mounts and polished using 0.05 µm grit. The grains were specifically cast with their polished surfaces parallel to their crystallographic *C*-axes. Analytical sessions, a second set of images was taken of the zircons in order to characterize the electron probe analysis locations. However, this study we emphasize that trace element chemistry can be used to distinguish the different types of zircon, (Cavosie et. al., 2006). Major and trace elements (Y, Ca, Al, Mg, Mn, and Fe) analysis of zircon crystals have been performed on a Cameca SX100 electron microprobe with a beam current of 40 nA and an acceleration voltage of 20 kV. All the analyses had been preformed at Curtin University, Western Australia.

RESULTS AND DISCUSSION

Genereal Features of the Studied Zircons

The separated zircons from granites of Gabal (G.) Bab El-Mekhaniq area, North Eastern Desert show the following features

1- Zircons of older granites are coloured, euhedral to subhedral prismatic with thick width and occasionally rounded.

2- The zircons separated from the younger granites comprise two populations according to colour, size and inclusions.

a- The first population is fine-grains of grayish to brown colour that containing fine needles of long prismatic zircon inclusions representing the residual zircon substance crystallized in the last stage of magma consolidation as well as apatite crystal inclusions (Fig.2).

b- The second population is coarse, range in color from colorless to dark pink cracked grains. This population is characterized with uranothorite inclusions which cause the weakness of the crystal structure due to the effect of radiations (Fig.3), as well; it is enriched in rare earth mineral, most probably of perovskite group of minerals as inclusion (Fig.4).

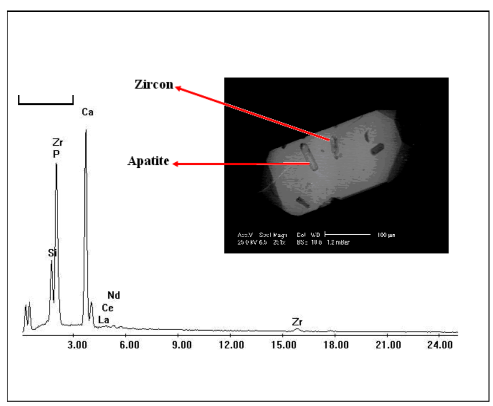

Fig.2 :SEM-EDAX semi quantitative compositional data of apatite inclusions in the studied zircons

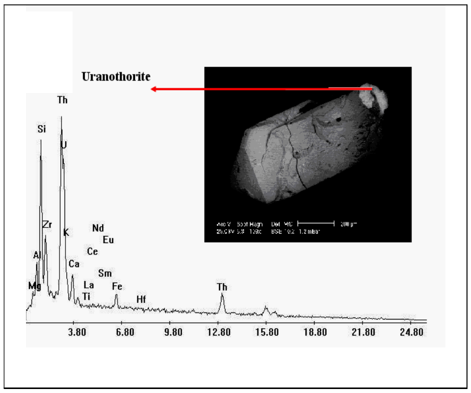

Fig.3:SEM-EDAX semi quantitative compositional data of Uranothorite inclusions in the studied zircons

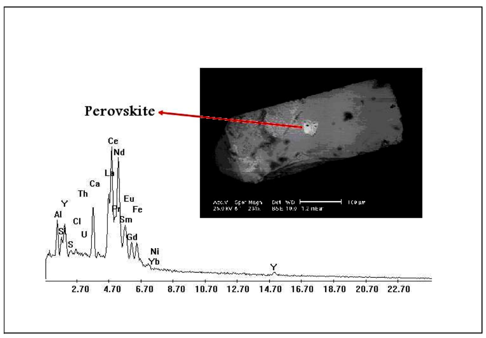

Fig.4:SEM-EDAX semi quantitative compositional data of Rare earth mineral, most probably of perovskite group of minerals in the studied zircons

Zircon Chemistry

Although zircon has a simple chemical formula $ZrSiO_4$, nevertheless its chemistry is more complicated. It posses a high ability to capture traces of REE, Y, P. and Hf as well as incorporates fine inclusions of other minerals (apatite, quartz, feldspars, REE minerals, U-Th minerals and opaques). The chemical analysis of the zircons exhibit systematic variation in Zr content. It decreases from older granite toward younger granite.

Major and trace elements contents in 24 zircons from the different granitoids of G. Bab El-Mekhaniq area are shown in Tables (1, 2 and 3). The data of the younger granites can be classified into two populations of the younger granites are (population 1 and population 2) to facilitate evaluation of REE, U, Th and Hf contents between different samples.

The zircons of the older granites and younger granite are different in the contents of Fe_2O_3, Al_2O_3, HfO_2, CaO, UO_2 and ThO_2 (tables 1,2 and3). Zircon crystals of younger granites compared to the older granitoids possess higher Al_2O_3 contents, and high CaO contents due to feldspar and apatite inclusions in the younger granites (Fig.2).

Regarding the complete solid solution series between Zircon ($ZrSiO_4$) and Hafnium ($HfSiO_4$), the concentration of Hf in Zircon

Table 1 : Chemical composition of the zircons of older granites

Oxides					Older granites
Al_2O_3	0.01	0.01	0.13	0.01	0.01
SiO_2	31.42	31.45	31.03	31.48	31.89
ZrO_2	64.29	64.75	66.38	66.14	64.10
ThO_2	0.02	0.03	0.04	0.03	0.01
UO_2	0.11	0.06	0.08	0.05	0.03
CaO	0.02	0.01	0.02	0.02	0.13
Fe_2O_3	0.18	0.15	0.45	0.12	0.17
HfO_2	1.57	1.75	1.81	1.36	1.52
∑REEs	n.d.	n.d.	n.d.	n.d.	n.d.
Total	97.63	98.22	99.95	99.21	97.74
ZrO_2/ HfO_2	40.95	37.00	36.67	48.63	42.17
ThO_2/ UO_2	0.18	0.50	0.50	0.60	0.33

Table 2 : Chemical composition of the population (1) zircons in younger granites (Wt %)

Oxides	Younger granites (population 1)								
Al_2O_3	1.22	0.02	0.01	0.89	0.79	0.85	0.02	0.86	0.22
SiO_2	28.67	37.04	37.43	29.07	32.37	32.91	35.18	32.22	28.67
ZrO_2	58.33	56.50	59.40	58.36	61.57	61.34	60.38	61.91	60.33
ThO_2	0.02	0.19	0.13	0.01	0.06	0.01	0.01	0.30	0.02
UO_2	0.65	0.58	0.23	0.32	0.43	0.26	0.02	0.51	0.65
K_2O	0.30	0.10	0.01	0.24	0.19	0.08	0.01	0.08	0.30
CaO	4.63	1.31	0.65	0.79	0.91	0.90	0.90	0.77	4.63
Fe_2O_3	4.07	1.28	0.43	10.68	0.26	0.87	0.99	0.98	4.07
HfO_2	1.45	1.78	1.42	1.79	1.88	1.60	1.55	1.37	1.45
∑REEs	0.18	0.20	0.16	0.24	0.27	0.19	0.18	0.15	0.17
Total	99.34	98.80	99.71	102.2	98.46	98.82	99.06	99.00	100.34
ZrO_2/ HfO_2	40.20	31.74	34.53	31.49	32.75	38.34	38.95	45.19	41.61
ThO_2/ UO_2	0.02	0.33	0.56	0.03	0.15	0.43	0.50	0.60	0.02

Table 3 : Chemical composition of population 2 zircons in younger granites (Wt %)

Oxides	Younger granites (population 2)									
Al_2O_3	0.83	1.43	n.d.	n.d.	n.d.	0.30	1.54	1.81	0.89	0.18
SiO_2	31.81	32.70	29.86	32.16	32.81	31.53	31.93	31.78	33.15	33.66
ZrO_2	60.87	59.62	59.75	60.57	61.92	61.40	59.24	57.38	62.55	60.64
ThO_2	0.17	0.10	0.33	0.01	0.09	0.02	0.17	0.57	0.18	0.01
UO_2	0.24	0.58	0.55	0.13	0.17	0.16	0.44	0.66	0.28	0.03
K_2O	0.05	n.d.	n.d.	0.04	0.01	0.21	0.04	0.41	0.06	0.01
CaO	1.97	1.43	1.19	1.08	0.32	1.01	2.25	2.12	0.21	0.83
Fe_2O_3	1.33	1.70	1.90	1.21	1.40	1.31	1.16	1.82	0.52	0.86
HfO_2	1.07	0.87	0.91	0.88	1.01	0.94	0.84	0.90	1.15	1.08
La_2O_3	0.28	0.27	0.47	0.01	0.09	0.60	0.16	0.31	0.21	0.37
Ce_2O3	0.28	0.39	0.63	0.34	0.14	0.75	0.74	1.03	0.68	1.11
Pr_2O_3	0.10	0.12	0.32	0.48	0.13	0.01	0.14	0.01	0.23	0.01
Nd_2O_3	0.01	0.01	0.38	0.19	0.10	0.02	0.19	0.23	0.18	0.20
Sm_2O_3	0.20	0.38	0.32	0.18	0.17	0.01	0.10	0.01	0.09	0.01
Gd_2O_3	0.40	0.05	0.55	0.40	0.13	0.01	0.01	0.05	0.01	0.01
Yb_2O_3	0.06	0.12	0.01	0.01	0.04	0.03	0.11	0.12	0.15	0.11
∑REEs	1.33	1.34	2.68	1.61	0.80	1.43	1.45	1.76	1.55	1.82
Total	99.67	99.77	97.17	97.69	98.53	98.31	99.06	99.21	100.54	99.12
ZrO_2/ HfO_2	69.97	65.52	67.90	59.97	65.87	65.32	70.52	63.76	54.39	56.15
ThO_2/ UO_2	0.71	0.17	0.60	0.08	0.53	0.13	0.39	0.86	0.64	0.33

depends on the degree of evolution of original magma (Wang et al, 2000). In the studied zircons the average of Hf concentration in zircons of the older as well as population (1 variable) is 1.60 wt% However, population (2) zircons of the younger granites are more depleted in Hf (av. 0.97 wt%), most probably due to substitution of U, Th and REE in this type of zircons.

Cation Substitutions

Zircon conforms to the general formula ABO4, where the position A represents the relatively large zirconium ion in eight-fold coordination with O, and position B represents the silicon ion in tetrahedral coordination with O. In position A, Zr can be replaced by tetravalent cations (M^{4+} = Hf, Th, U), trivalent cations (M^{3+} = REE, Y, Fe), and divalent cations (M^{2+} = Ca, Fe, Mg, Mn). As summarized by Hoskin & Schaltegger (2003), there are several mechanisms of substitution, including coupled substitutions where divalent and trivalent cations may be incorporated and simple isovalent replacement of Zr by other tetravalent cations.

The divalent (M^{2+}) and trivalent (M^{3+}) cations have been considered as involved in the xenotime–zircon series, providing a mechanism of charge balance for the REE and Y in excess of P:

$$(Mg, Fe)^{2+}_{(int)} + 3(Y, REE)^{3+} + P^{5+} = 3Zr^{4+} + Si^{4+}$$

$(Al, Fe)^{+3}_{(int)} + 4(Y, REE) + P^{5+} = 4Zr^{4+} + Si^{4+}$ (Hoskin and Black 2000)

This fact reflects the inability of the Si site to incorporate enough P to charge-balance the REE^{3+} ions (Finch et al. 2001). According to Hoskin et al, (2000), a limit on the amount of $LREE^{3+}$ substitution for Zr^{4+} is due to strain at the Zr site.

On the other hand, Y and REE can be a result of one structural site replacement:

$$(Y,REE)^{3+} + (Nb,Ta)^{5+} = 2\,Zr^{4+}$$

The abundance of each Nb and Ta in zircon is typically very low, that they are not detected in the studied samples.

Evolution of Hf, U, and Th substitution in zircons of different granite types

ZrO_2/HfO_2 ratio range from 31.75 to 48.63 in zircons from older granites and population 1 of younger granites. The same ratios are ranging from 54.39 to 70.0 in zircons of population 2 of younger granites, this may be due to presence of U, Th and REE bearing inclusions. Plotting ZrO_2 against ZrO_2/HfO_2 (Fig.5) reveals a decoupling of Zr and Hf in the direction of enrichment in Hf with the degree of differentiation of the host granite (in agreement with Linnen and Keppler, 2002; and Lowery et al. 2006).

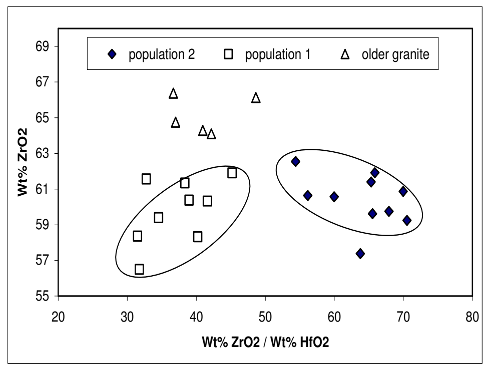

Fig.5 : ZrO_2 (Wt%) Vs ZrO_2/HfO_2

The studied zircons contain minor amounts of uranium and thorium, where U^{+4} and Th^{+4} can replace Zr^{+4} in its structure, but U^{+4} is more preferred than Th^{+4} due to its close ionic radius. The substitution of U and Th may take place in the trigonal sites of dodecahedron Zr (ZrO_8) (Frondel, 1953). In general Zr is considered as a good U- collector mineral. The average of UO_2 contents are varying from 0.02 % and 0.28 % in zircons of older granites and younger granites respectively. This variation depends on the degree of isomorphous replacement or U-Th bearing inclusions (Zhang et al, 2005).

In this work, zircons of younger granites show higher ThO_2 and UO_2 contents due to the presence of uranothorite inclusions which may indicate a relatively U and Th saturation environment (Fig. 3) and Table (3). This may be suggesting that different chemical fractionations took place within and between zircon sites both in the fluids and silicate melts participated. In population (2) high Th/U ratios are apparently associated with radiation damage of zircon grains.

As shown in Figure (6), average values of UO_2+ ThO_2 + ∑REEs and the ZrO_2 contents for zircons from younger granite show significant variation with remarkable gaps. Such gaps appear to indicate two populations of the zircons, which are referred to as population (1) and population (2), respectively. No straightforward evolution and generation separation have been identified in the older granite zircons.

Evaluation of LREE enrichment in population 2 zircons

Population (2) zircons characteristics lead to conclude that LREE enrichment in these domains dose not representate the magma composition. One outstanding question is the cause of the LREE enrichment. A subset of analyses in population (2) zircons appears to result from incorporation of subsurface mineral inclusions in the analyzed volume. It is clear that LREE - bearing mineral inclusions (e.g. perovskite) (Fig. 4) have been incorporated into the analyzed volume. The above observation indicates that several analysis of the population (2) analysis appear to have encountered sub-surface LREE-bearing inclusions.

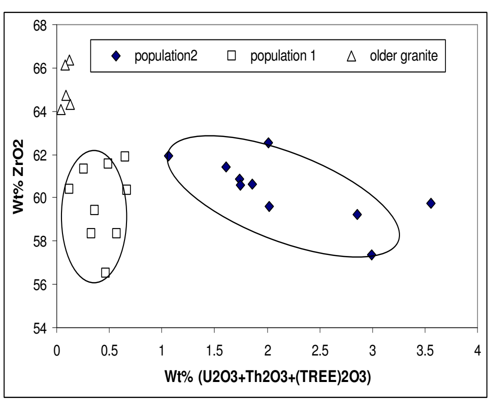

Fig. 6 : ZrO_2 (Wt%) Vs U_2O_3+Th_2O_3+TREE (Wt%)

CONCLUSION

Two populations of zircon crystals have been distinguished in younger granites owing to chemical composition. Population (1) zircons consists of euhedral and elongate crystals which is higher in HfO_2 and population (2) zircon that occurs as equant and smaller crystals with high content of UO_2, ThO_2 and REEs.

The depletion of Hf (av. 0.97 wt %) in population (2) zircons may be due to cation substitution of U, Th and REE in this type of zircons

The two populations of zircons are characterized by high content of Al_2O_3, Fe_2O_3, ThO_2, UO_2, HfO_2, CaO and lower contents of Zr if compared with zircons of older granites of the studied area.

Mechanisms of substitution may be broadly correlated with the degree of magma differentiation. In the younger granites, the Zr site in zircon is preferentially occupied by (Th + U) rather than (Y + REE). Divalent cations are also progressively introduced, and the total degree of substitution is high.

Different inclusions in zircons may refer to isomorphic substitution as a possible mechanism. population (2) zircons are characterized by Th, U and LREEs saturated environment in accordance with the presence of U-Th bearing inclusions (uranothorite) and LREEs bearing mineral (most probably perovskite) which suggests an environment saturated in Th-bearing phases (Černy and Siivola, 1980). The enrichment of LREEs in population (2) zircons may appear to result from incorporation of sub-surface mineral inclusions in the analysis volume.

The substitution of U and Th may take place in the trigonal sites of zircon so the variation in U and Th contents depend on the degree of isomorphous replacement of these elements as well as presence of uranothorite inclusions which may indicate a relatively U and Th saturation environment.

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حركة واحلال العناصر الشحيحة فى الزركون
جرانيت جبل باب المخينق- شمال الصحراء الشرقية- مصر

عزة فهمي الوكيل

إن دراسة التركيب الكيميائي للزركون بجرانيت جبل باب المخانيق هو الهدف الرئيسي لهذه الدراسة ، فمعظم الاهتمامات تدفع لدراسة العناصر الشحيحة فى الزركون لذا تم التحليل بالميكروسكوب الالكترونى لعينات من الصخور القديمة والصخور الحديثة (مونزوجرانيت وسيانوجرانيت) من منطقة جبل باب المخينق – جنوب وادى الاطرش بالصحراء الشرقية.

أكد تحليل الميكروسكوب الالكترونى ان زركون الجرانيتات الحديثة يتميز بمحتوى عالى من CaO, HfO_2, UO_2, ThO_2, Fe_2O_3,Al_2O_3 وكميات قليلة من ZrO_2 بالمقارنة بالجرانيتات القديمة.

كذلك اثبتت التحاليل وجود محتوى عالى من (UO_2+ThO_2) ومحتوى اقل من (Zr/Hf) فى زركون الجرانيتات الحديثة وتم التعرف على نوعين من الزركون، يحتوى النوع الاول من الزركون على HfO_2 اعلى من النوع الثانى الذى يحتوى على محتوى عالى من UO_2+ThO_2+REEs يرجع الإثراء في والعناصر الأرضية النادرة الخفيفة إلى وجود مكتنفات من معادن حاملة لها داخل الزركون.

بدراسة نسبة Zr/Hf في الزيركون يمكن استنتاج أن الجرانيت الحديث قد نشأ في قشرة أرضية تحت درجة حرارة أقل من التي نشأ بها الجرانيت الفديم مما أدى إلى عمليات إحلال بعض العناصر محل الزركون .