# **Mineralogy and mineral chemistry of rare-metal pegmatites at Abu Rusheid granitic gneisses, South Eastern Desert, Egypt**

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#### **Abstract**

The Abu Rushied area, situated in the South Eastern Desert of Egypt is a distinctive occurrence of economically important rare-metal mineralization where the host rocks are represented by granitic gneisses. Correspondingly, mineralogical and geochemical investigation of pegmatites pockets scattered within Abu Rusheid granitic gneisses revealed the presence of Hf-zircon, ferrocolumbite and uranyl silicate minerals (uranophane and kasolite). Electron microprobe analyses revealed the presence of Nb-Ta multioxide minerals (ishikawaite, uranopyrochlore, and fergusonite), uraninite, thorite and cassiterite as numerous inclusions in the recorded Hf-zircon and ferrocolumbite minerals.

Abu Rusheid pegmatites are found as small and large bodies that occur as simple and complex (zoned) pegmatites. Abu Rusheid rare-metal pegmatites occur as steeply dipping bodies of variable size, ranging from 1 to 5 m in width and 10 to 50 m in length. The zoned pegmatites are composed of wall zone of coarser granitic gneisses, intermediated zone of K-feldspar and pocket of mica (muscovite and biotite), and core of quartz and pocket of mica with lenses of rare metals.

The zircon is of bipyramidal to typical octahedral form and short prisms. Because the zircon of the investigated Abu Rushied pegmatite frequently contains hafnium in amounts ranging between 2.31 and 11.11%, the studied zircon was designated as Hf-rich zircon. This zircon commonly exhibits a normal zoning with rims consistently higher in Hf than cores. The bright areas in the crystal either in core or rim showed a remarkable enrichment in hafnium content (8.83–11.11%) with respect to the dark zones (3.19%). The investigated ferroclumbite commonly exhibits zoning; the dark zone is low in the Ta and U but the light zone is enriched in Ta (13%) and U (1%). EMPA analyses indicate the chemical composition of ishikawaite with U ranging from 0.68 to 0.79 per formula unit. Uranopyrochlore species has dominant uranium in the A-site where it ranges from 12.72 to 16.49% with an average of 14.84%. The calculated formula of the studied fergusonite is  ${}^{\rm A}({\rm Y}_{0.303} \tilde{\Sigma}$ REE<sub>0.014</sub>  ${\rm U}_{0.135}$  Th<sub>0.063</sub> Ca<sub>0.013</sub> Pb<sub>0.006</sub> Si<sub>0.213</sub>  $\rm Zr_{0.035}~Hf_{0.048}~Fe_{0.105})_{\Sigma 0.935}~^{B} (Nb_{0.61}~Ta_{0.084}~Ti_{0.01})_{\Sigma 0.704}~O_{4}.$ 

The presence of uraninite (high Th, and REE contents) and thorite, indicates that these minerals magmatic processes and followed by hydrothermal processes which are responsible for the precipitation of Nb-Ta multioxide minerals. Uranophane and kasolite of Abu Rusheid pegmatites are most probably originated from hydrothermal alterations of the primary uraninite. Abu Rushied pegmatites are characterized by being of ZNF-type due to their marked enrichement in  $Zr$ , Nb, and F, with a typical geochemical signature:  $Zr$ , Nb  $>>Ta$ , LREE, Th, P, F. Accordingly, the mineralized Abu Rushied pegmatite can be considered as a promising target ore for its rare metal mineralization that includes mainly Nb, Ta, Y, U, and REE together with Zr, Hf, Sn and Th.

#### **Introduction**

Rare-metal mineralization is particularly and genetically associated with post – orogenic, geochemically distinctive granitoids (TISCHENDORF, 1977). Abu Rushied - Sikeit area represents a small part of the Precambrian basement of the southeastern desert and is located some 90 km southwest of Marsa Alam on the Red Sea coastal plane (Fig. 1). The studied mineralization which is restricted to psammitic gneissose type has been attributed to a metasomatic process associated with Nb-Ta mineralization (Hassan, 1973). The type and grade of the rare metal mineralization is greatly variable along the host rock. The ori-

gin of the psammitic gneiss host rock is indeed controversial where several authors considered it as a metamorphosed sedimentary unit of quartzofeldspathic composition (HASSAN, 1964; ABDELL Monem & Hurley, 1979; El Gemmizi, 1984; El-Ramly et al., 1984; Eid, 1986; Saleh, 1997; Abd El-Naby & Frisch, 2006 beside Dawood, 2010). Some authors described these rocks as gneissic granites (Ibrahim et al., 2000; Raslan, 2008), cataclastic granites (Ibrahim et al., 2007 a,b) and peralkalic granitic gneisses and cataclastic to mylonitic rocks (Ali et al., 2011). Ibrahim et al., (2000) considered it as a highly mylonitic gneissose granitic rock, ranging in composition from granodiorites to adamellites.



Fig. 1. Geological map of Abu Rushied area, South Eastern Desert, Egypt, (modified after IBRAHIM et. al., 2004).

Several rare metal mineralization occurrences including Nb-Ta, U-Th and Zr-Hf minerals have been recorded in different localities of the Eastern Desert namely; El Naga, Abu Khurg, Abu Dabbab, Noweibi and Abu Rushied localities. These mineralizations are however mainly restricted to the granite pegmatite bodies associated with the younger granite that are widely distributed in the Eastern Desert (Sayyah et al. 1993; Omar 1995, Ibrahim et al., 1996, Abdalla et al., 1998, Ibrahim, 1999, Attawiya et al., 2000, Ammar, 2001; Abdalla & El Afandy, 2003; Raslan, 2005, 2008; abd el wahed et al., 2005; Abd El Wahed et al., 2006; Abdel

WARITH et al., 2007; RASLAN et al., 2010a,b; ALI et al., 2011).

Relevant literatures indicate that Nb-Ta mineralization in Egypt has a direct connection with albite granites in the Eastern Desert (SABET  $&$ Tsogoev, 1973) Such type of granite is commonly termed "apogranite", which is believed to be a special type of metasomatic granitoid (Beus, 1982).

According to Cerny (1990) pegmatite classification, the rare earth elements (REE) subclass is characterized by Niobium-Yttrium –Fluorine family (NYF) and Zirconium-Niobium-Fluorine family (ZNF) signatures. The NYF pegmatite are

distinguished by the signature: Y, Nb>Ta, HREE, U, Th and F, meanwhile, the ZNF pegmatites can be distinguished by the signature: Zr, Nb>>Ta, Y. Th, P and F. From the exploration point of view, the post-orogenic,  $A_2$ -type granites are the most favorable sites for localization of rare metal pegmatitic mineralization of NYF affinity. These granites are characterized by mineralogical and geochemical signatures, i.e. they are transolvus, alkaline, metaluminous to mildy peraluminous with annite-siderophyllite mica as a sole mafic mineral (Abdalla & El Afandy, 2003).

Hassan (1964) studied geology and petrography of the radioactive minerals and rocks in wadi Sikait-wadi El Gemal area. Also, Hassan (1973) and Hilmy et al. (1990) studied geology, geochemistry and mineralization of radioactive columbite-bearing psammitic gneiss of wadi Abu Rusheid. EL-GEMMIZI (1984), SALEH (1997) and Ibrahim et al. (2004) studied the area and recorded several types of mineralization, such as Ta-Nb, zircon, thorite, and secondary uranium minerals. Ibrahim et al. (2007a,b) studied the geochemistry of lamprophyres hosting uranium and base-metal mineralization within the shear zones in the Abu Rusheid area. Raslan (2005) identified columbite, Hf rich zircon and dark Li-mica (zinnwaldite) from Abu Rushied mineralized gneiss. The author has further been able to identify ishikawaite from Abu Rushied mineralized gneiss for the first time in Egypt (Raslan, 2008).

Dawood (2010) studied the mineral chemistry and genesis of uranyl minerals associated with psammitic gneisses, Abu Rusheid area, and concluded that the composition and genesis of uranyl mineralization associated with Abu Rusheid gneisses provide additional information about the behavior of radionuclides in arid environments at very oxidizing conditions. Separated zircon grains from the rocks gave U/Pb age of 1770 Ma that interpreted as a probable age of the crustal area that supplied the detritus forming the original sediments (ABDEL-MONEM & HURLEY, 1979). Ali et al. (2011) studied the mineralogy and geochemistry of Nb-, Ta-, Sn-, U-, Th-, and Zr-Bearing granitic rocks from Abu Rusheid Shear Zones, and concluded that the field evidence, textural relations, and compositions of the ore minerals suggest that the main mineralizing event was magmatic (629  $+/- 5$  Ma, CHIME monazite), with later hydrothermal alteration and local remobilization of high-field-strength elements.

The aim of the present study is to identify the mineralogical and geochemical characteristics of the radioactive and economic minerals of Abu Rushied rare-metal pegmatites.

## **Geologic setting**

The tectonostratigraphic sequences of the Precambrian rocks in Abu Rushied area are arranged as follows: (1) ophiolitic mélange, consisting of ultramafic rocks and layered metagabbros with a metasedimentary matrix; (2) cataclastic rocks

are composed of protomylonites, mylonites, ultramylonites, and silicified ultramylonites, (3) mylonitic granites; and (4) kinematic granitic dykes and veins (Ibrahim et al., 2004). The metasediments are represented mainly by separated successions of highly foliated mica schist locally thrusted over the psammitic gneisses (Fig. 1). Tourmaline mineralization occurs in different parts of the metasediments either as disseminated crystal clusters or as discontinuous tourmalinite bands (Harraz & El-Sharkawy, 2001). The ophiolitic mélange represents the hanging wall of the major thrust in the study area. It comprises a metamorphosed sedimentary matrix enclosing amphibolite sheets, allochthonous serpentinite and gabbroic masses, as well as quartzitic bands. Amphibolites and metagabbros are probably related to the calc-alkaline metagabbros associated with Hafafit gneisses (EL-RAMLY et al., 1993). Abu Rusheid granitic gneisses are highly mylonitized and dissected by several shear zones mostly oriented to NW-SE directions (Fig. 1). Brecciation resulting from faulting reactivation is found in some parts along the shear zones. The psammitic gneisses show a well developed planer banding, gneissosity and folding. Lineation, defined by mineral streaking is well marked on the foliation surfaces (Hassan, 1973). Small size quartz and pegmatitic veins are common and seem to be developed from the gneiss through mobilization and crystallization as they fade out into the gneiss with no sharp contacts (Hassan, 1973).

The Abu Rusheid pegmatites of granitic gneisses were surveyed on a 5x20 m grid. Many vugs are formed in the studied area (especially close to the contact of metasediments and two mica granites) as a result of leaching processes that were filled by pegmatites (Ibrahim et al., 2004). Greisenization is common in contact zones with other rocks (metasediments and two mica granites). Abu Rusheid pegmatites are very coarse to coarse in size and pink to dark redish in colour; they Crop Out Along The Eastern Flank Of Wadi Abu Rusheid Around Khour-Abalea As Elongated Scattered Bodies (Fig. 1).

Abu Rusheid rare-metal pegmatites are commonly found witinin the granitic gneisses of the studied area. They are found as small and large bodies and occur as simple and complex pegmatites. Abu Rusheid pegmatites occur as steeply dipping bodies of variable size, ranging from 1 to 5 m in width and 10 to 50 m in length. The zoned pegmatites are composed of wall zone of coarser granitic gneisses, intermediated zone of K-feldspar and pocket of mica (muscovite and biotite), and core of quartz and pocket of mica with lenses of rare metals (Fig. 2). These rocks are very coarse grained, mainly observed in the granitic gneisses near the contact with ophiolitic mélange and two mica granites. Mineralogically, they are mainly composed of intergrowth of K-feldspar, milky quartz, plagioclase (albite) together with small pockets of mica (muscovite and biotite). Field radiometric measurements indicate that radioactivity of Abu Rusheid simple pegmatites are



Fig. 2. Sketch showing the pegmatites of Abu Rusheid area, South Eastern Desert, Egypt.

more than twice that of their enclosing country rocks (granitic gneisses). These pegmatites are also found as zoned bodies ranging from 5 to 10 m in width and extend to 50 to 100 m in length, and trending in a NNW-SSE direction.

#### **Sampling and techniques**

Twenty mineralized pegmatite samples were collected from the study area and prepared for mineralogical and geochemical investigations. 20 polished thin sections were prepared and studied under reflected and transmitted light in order to determine mineral association and mineral chemistry. In addition, representative bulk composite sample of Abu Rushied pegmatites was subjected to various mineral separation steps: disintegration (crushing, grinding), desliming, sieving, followed by heavy liquid separation using bromoform (specific gravity. 2.85). The heavy minerals were analyzed using Environmental Scanning Electron Microscope (ESEM) supported by energy disperssive spectrometer (EDS) unit (model Philips XL 30 ESEM) at the laboratory of the Nuclear Materials Authority (NMA). The instrument enables analyses of wet, oily, dirty, nonconductive and rough samples in their natural state without modification or preparation. However, the application is limited to qualitative and semiquatntitative determinations. The analytical conditions were 25-30 kV accelerating voltages, 1-2 micron beam diameter and 60-120 second counting times. Minimum detectable weight concentration of elements from 0.1 to 1 wt % was obtained. Precision was well below 1 %. The relative accuracy of quantitative result was 2-10 % for elements Z>9 (F), and 10-20 % for the light elements B, C, N, O and F.

Also, polished thin-sections of some mineral grain varieties were analyzed using a Field Emission Scanning Electron Microscope (JEOL 6335F) at the Particle Engineering Research Center (PERC), University of Florida, USA. This instru-

ment is fitted with an Oxford Energy Dispersive X-ray Spectrometer (EDS) for elemental analysis of micro areas, a backscattered electron detector that allows compositional analysis, and a cathode luminescence detector that can image complex characteristic-visible spectra for detailed molecular structure information. The applied analytical conditions involved 0.5 to 30 accelerating voltage, 1.5 nm (at 15 kV) / 5.0 nm (at 1.0 kV). Imaging modes are secondary electron imaging (SEI) and backscatter electron imaging (BSI). This instrument can be available for operation from remote locations, X-ray microanalysis of small areas, lines scans of relative concentrations for multiple elements and for X-ray maps of relative concentrations for multiple elements.

Backscattered electron images were collected with the scanning electron microscope-energy dispersive spectrometry (BSE) (model JEOL 6400 SEM) at the Microscopy and Microanalyses Facility, University of New Brunswick (UNB), Canada. Mineral compositions were determined on the JEOL JXA-733 Superprobe; operating conditions were 15 kV, with a beam current of 50 nA and peak counting times 30 second for all elements. Standards used in this study were, as follows: jadeite, kaersutite, quartz, and apatite (for Na, Al, Si, P, and Ca, respectively),  $SrTiO<sub>3</sub>$  (for Ti),  $CaF<sub>2</sub>$  (for F), Fe, Nb, Hf, Ta, Sn, Th, and U metals (for Fe, Nb, Hf, Ta, Sn, Th, and U, respectively), YAG (for Y), cubic zirconia (for Zr), La-, Ce-, Nd-, Sm-, Pr-, Er-, Gd-, Eu-, Tb-, Dy-, and Yb- Al; Si-bearing glass, for (La-, Ce-, Nd-, Sm-, Pr-, Er-, Gd-, Eu-, Tb-, Dy-, and Yb-) and crocoite (for Pb).

#### **Results and discussion**

Microscopic investigation, scanning electron microscope and electron microprobe analyses have been used to determine the mineralogical and geochemical characteristics of the recorded minerals in Abu Rushied pegmatites. Mineralogical investigation of pegmatite pockets scattered within Abu Rusheid gneissose granite revealed the presence of Hf-zircon, ferrocolumbite and uranyl silicate minerals (uranophane and kasolite). Hf-zircon is the most dominant mineral in the representative bulk composite sample followed by ferrocolumbite and uranyl silicate minerals. Additionally, EMPA analyses revealed the presence of Nb-Ta oxide minerals (ishikawaite, uranopyrochlore and fergusonite), uraninite, thorite and cassiterite as numerous inclusions in the recorded Hf-zircon and ferrocolumbite minerals. The detailed mineralogical and geochemical characteristics of the studied minerals showed the following.

## **Microscopic and scanning electron microscope studies**

# Zircon

A unique type of zircon occurs in the Abu Rushied radioactive pegmatites. Zircon crystals of the studied radioactive pegmatite are generally characterized by their coarse size and distinctive habit. They are commonly pale to deep brown in colour under binocular microscope and generally opaque. The most common habit is the bipyramidal form with various pyramidal faces and outgrowths. Some zircon crystals are however characterized by extremely short prisms and are more or less equidimensional and exhibiting square cross section (Figs. 3 A-D). The crystals are characterized by a length/width ratio of 1:1 to 0.5:1. Some grains of the studied zircon show in most cases secondary growths, multiple growth and fused aggregations (Figs. 3E, F). The surfaces of crystals are generally rough and dull. It is referred to the pyramidal combination with extremely short prisms as mud zircon (El-Gemmizi, 1984) and to the prismatic type with no tendency to be elongated as murky type (Williams et al., 1956). In thin section, the studied zircons appear dull grayish brown and commonly show a welldeveloped euhedral shape except that one of the pyramidal faces is missing. Some crystals are characterized by sieve texture due to inclusions of other minerals such as feldspars (Figs. 3 G, H).

Several zircon crystals were subjected to semiquantitative analyses using environmental scanning electron microscope (ESEM). While the ESEM microphotographs reflect the morphological features of the investigated zircon as well as its inclusions, the EDAX analyses confirm the semiquantitative chemical composition of zircon and its inclusion respectively (Figs. 3I, J). The major elements in zircon include Zr (46.3 %), Si (18.1 %), Fe (17.2 %) and Hf (3.5 %). On the other hand, several zircon crystals have also been subjected to semiquantitative analyses using a fieldemission scanning electron microscope and the obtained SEM data (Figs. 4 A-F) show that both Zr and Si are the essential components. Other elements present in small to minor amounts include Fe, Hf, U, and Th. While the distribution of Zr, Si and Hf within the crystal is homogeneous, the distribution of uranium and thorium is actually heterogeneous.

#### Ferrocolumbite

Minerals of the columbite-tantalite group have the general formula  $AB_2O_6$ , with the A site occupied by Fe, Mn and a smaller quantity of Mg, Na and trivalent ions, and the B site occupied by Nb, Ta and small amounts of Ti and W. The main trends known from the literature are the isovalent substitutions  $\mathrm{Fe} \leftrightarrow \mathrm{Mn}$  in the A site, and  $Nb \leftrightarrow Ta$  in the B site, with corresponding end members ferrocolumbite, manganocolumbite, ferrotantalite and manganotantalite (ERCIT,  $1994$ ; Ercit et al*.*, 1995).

Ferrocolumbite grains were detected in the studied sample of Abu Rusheid pegmatite. The grains are generally black in colour and possess a brilliant metallic luster under binocular microscope. The grains are massive, rounded to subrounded and range in size from 15 to 200  $\mu$ m. RASLAN (2005) identified ferrocolumbite grains in the mineralized Abu Rushied gneiss and revealed that the grains are usually characterized by the presence of surface cavities rich in iron. Several columbite crystals have been subjected to semiquantitative analyses using a field-emission scanning electron microscope and the obtained SEM data show that both Nb and Fe are the essential components together with minor amount of Ta, Th and Mn. SEM data revealed that Ta is actually enriched in the bright zone of the crystal. The scan line within ferrocolumbite grain and scan map confirm that the distribution of Nb, Fe and Mn is generally homogeneous with respect to Th and Ta, which is actually heterogeneous (Figs. 5A-F). According to KNORRING & HORNUNG (1961) Nb and Ta mineralization are generally associated with Hf- rich zircon; a matter, which is in agreement with the Abu Rushied mineralized pegmatites.

#### **Electron microprobe analyses**

#### Zircon

The chemical composition of the studied zircon and the microprobe spots are shown in figures (6A, C, G, H). The obtained microprobe analyses (Table 1) gave an average in wt%:  $ZrO_2$ , 60.33;  $\text{SiO}_2$ , 31.85; HfO<sub>2</sub>, 4.60; U<sub>O2</sub>, 0.185; ThO<sub>2</sub>, 0.167;  $Y_2O_3$ , 0.195; FeO, 0.199 and a total REE of 0.505 with an average sum of 98.85 wt%. The microprobe data confirm that the Hf content in the studied zircon is generally increased from the core to the rim of crystals. The bright areas in the crystal showed a remarkable enrichment in hafnium content (8.83 and 11.11%) with respect to the dark zones (3.19%). Table 1 shows chemical empirical formula that is recalculated on the basis of 4 oxygen; viz,  $(Zr_{0.94}Hf_{0.044}Th_{0.003}U_{0.007}\angle REE_{0.11}\Sigma_{1.10}$  $(Si_{0.993}P_{0.006}Al_{0.003})_{\Sigma1.002}$ .

It is actually noteworthy that the EMPA analyses revealed the presence of Nb-Ta oxide minerals (ishikawaite, uranopyrochlore, and fergusonite), uraninite, thorite and cassiterite as numerous inclusions in the studied Hf-zircon. Because the zircon of the investigated Abu Rushied pegmatite frequently contains hafnium in amounts ranging between 2.31 and 11.11 wt%, the studied zircon was designated as Hf-rich zircon according to the scheme of CORREIA NEVES et al. (1974).

The obtained microprobe analyses of zircon from Abu Rushied pegmatite were plotted in the Zr-Hf-(Y, HREE, U, Th) ternary diagram and  $ZrO<sub>2</sub>$ versus  $HfO<sub>2</sub>$  diagram. The shown trends are modified from Kempe et al. (1997). The granite box, comprising Zr-Hf ranges in granites from WEDE-POHL (1978). The letters show that all the data point plot in the magmatic field (MZ) (Figs. 7, 8).

Kempe et al. (1997) considered that both magmatic and metasomatic mechanisms or a combination of them were responsible for yielding extreme Zr/Hf fractionation and hence the formation of Hf-rich zircon.

#### Ferrocolumbite

The chemical composition of the studied ferrocolumbite and the microprobe spots are



tron microscopy photomicrographs for Abu Rushied zircon, A & B, Short to equidimentional zircon crystals with a distinctive bipyramidal form. C & D, Multiple growths of bipyramidal zircon. E, Multiple growths of bipyramidal zircon with iron inclusions. F, Zircon crystal with well developed pyramidal faces. Note the bright inclusions rich in Nb and U. G & H. Thin section images of zircon crystals with one of the pyramidal faces missing, Polarized Light. Note the inclusions of silicates. I & J, EDX analyses of zircon and its inclusions respectively.

Fig. 3. A-F, Scanning elec-

shown in figures (6 B, E). The obtained microprobe analyses (Table 2) have resulted in the following averages in wt%:  $Nb_2O_{5}$ , 68.34; Ta<sub>2</sub>O<sub>5</sub>, 9.13%; MnO, 4.06%. Minor amounts of Ti, Th, U, Y, and REE were reported as substitution in fer-

rocolumbite. The calculated empirical formula of ferrocolumbite is  $({\rm Fe}_{0.52}\ {\rm Mn}_{0.13}\ {\rm Na}_{0.002}\ {\rm U}_{0.005}\ {\rm Th}_{0.004}$  ${\rm Pb}_{0.006} \, {\rm Zr}_{0.004} \, \Sigma {\rm REF}_{0.006} )_{\Sigma 0.672} \, {\rm ^B(Nb_{1.07} \, Ta_{0.139} \, Ti_{0.017}^{\rm s})_{\Sigma 1.226}}$  $O_6$ . Zoned ferrocolumbites are found in the studied pegmatite, tantalum (13wt%) and ura-



Fig. 4. A - Zircon crystal with well developed pyramidal faces, Polarized light. B - BSE image showing scan line within zircon. C - EDX spectrum of zircon D - The corresponding scan line elemental analyses results. E - Circular diagram showing the chemical composition of the investigated zircon. F - The corresponding elemental scan map.

nium (1wt%) are enriched in the bright zone with respect to the dark zone. The microprobe analyses were plotted on the  $Fera_2O_6$  -  $FeNb_2O_6$  - Mn Nb<sub>2</sub>O<sub>6</sub> - MnTa<sub>2</sub>O<sub>6</sub> quadrilateral diagram (CERNY & ERCIT, 1985). The latter show that all the data point plot in the ferrocolumbite field (Fig. 9). EMPA analyses revealed the presence of Nb-Ta oxide minerals (ishikawaite, uranopyrochlore, and fergusonite), uraninite, and thorite as numerous inclusions in the studied ferrocolumbite.

## Uranyl silicate minerals

Uranyl silicates are the most abundant group of uranium minerals. The uranyl silicate minerals can be divided into several categories on the basis of their uranium and silicon ratios (STOHL & Smith, 1981). Three categories, with uranium to silicon ratios of 1:1, 1:3, and 2:1, are well defined and reported by STOHL  $(1974)$ ; STOHL & SMITH (1974). Kasolite and uranophane are the



Fig. 5. A - BSE microphotograph showing scan line within collumbite crystal. B - BSE image showing enlarged area within that crystal. Note the bright zone rich in Ta. C - EDX analyses of collumbite. D - Scan line elemental analyses of columbite. E - Histogram showing the chemical composition of the investigated collumbite. F - The corresponding elemental scan map.

members of the first group with uranium to silicon ratio 1:1. Kasolite is distinguished by its bright colors (canary lemon, yellow and brown of different intensities). These minerals are close in their physical properties and morphological features and characterized by their softness to crushing. However, kasolite grains, compared to other uranium secondary minerals are relatively harder (Raslan, 1996). Kasolite is generally distinguished from the other uranium silicates by its crystal habit and luster. It is a hydrated silicate of lead and hexavalent uranium and is the only uranyl silicate with lead as major cation. These grains usually occur as massive granular

forms composed of druses of rod like crystals. They are characterized by their waxy or greasy luster under binocular microscope. EPMA analyses of the kasolite (Fig. 6 F and Table 3) reflect the major elements in the mineral;  $UO<sub>2</sub>$  (50.16%), PbO (36.86%) and SiO2 (10.42%) associated with quartz, minor amounts of REE, Hf, and Y, were reported as minor elements in kasolite.

The composition of analyzed kasolite (Table 3) can be expressed in the following formula:  $(Pb_{0.374})$  $\sum \text{REE}_{0.009}$ ) <sub> $\Sigma$ 0.38</sub>O. 3(U<sub>0.853</sub>) O<sub>3</sub>.3 (Si<sub>0.322</sub>) O<sub>2</sub>.4H<sub>2</sub>O. The REEs occupy the Pb sites in the lattice.

Under binocular microscope, uranophane grains are generally massive with granular form.



Fig. 6. SEM-BSE images of A - Zoned zircon crystal and location of microprobe spots. B - Subhedral cloumbite crystal associated with prismatic zircon. C - Columbite crystal enclosed zircon D - Enlarged area in that columbite crystal with uranopyrochlore and fergusonite inclusions. E - Zoned columbite and associating fergusonite inclusion. F - Acicular crystals of uranophane and kasolite. G - Ishikawaite and uranopyrochlore inclusions in zircon. H - Thorite inclusions in zircon. I - Minute uraninite inclusions enclosed in ferrocolumbite. J - Large cassiterite crystal associating ferrocolumbite.

Their luster is dull and greasy. These grains are distinguished by their bright colors (canary to lemon yellow) with pale yellow streak and found in the form of fissures and fracture fillings (Fig. 5 D). Raslan (2009b) identified dark colored iron aniferous grains in some radioactive granite plutons in the Eastern Desert of Egypt. These grains are mainly composed of uranophane and beta-uranophane, coated and stained with limonite. Raslan (2004) remarked that the presence

of both uranophane and beta-uranophane as a mixture in some samples is attributed to the presence of both habits (massive granular and fibrous acicular crystals) as intergrown mixtures. The EPMA analyses of the crystals (Fig. 6F and Table 3) reflect the chemical composition of uranophane; these results indicate that the major elements are  $UO_2$  (75.11 %),  $SiO_2$  (15.98 %), and CaO (4.68 %). Also, minor amounts of REE, Y and K, were reported as substituents for U (Table 3). The



 $\frac{1}{2}$ from hydrothermal alterations of primary minepressed in the following formula  $Ca_{0.13}(U_{1.27}O_2)$  $2(Si_{0.463}O_3)_2$  (OH)<sub>2</sub> 5H<sub>2</sub>O. Uranophane and kasolite composition of analyzed uranophane can be exof Abu Rusheid pegmatites are mainly originated

Zircon He U+Th+Y+HREE zr 90  $\frac{80}{4}$  WA  $\frac{1}{80}$ 90  $70$   $70$   $70$ <u>A</u> Mz Hz zr He U+Th<sup>4</sup>Y+HREE Solid solid series Xenon Solid solustion series Zr<br>N  $\mathcal{U}$   $\mathcal{U}$  $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$   $\mathbb{Z}$  $\sqrt[8]{7}$   $\sqrt[8]{7}$   $\sqrt[8]{80}$  $\lambda$  $\mathcal{L}_{\mathcal{B}}\mathcal{L}_{\mathcal{B}}\mathcal{L}_{\mathcal{B}}\mathcal{L}_{\mathcal{B}}$  $\text{Zr}_{\rightarrow\text{\&}}$ ZIT WEBSTER  $\sqrt{\frac{1}{N}}$  $\sum_{\mathcal{S}} \sum_{\mathcal{S}} \mathcal{S}^{\mathcal{S}}$  $\sqrt{2}$  $\blacktriangle$ r<br>Emerg  $\mathbf{\Delta}_{70}$ 

Core • Core  $\bullet$ Mz Magmatic zircon Light Dark Hz Hydrothermal zircon  $\mathcal{L}_{\mathcal{A}}$ 

positions in rare-metal pegmatites, Eastern Desert, Egypt. Intersional micropresents and interpretative boundary that h-<br>mits the compositional gap between the two zircon series. The shown trends magmatic zircon  $(MZ)$  and hydrothermal zircon  $(HZ)$  by  $K_{\text{EMDE}}$  of al. (1997) and Appellie of al. (2009) Fig. 7. Zr, Hf, (U, Th Y, HREE) ternary diagram of zircon com-The sold line represents an interpretative boundary that li-

(HZ) by KEMPE et al. (1997) and ABDALLA et al. (2009).

that it did not deposit from the circulating  $\blacksquare$ Solution series that it did not deposit from the groundwater (Osmond et al., 1999).<br>The groundwater (Osmond et al., 1999). Granitic box Granitic box ZrO wt% **2** 70 ZrO wt% **2** 70 60 60 **I Thoritical fracionation correspounding to the Solid solution Series Zircon Hafnon I** Thoritical fracionation correspounding to the crystal faces of studied uranophane indicates

ral (uraninite-High Th). The absence of distinct



Fig. 8.  $ZrO<sub>2</sub>$  versus HfO<sub>2</sub> diagram of Zircon from rare metals pegmatites, South Eastern Desert, Egypt. The shown trends are modified from KEMPE et al. (1997) and ABDALLA et al (2009). The granite box, comprising Zr-Hf ranges in granites from WEDEPOHL (1978).



Table 3. Selected EMPA analyses of uranophane and kasolite from Abu Rusheid pegmatites, South Eastern Desert, Egypt.



# Uraninite

Uraninite is a common accessory mineral in pegmatites and peraluminous granites, and is probably the most important source of dissolved U in groundwaters emanating from weathered granitic terrains (FRONDEL, 1958; FÖRSTER, 1999). The EPMA analysis (Fig. 6 I and Table 4) was used to characterize the chemical composition of uraninite. The EPMA results indicate that the major elements in uraninite are  $UO<sub>2</sub>$ (70.00 wt%), ThO<sub>2</sub> (10.13%), and PbO (6.18 %) within elemental composition of columbite  $(Nb<sub>2</sub>O<sub>5</sub> = 5.98\%),$  Ta<sub>2</sub>O<sub>5</sub> (1.96%) and FeO (2.11%) Also, minor amounts of LREE and Y were reported as substitution in columbite. The chemical formula of the investigated uraninite is  $(U_{1.20}Pb_{0.058}Th_{0.185})_{\Sigma1.44}O_2.$ 

## Thorite

Thorite was found as numerous subhedral to anhedral inclusions in zircon, 5 to 10 µm in size (Fig. 6H). The EPMA analyses for these inclusions





**Mn/(Mn+Fe)**

Fig. 9. Chemical composition of the columbite-tantalite from rare metal pegmatites in the Abu Rusheid area, plotted on the  $FeTa_2O_6-FeNb_2O_6-MnNb_2O_6-MnTa_2O_6$  quadrilateral diagram (CERNY & ERCIT, 1985). Abu Rusheid ferroclumbite in the pegmatites is represented by the closed circles.

reflect the chemical composition of uranothorite (Table 4). These results indicate that the major elements in thorite are ThO<sub>2</sub> (73.54%),  $SiO<sub>2</sub>$  $(13.02\%)$ , U  $(0.71\%)$ ,  $Y_2O_3$   $(4.67\%)$ , and FeO (2.11). Also, minor amounts of LREE and K were reported as substituents in thorite. According to FRONDEL & CUTTITO  $(1955)$ , huttonite and thorite form hydrothermally over a temperature range (300 °C to 700 °C); the formation of huttonite is favoured by alkaline conditions and thorite by acid conditions. Several authors reported the presence of thorite inclusions in rare metal mineralization and accessory heavy minerals separated from some Egyptian pegmatites (Ali et al., 2005; Abdel Warith et al., 2007; Raslan et al., 2010a, b). Electron microprobe analysis confirmed the presence of thorite whose composition corresponds to the empirical formula:  $(Th_{1,32} U_{0,012})$  $Y_{0.121}\rangle_{\Sigma1.45}(\mathrm{Si}_{0.405}P_{0.038})_{\Sigma0.443}\mathrm{O}_4$ . Uranium, rare earths, Y, Pb and Al substitute Th sites in the crystal lattice.  $PO_4$  is known to substitute for  $SiO_4$ .

# Ishikawaite (uranium-rich samarskite)

Samarskite is a group of the Nb-Ta mineral varieties occurring in pegmatite granites and hav $_{(m+n)}$  where A represents Fe<sup>2+</sup>, Ca, REE, Y, U and Th while B represents Nb, Ta and Ti. According to Hanson et al. (1999), the complete metamict state, alteration and the broad variation of cations in A-site of these mineral varieties render their crystal structure a problematic case. Therefore, these authors have proposed a nomenclature for the samarskite group of minerals based on their classification into three species. Thus, if the REE + Y are the dominant, the name samarskite- $(REE + Y)$ should be used with the dominant of these cations as a suffix. If U + Th are the dominant, the mineral is properly named ishikawaite whereas if Ca is the dominant cation, the mineral should be named calciosamarskite. Hanson et. al. (1999) have also reported that ishikawaite and calciosamarskite are depleted in the light rare-earth elements (LREE) and enriched in the heavy rare-earth elements (HREE) together with Y. Recently, samarskite-(Yb) has been identified as a new species of the samarskite group (William et al., 2006) i.e. an Yb-dominant analog of samarskite-Y. On the other hand, samarskite-Y has also been described as a mineral with Y + REE dominant at A-site (Nickel & Mandarino,

Table 4. Selected EMPA analyses of uraninite, thorite, and ishikawaite from Abu Rusheid pegmatites, South Eastern Desert, Egypt.



ing the general formula  $A_m B_n O_2$ 

1987). Raslan et al. (2010a) identified samarskite-Y from the pegmatite bodies of Gebel Ras Baroud granite and from the surrounding wadi stream sediments (Raslan, 2009b). Finally, it has to be mentioned that Warner & Ewing (1993) have proposed that samarskite should be formulated as an  $ABO<sub>4</sub>$ . It is interesting to mention that ishikawaite with an average assay of about 50%  $Nb<sub>2</sub>O<sub>5</sub>$  and 26%  $UO<sub>2</sub>$  has been identified for the first time in Egypt in the mineralized Abu Rushied gneissose granite (Raslan, 2008). The author describes Ishikawaite as black translucent massive grains of anhedral to subhedral and granular form, which are generally characterized by a dark brown streak and by a resinous to vitreous luster (Raslan, 2008).

In the present study, ishikawaite occurs as euhedral to subhedral minute crystals with sizes ranging size from 5 to 10 µm. They are present as inclusions in columbite (Fig. 6G). They are distinguished by their bright colour in SEM-BSE images. The EPMA data for ishikawaite are represented in Table 4. These results indicate that the major elements in ishikawaite are  $UO<sub>2</sub>$  $(44.73\%), \; \mathrm{Nb}_2\mathrm{O}_5 \; (30.79\%), \; \mathrm{Ta}_2\mathrm{O}_5 \; (2.53\%), \; \mathrm{FeO}$  $(3.38\%)$ , ThO<sub>2</sub>  $(5.08\%)$ . Also, minor amounts of

LREE, and Y were reported as substitution in ishikawaite. Analytical results indicate a structural formula of  ${}^A(U_{0.74}Fe_{0.11}Y_{0.004}Ce_{0.002}Ca_{0.02})_{\Sigma 0.88}$  $B(Nb_{0.513}$  Ta<sub>0.035</sub> Ti<sub>0.06</sub>)<sub> $\Sigma$ 0.61</sub>O<sub>4</sub> for ishikawaite with U ranging from 0.68 to 0.79 per formula unit. In the meantime, the two microprobe analyses were plotted on the ternary diagram of Hanson et al. (1999), which shows the A-site occupancy of samarskite-group minerals (Fig. 10). The latter shows that all the data points plot in the ishikawaite field.

From the analytical data it is quite clear that the studied mineral reflects the chemical composition of a U-rich samarskite variety in the Abu Rushied pegmatite, which is ishikawaite as indicated by the following evidence:

- 1. Both samarskite-Y and ishikawaite have a dominant Nb in the B-site and the distinction between either variety must be based on the content of B-site occupancy. The obtained EMPA data revealed that  $Nb<sub>2</sub>O<sub>5</sub>$  is the dominant in the investigated mineral; in wt% it ranges from 30.14 to 31.44 with an average of 30.79%. Thus, the studied mineral falls actually within the compositional limits of both samarskite-Y and ishikawaite.
- 2. The samarskite group of minerals must comprise only those that have Nb > Ta and Ti in the B-site (Hanson et al., 1999), and the studied mineral contains an average Ta  $+$  Ti = 4.33%  $<$  $Nb = 30.79\%$ .
- 3. Samarskite-Y has been described as a mineral with  $Y + REE$  dominant at the A-site (NICKEL & Mandarino, 1987). According to Fleischer & Mandarino (1995), the currently accepted formula of the ishikawaite species is �(U, Fe, Y, Ca) (Nb, Ta) O4] and that ishikawaite was first described as a uranium-rich, REE-poor mineral by Kimura (1922). Also, Cerny & Er $crr$  (1989) have described ishikawaite as a probable uranium-rich variety of samarskite.



Fig. 10. Ternary diagram showing A-site occupancy of samarskite-group minerals after Hanson et al., (1999). Ishikawaite in the Abu Rusheid pegmatites is represented by the closed square.

- 4. The investigated mineral is actually rich in both uranium and thorium, where the former ranges from 43.24 to 46.22% with an average of 44.73%, whereas the latter varies from 4.64 to 5.52% with an average of 5.08%.
- 5. Hanson et al., (1999) have proposed a nomenclature for the samarskite group of minerals. They thus classified this group of minerals into three species. If  $REF+Y$  is dominant, the name samarskite-(REE + Y) should be used with the dominant of these cations as a suffix. If  $U + Th$  is dominant, the mineral is properly named ishikawaite, whereas if Ca is dominant, the mineral should be named calciosamarskite. They also reported that ishikawaite and calciosamarskite are depleted in light rare earth elements (LREE) and enriched in the heavy rare-earth element (HREE) Y. The studied Abu Rushied samarskite species contain a Y content ranging from 0.205 to 0.209% with an average of  $0.207\%$ , which reflects the enrichment of HREE.
- 6. The investigated samarskite variety separated from the Abu Rushied radioactive pegmatite is characterized by dominant U + Th, Nb > Ta + Ti and relatively rich in Y.
- 7. In summary, the studied mineral most probably falls within the compositional limits of other ishikawaites cited in the previous literature.

### Uranopyrochlore

Pyrochlore group minerals are characteristic constituents of carbonatites, phoscorites and related metasomatic rocks. These minerals show a wide compositional range with respect to A- and B-site cation substitutions. General formula can be written as  $A_{2-m} B_2 O_6 Y_{1-n} \cdot pH_2 O$ , where  $A = Na$ , Mg, K, Ca, Mn, Fe<sup>2+</sup>, Sr, Sb, Cs, Ba, REEs, Pb, Bi, Th and U;  $B = Nb$ , Ta, Ti, Zr, Sn, W, Fe<sup>3+</sup> and Al; and  $Y = F$ , OH, or O (LUMPKIN & MARIANO, 1996).

Three pyrochlore subgroups are defined, depending on the predominant cation in the B site. Niobium exceeds Ta in the pyrochlore subgroup, whereas Ta exceeds Nb in the microlite subgroup. Both pyrochlore and microlite subgroups have  $(Ta + Nb) > 2Ti$ , whereas the betafite subgroup is characterized by  $2Ti > (Ta + Nb)$ . U substitutions at the A site, and metamict pyrochlore are common. Although virtually all these minerals contain some U, only two minerals of pyrochlore group contain U as an essential constituent uranmicrolite and uranopyrochlore (HOGARTH, 1977; Lumpkin & Ewing; 1995). Atencio et al. (2010) proposed a new scheme of nomenclature for the pyrochlore subgroup, based on the ions at the A, B and Y sites. They recommended five groups based on the atomic proportions of the B atoms Nb, Ta, Sb, Ti and W. The recommended groups are pyrochlore, microlite, romeite, betafite and elsmoreite respectively.

Uranopyrochlore occurs as minute subhedral to anhedral crystals in columbite, and range in size from  $5$  to  $10 \mu m$  (Figs.  $6$  B, D, G). The EPMA analyses of the crystal reflect the major elements

in uranopyrochlore are  $Nb<sub>2</sub>O<sub>5</sub>$  (35.28%),  $Ta<sub>2</sub>O<sub>5</sub>$  $(20.03\%)$ , UO<sub>2</sub>  $(14.84)$ . Also, minor amounts of Th, Y, and LREE were reported as substitutions in pyrochlore (Table 5).

In the studied pyrochlore species, the average of Nb attains 35.28% which is much higher than the average of Ta (20.03%). The obtained EPMA data revealed that the average of Nb and Ta attains 55.31% which is much higher than the average of 2Ti (3.07%). The studied pyrochlore species has dominant uranium at the A-site where it ranges from 12.72 to 16.49% with an average of 14.84%. Therefore, the defined pyrochlore species in the present work belongs actually to the compositional limits of uranopyrochlore minerals species as specified in the literature. The chemical formula of the uranopyrochlore, as indicated from the EMPA data, is  $\rm {}^A(U_{0.243}$  Th<sub>0.01</sub> Ca<sub>0.021</sub>  ${\rm Na}_{0.002}$   ${\rm Pb}_{0.01}$  $\sum$ REE $_{0.012}$  Y<sub>0.104</sub> Fe<sub>0.07</sub> Sn<sub>0.001</sub> Mn<sub>0.001</sub>)<sub> $\Sigma$ 0.474</sub>  $B(Nb_{0.505}$  Ta<sub>0.292</sub> Si<sub>0.128</sub> Zr<sub>0.005</sub> Ti<sub>0.121</sub>)<sub>∑1..05</sub> O<sub>6</sub>. The obtained microprobe analyses were plotted on the ternary diagram of HOGARTH (1977) which shows the pyrochlore group minerals (Fig. 11). The latter shows that all the data points plot in the pyrochlore field.



Fig. 11. Ternary diagram showing the pyrochlore group minerals after Hogarth (1977). Uranopyrochlore in the Abu Rusheid pegmatites is represented by the closed square.

## Fergusonite

The fergusonite group consists of REE-bearing Nb and Ta oxides, many of which are metamict and therefore commonly poorly characterized. The structure of fergusonite group is comparable to that of samarskite group but with large A-sites. Most of these minerals are monoclinic, although orthorhombic and tetragonal unit cells arise from cation ordering. Similar to other (Y, REE, U, Th)-(Nb, Ta, Ti) oxides, fergusonite (ideal formula: YNbO4), occurs typically as an accessory component in granites (POITRASSON et al., 1998) and granitic pegmatites (ERCIT, 2005) Due to its actinide content of several weight percent, fergusonite is commonly found in a highly radiation- damaged state (Ervanne, 2004) which is accompanied by major changes of physical properties and generally lowered chemical resistance. Correspondingly, fergusonite and other Nb-Ta-Ti

oxide minerals are often affected by post-growth chemical alteration (Ewing, 1975; Ercit, 2005).

The obtained EPMA chemical analyses and SEM-BSE images (Figs.6 D & E and Table 5) indicate that this fergusonite phase is predominantly composed of Y, Nb, Ta, REE, U and Th. (Table 5). The calculated formula of the studied fergusonite is  ${\rm ^{A}(Y_{0.303}\sum\!E\rm E}_{0.014}\rm U_{0.135}\rm \ Th_{0.063}\ Ca_{0.013}\ Pb_{0.006}\ Si_{0.213}}$  $\rm Zr_{0.035}\rm\,Hf_{0.048}\rm\,Fe_{0.105})_{\Sigma 0.935}\rm\,{}^{B} (Nb_{0.61}\rm\,Ta_{0.084}\rm\,{}^{T}\!i_{0.01})_{\Sigma 0.704}\rm O_{4}.$ 

## Cassiterite

Cassiterite occurs as large anhedral crystals  $(200 \mu m)$  with commonly associating ferrocolumbite. The obtained EPMA chemical analyses indicate that Sn is the most predominant element (96.79-101.9 wt%) together with minor amounts of Ta, Nb, Ce, La, Ca, Fe and Mn. (Fig. 6 J and Table 5).

## **Conclusions**

- 1 An economically important rare-metal mineralization is recorded in the pegmatite bodies of Abu Rusheid gneissose granite, South Eastern Desert, Egypt.
- 2 Field surveys indicate that the Abu Rusheid rare-metal pegmatites occur as steeply dipping bodies of variable size, ranging from 1 to 5 m in width and 10 to 50 m in length and are also found as zoned bodies ranging from 5 to 10 m in width and extend 50 to 100 m in length, and trend in a NNW-SSE direction. They are mainly composed of intergrowth of milky quartz, K-feldspars and plagioclase (albite) together with large pockets of muscovite and biotite.
- 3 The zircon is of bipyramidal to typical octahedral form with complete absence of prism, thus the zircon crystals have a length/width ratio of 1:1-0.5-1. Because the zircon of the investigated Abu Rushied pegmatite frequently contains hafnium in amounts ranging between 2.31 and 11.11 wt%, the studied zircon was designated as Hf-rich zircon. The bright areas in the crystal either in core or rim showed a remarkable enrichment in hafnium content (8.83-11.11%) with respect to the dark zones (3.19%). Ishikawaite, uranopyrochlore, columbite and thorite are common inclusions in zircon.
- 4 The investigated ferroclumbite commonly exhibits zoning; the dark zone is low in Ta and U but the light zone is enriched in Ta (13 Wt%) and U (1 wt%). Uraninite, uranopyrochlore, fergusonite and zircon are common inclusions ferrocolumbite.
- 5 The field evidence, textural relations, and compositions of the rare-metal pegmatites suggest that the main mineralizing event was magmatic with later hydrothermal alteration and local remobilization of high-field-strength elements. In the studied pegmatites, the recorded uraninite, characterized by high-Th and REE contents together with thorite, these latter



Table 5. Selected EMPA analyses of fergusonite, uranopyrochlore and cassiterite from Abu Rusheid pegmatites, South Eastern Desert, Egypt.

minerals indicate that the minerals are formed by magmatic processes and followed by hydrothermal processes; the latter hydrothermal precipitation rich in Nb-Ta which post-dated precipitation of uranopyrochlore, ferrocolumbite and ishikawaite. Magmatic uraninite commonly contains Th and REE, whereas these elements are largely absent from hydrothermal and low- temperature sedimentary uraninite (FRONDEL, 1958). Uranophane and kasolite of Abu Rusheid pegmatites are mainly originated from hydrothermal alterations of primary mineral (uraninite-High Th).

- 6 Abu Rushied pegmatites are characterized by being of ZNF-type due to their marked enrichement in Zr, Nb, and F, with a typical geochemical signature: Zr, Nb >> Ta, LREE, Th, P, F.
- 7 The Abu Rusheid rare-metal pegmatites are actually considered a promising ore material for its rare-metal mineralizations that include mainly Nb, Ta, Y, U, Th, Sn, Zr, Hf, and REE (especially HREE).

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