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Radiogenic Isotope Fractionation as an Indication for Uranium Mobility in the Granites of El Shallal Area, West Central Sinai, Egypt

Yehia H. Dawood*

Fac. of Earth Sciences, King Abdulaziz Univ., Jeddah, KSA
yhdawood@yhaoo.com

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Abstract, The granitic rocks at Wadi El Shallal area are categorized into metaluminous hornblende-biotite granites and peraluminous two-mica granites. The chemical analyses indicate that the two-mica granites are slightly altered and characterized by high uranium and thorium contents (averages 20.27 and 27.12 ppm, respectively). Compared to the hornblende-biotite granites, the $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ activity ratios of the two-mica granites display values that depart significantly from equilibrium. The sample plots of the two granitic types are distributed in a trend reflecting more ^{234}U loss from the two-mica granites. Leaching experiments confirmed that the area around mineral grain boundaries are more enriched in ^{234}U relative to its parent ^{238}U as a result of α-recoil process that displaces the daughter nuclei outside the grain surface. This daughter (^{234}U) is ejected from the grain into the open spaces and therefore, subjected to leaching by fluids. The observed uranium series disequilibrium in the granitic rocks from El Shallal area reflects isotopic fractionation and a recent migration of uranium in the two-mica granites. The high uranium contents and U-Th isotopic composition of the two-mica granites fairly suggest that these rocks are probably the source for uranyl mineralization of the nearby Um Bogma Formation reflecting a history of leaching, mobilization and precipitation by meteoric and groundwater. However, more intensive investigations for uranium

*Dept. Geology, Fac. Science, Ain Shams Univ., Cairo, Egypt.

series disequilibrium of local groundwater and uranyl mineralization of Um Bogma Formation are recommended.

Keywords: U-series disequilibrium; Activity ratios; $^{234}U/^{238}U$; $^{230}Th/^{234}U$; Uranium mobility; Granite; Sinai.

Introduction

Uranium-series disequilibrium has been widely used to study Earth's surface processes (*e.g.* Rosholt, 1982; Osmond *et al.*, 1999; Porcelli and Swarzenski, 2003; Swarzenski *et al.*, 2003 and Dosseto *et al.*, 2008). Generally, the weathered rocks deviate from secular equilibrium due to the differences in radionuclides mobility during weathering processes. This relative mobility is believed to be $^{234}U > ^{238}U > ^{230}Th$, and consequently the weathered rocks are estimated to have $^{234}U/^{238}U<1$ and $^{230}Th/^{238}U>1$ (Chabaux *et al.*, 2003 and Dosseto *et al.*, 2008). The extent of disequilibrium depends on both the intensity and age of weathering processes. For example, $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ activity ratios can be used to record weathering events up to 1 Ma and 300 ka old, respectively, because ^{234}U and ^{230}Th, the daughter nuclides of each system, have half-lives of 245 and 75 ka, respectively (Dosseto *et al.*, 2008). The granitic rocks outcropping at the Earth's surface frequently contain daughter isotopes in disequilibrium with their parents (Latham and Schwarcz, 1987; Villar *et al.*, 1997; Osmond *et al.*, 1999 and Dawood, 2001). Chemical processes and the alpha-recoil effects are the main factors responsible for the uranium-series disequilibrium. It is reasonable to determine the time of the disequilibrium event by measuring the extent to which the isotope system has returned to the state of secular equilibrium. Both ^{238}U and ^{234}U isotopes belong to the same radioactive decay chain and disequilibrium between them is due mainly to water–host rock interaction, leading to preferential mobilization of ^{234}U. Therefore, the determination of ^{238}U and ^{234}U has been extensively used to study characteristics of host rocks and rock-water interactions, including the determination of rock dissolution rates and other related physical and chemical processes (Osmond *et al.*, 1999; Luo *et al.*, 2000 and Bourdon *et al.*, 2003).

The main objective of this study has been to analyse the uranium and thorium isotopes in the two granitic rock types from Wadi El Shallal area in order to assess geologically recent uranium mobility. This is expected to give insight into the possibility for these granites to be a uranium source for the nearby Um Bogma Formation.

Geological Setting

Geologically, southern Sinai consists mainly of an intricate complex of high and very rugged igneous and metamorphic mountains of the Arabian-Nubian Shield. These mountains form the hard core of Sinai Peninsula and rise to great height (*e.g.* Gebel Katherina, 2641 m above sea level). Immediately overlying this massif are the plateaus of low dip slopes consisting of the dark-colored purplish and reddish sandstones of Um Bogma Formation which is occasionally interbedded with thin bands of fossiliferous limestones of Carboniferous age (Fig. 1). To the north of Um Bogma plateau lies the great El-Tih plateau. The eastern and western scarps of this massif form part of the Great Rift Valley occupied by the Gulfs of Aqaba and Suez, respectively.

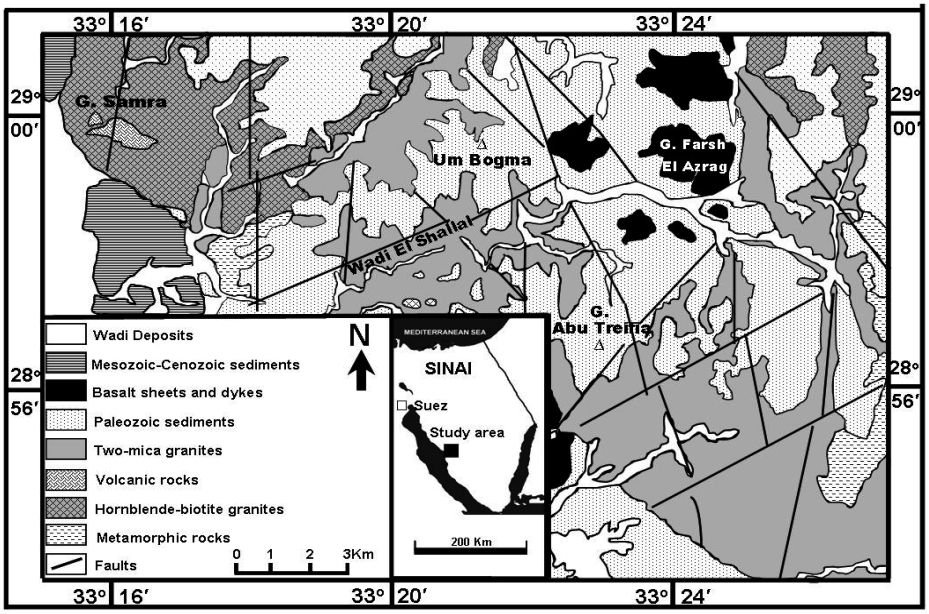

Fig. 1. Geologic map of the study area showing the outcrops of the granitic rocks.

The drainage system of the study area is mainly controlled by the existing structures. Wadi El-Shallal is one of the wadis which drain their water during rainy periods to the Gulf of Suez. The encountered rocks at this wadi are composed mainly of metamorphic assemblage, hornblende-biotite granites, rhyolite, two-mica granites, Paleozoic sediments, basaltic sheets and Mesozoic-Cenozoic sediments. The metamorphic rocks are composed of mica schist, metapelites, biotite gneisses and migmatites. In addition to garnet porphyroblasts, these rock varieties contain cordierite, sillimanite and staurolite indicating genesis from Al-rich pelites under high temperature and low to medium pressure of metamorphism in amphibolite facies (Ibrahim, 1997 and Ibrahim *et al.*, 2000). The hornblende-biotite granites are dated as 653±26 Ma using K-Ar techniques (Abdel Kariem and Arva-Sos, 1992). They are medium to coarse grained rocks that form moderate to high relief (about 700 m above sea level). The hornblende-biotite granites are highly fractured jointed and enclose xenoliths of older rocks such as gneisses and migmatites. Petrographically, these rocks are composed of plagioclase (An_{18-28}), hornblende and biotite in addition to minor quartz and orthoclase. The accessory minerals are represented by zircon, apatite, titanite, monazite and magnetite.

The younger granites in southern Sinai were emplaced over a restricted time span between ~580 and 595 Ma using SHRIMP U-Pb zircon method (Ali *et al.*, 2008). At Wadi El Shallal area, the younger granites are intruded and infrequently form apophyses in the older rocks. They are represented by medium to coarse grained, pink colored two-mica granites. These rocks are slightly altered, jointed and occasionally cavernous. They stand in higher relief compared to the older granites. Petrographically, the two-mica granites are composed of quartz, orthoclase, perthite, plagioclase (An_{6-14}), biotite and muscovite. Zircon, titanite, monazite, fluorite, allanite and magnetite represent the essential accessory phases. Some secondary minerals have been formed due to alteration for instance chlorite, kaolinite, sericite and epidote.

The two granitic rock types are intruded by different types of dike swarms, felsite, quartz and pegmatitic veins. The two-mica granites are overlain by Paleozoic sedimentary successions which are represented

mainly by Ordovician to Carboniferous rocks. The latter rocks comprise the Lower sandstone Series, the Limestone Series and the Upper Sandstone Series. Uranyl minerals represented mainly by zippeite, uranophane, carnotite, meta-autunite, meta-torbernite, were reported from the middle carbonate rock units (Um Bogma Formation) (Hussein *et al.*, 1971; El-Reedy *et al.*, 1988; Afifi, 1991; Hussein *et al.*, 1992; El Agami, 1996; Abdel Monem *et al.*, 1997 and El Galy *et al.*, 2008).

Sampling and Analytical Procedures

Fifteen representative rock samples were collected from Wadi El Shallal granites. The petrography of these rocks was examined using transmitted light microscopy. Eight rock samples from the two-mica granites and seven from the hornblende-biotite granites were analyzed for major elements and U-Th isotopic compositions (Tables 1 and 2). The major elements were measured using two analytical methods. Si was analyzed using Unicam UV2/100 Spectrophotometer and the rest was analyzed using Perkin–Elmer Atomic Absorption Spectrophotometer (AAS). Absolute accuracy has been assessed by comparison with international USGS rock standards analyzed along with the samples and is generally better than 2%.

Uranium and thorium were determined using alpha-spectrometric techniques. The uranium and thorium isotope analyses were carried out for the whole-rocks and leachates. Two grams of the pulverized whole rock samples were dissolved using a mixture of HNO_3 and HF in a pressure container at 140^{o}C for two weeks. Since the whole-rock contains uranium in two different forms (fixed uranium in accessory minerals and labile uranium along cleavage and fracture planes) which could not be separated mechanically, leaching procedures were carried out to determine the amount of labile uranium and to compare the isotopic composition of the different phases. Leaching procedures have been done using two chemical reagents representing acidic and alkaline media. Four grams from pulverized rock samples were subjected to leaching by 1N HNO_3 (Weijden *et al.*, 1985) and 0.2N K_2CO_3 (Green, 1994). The leaching process was carried out for 24 h at 70^{o}C. During this time the sample was periodically stirred and the normality adjusted. The detailed

procedure for uranium and thorium isotope analyses is described in detail by Lally (1992). ^{236}U and ^{229}Th spike tracers were added to the samples before digestion. The solutions obtained by complete dissolution of the whole rocks and by leaching procedures were left for three days to reach isotopic equilibrium. A carrier consisting of ferric nitrate was added, then the sample was heated to boiling and the pH increased to a value of about 10 by the addition of ammonium hydroxide. The actinides including uranium and thorium co-precipitate with ferric hydroxide floc. After separation of the floc by centrifugation, the iron was separated from the actinides by solvent extraction with diethyl ether. The anion exchange procedures were performed for uranium-thorium separation and purification. The resin used is Bio-Rad AG 1X8, 100–200 mesh; chloride form. Both hydrochloric and nitric acid stages of anion exchange were used. Thin sources of purified uranium and thorium fractions were prepared by electrodeposition onto stainless steel discs for subsequent isotope measurements by alpha spectrometry available at the Department of Earth and Environmental Sciences, University of Illinois at Chicago (UIC). The uncertainty of the measurements was calculated using standard error formulae based on counting statistics (Friedlander and Kennedy, 1956).

Results

Major Elements

The results of the chemical analyses of the two granitic rock types are given in Table 1. The normative mineral compositions are plotted in the orthoclase-albite-anorthite diagram (Fig. 2). It classifies the collected samples into granite and granodiorite-trondhjemite, respectively. Figure 3 shows the plots of the studied granitic rocks in the Shand Index diagram according to Maniar and Piccoli (1989). It indicates that the two-mica granite is predominantly peraluminous whereas the hornblende-biotite granite is metaluminous. A possible magma source of the studied granitic rocks could be recognized from the plot of SiO_2 vs. K_2O+Na_2O-CaO (Fig. 4, after Frost *et al.*, 2001). It shows that the two-mica granite was formed by partial melting of sedimentary source (S-type) whereas the hornblende-biotite granite is of igneous source (I-type).

Table 1. Major elements and Chemical Indices of Alteration for Wadi El Shallal granites.

Major oxides (%)	Two-mica granites								Hornblende-biotite granites						
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
SiO_2	73.15	74.00	73.24	73.35	73.89	73.62	73.78	72.41	63.60	63.39	59.80	60.34	61.54	62.99	60.26
TiO_2	0.13	0.17	0.13	0.10	0.12	0.11	0.09	0.10	0.15	0.17	0.21	0.24	0.22	0.19	0.25
Al_2O_3	13.77	13.80	14.25	14.27	13.81	14.06	14.26	14.00	15.53	15.46	16.02	15.52	14.94	15.20	15.83
Fe_2O_3	1.18	1.19	1.06	0.90	0.75	0.66	0.77	1.30	4.78	4.64	5.53	5.61	5.67	4.91	5.70
FeO	0.49	0.47	0.50	0.53	0.62	0.65	0.69	0.52	0.81	0.91	1.28	1.14	0.96	0.83	1.13
MnO	0.02	0.03	0.02	0.03	0.02	0.02	0.03	0.02	0.03	0.03	0.05	0.06	0.04	0.08	0.06
MgO	0.62	0.60	0.52	0.43	0.37	0.46	0.50	0.62	1.91	2.05	2.71	2.65	2.54	2.16	2.72
CaO	1.20	1.09	1.13	0.92	0.95	1.01	0.96	1.50	4.26	4.43	5.14	4.84	4.83	4.16	4.87
Na_2O	3.83	3.50	3.60	3.91	3.94	3.90	3.81	3.84	4.47	4.49	4.84	4.89	4.72	4.81	4.86
K_2O	4.63	4.51	4.79	4.98	4.84	4.68	4.42	4.90	3.33	3.22	3.34	3.49	3.29	3.36	3.16
P_2O_5	0.16	0.17	0.14	0.12	0.14	0.15	0.13	0.16	0.15	0.14	0.18	0.19	0.16	0.17	0.16
L.O.I.	0.59	0.47	0.46	0.51	0.49	0.61	0.51	0.49	0.98	1.03	0.87	0.97	1.06	1.13	0.95
Total	99.77	100.00	99.84	100.05	99.94	99.93	99.95	99.86	100.00	99.96	99.97	99.94	99.97	99.99	99.95
CIA	0.506	0.523	0.520	0.514	0.507	0.514	0.527	0.495	0.445	0.451	0.435	0.431	0.428	0.444	0.440

CIA: Chemical Index of Alteration

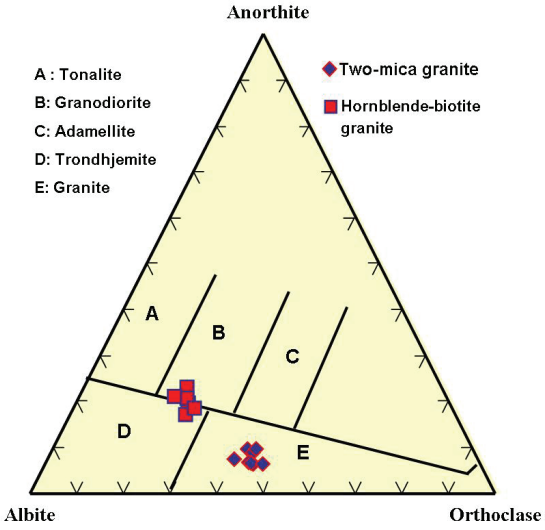

Fig. 2. Nomenclature of El Shallal granites based on normative mineral composition (after O'Connor, 1965).

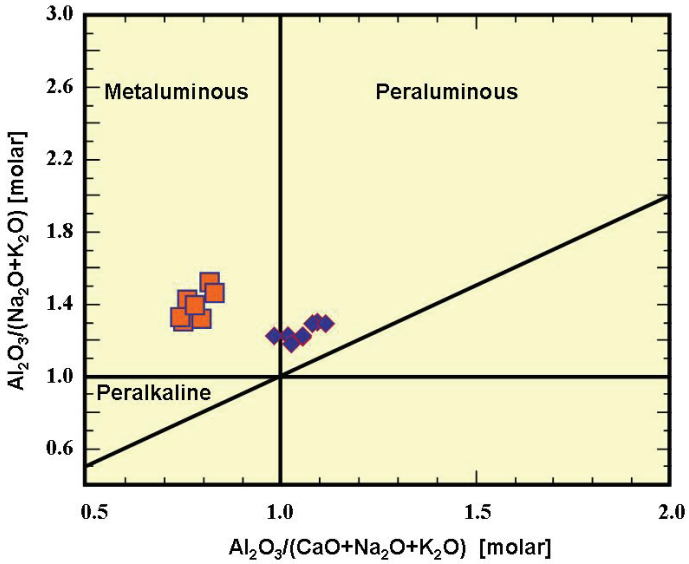

Fig. 3. The Shand index of El Shallal granites after Maniar and Piccoli (1989). See Fig. 2 for Symbols.

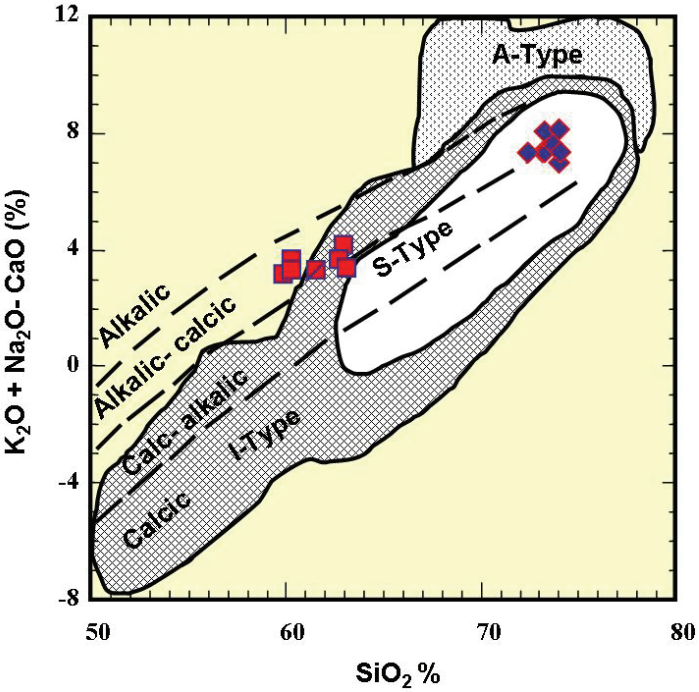

Fig. 4. Na_2O+K_2O-CaO vs. SiO_2 plot showing the composition of El Shallal granites (after Frost *et al*., 2001). Symbols as in Fig. 2.

U-Th Isotopic Composition

The results of uranium and thorium isotope analyses are given in Tables 2-3. In general, the uranium and thorium radioelement contents of the studied two-mica granites are higher than the corresponding contents of the hornblende-biotite granites. The average uranium content of the two-mica granites is 20.3 ppm and that of thorium is 27.1 ppm. Conversely, the average uranium content of the hornblende-biotite granites is 4.9 ppm and that of thorium is 6.51 ppm (Table 2). In both rock types, the concentrations of the two radioelements obtained from nitric acid leach are remarkably higher than the corresponding values of the potassium carbonate leach (Table 3). A plot of uranium concentration of 1 N nitric acid leachates versus whole rock uranium for the studied two granitic types (Fig. 5) shows that around 50% of the uranium in the two-mica granites is readily leachable and therefore located around grains or on grain surfaces whereas, around 25% of the uranium in the hornblende-biotite granites is leachable, *i.e.* more uranium can be leached from the two-mica granites than the hornblende-biotite granites.

Table 2. Isotope data for the whole granitic rocks of Wadi El Shallal area.

	S. N.	Whole rock			
		U (ppm)	Th (ppm)	$^{230}Th/^{234}U$	$^{234}U/^{238}U$
Two- mica granites	1	25.38±1.10	40.24±3.51	1.55±0.12	0.69±0.03
	2	9.14±1.20	16.61±2.10	1.66±0.08	0.78±0.05
	3	16.23±0.98	27.45±2.98	2.07±0.13	0.81+0.04
	4	23.4±0.50	33.07±4.02	1.42+0.05	0.54±0.02
	5	29.11±0.65	14.15±1.20	1.61±0.15	0.87±0.05
	6	22.08±0.46	19.95±3.00	1.76±0.05	0.58±0.02
	7	9.70± 0.33	45.30±3.33	2.15±0.09	0.33±0.01
	8	27.13±1.05	20.22±2.47	1.59±0.13	0.63±0.05
Hornblende-biotite granites	9	5.01±0.22	4.3±0.61	1.08±0.12	0.91±0.04
	10	3.68±0.11	6.3+0.52	1.11±0.11	0.93±0.07
	11	3.61±0.24	3.75±0.24	1.03±0.08	0.89±0.03
	12	5.34±0.63	7.11±0.95	1.17±0.14	0.94±0.04
	13	6.21±0.33	9.16±0.63	1.24±0.15	0.83±0.05
	14	4.2±0.19	6.88±0.29	1.13±0.07	0.98±0.07
	15	6.5±0.64	8.1±0.68	1.30±0.06	0.73±0.04

The $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ activity ratios for the whole-rock of both rock varieties display values out of equilibrium. Compared to the hornblende-biotite granites, the isotopic ratios of the ^{238}U series in the two-mica granites significantly deviates from equilibrium (averages of $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ activity ratios are 0.65 and 1.73, respectively) whereas the corresponding ratios for the hornblende-biotite granites average 0.89 and 1.15, respectively (Table 2). On the contrary to the $^{234}U/^{238}U$ activity ratios, the $^{230}Th/^{234}U$ ratios of the acid leachates are usually higher than the corresponding ratios of alkaline leachates (Table 3).

Table 3. Isotope data of the 1N nitric acid and 0.2N potassium carbonate leachates of Wadi El Shallal granites.

	S. N.	1N H_2NO_3 Leachates				0.2N K_2CO_3 Leachates			
		U (ppm)	Th (ppm)	$^{230}Th/^{234}U$	$^{234}U/^{238}U$	U (ppm)	Th (ppm)	$^{230}Th/^{234}U$	$^{234}U/^{238}U$
Two- mica granites	1	13.19±0.40	16.1±1.20	0.97±0.12	0.83±0.04	6.09±0.40	1.28±0.30	0.18±0.04	1.51±0.08
	2	3.93±0.14	9.14±0.80	0.45±0.04	0.92±0.04	2.29±0.10	0.35±0.08	0.08±0.01	1.80±0.07
	3	8.2±0.47	17.96±1.80	0.90±0.11	0.84±0.03	3.57±0.10	0.71±0.13	0.20±0.03	1.44±0.04
	4	12.87±0.40	19.18±1.30	0.91±0.10	0.50±0.02	6.32±0.30	1.30±0.20	0.13±0.02	1.58±0.05
	5	13.65±0.90	7.64±0.90	0.82±0.07	0.88±0.03	8.44±0.70	0.31±0.06	0.16±0.02	1.32±0.04
	6	12.59±0.50	9.58±0.80	0.51±0.03	0.91±0.03	4.42±0.30	0.45±0.13	0.13±0.02	1.13±0.05
	7	5.09±0.30	14.95±0.60	0.73±0.04	0.89±0.04	2.23±0.13	1.13±0.09	0.3±0.04	1.21±0.06
	8	12.57±0.70	7.88±0.90	0.66±0.05	0.95±0.03	6.65±0.40	0.52±0.11	0.12±0.01	1.29±0.08
Hornblende-biotite granites	9	1.43±0.12	2.37±0.12	0.61±0.03	0.86±0.03	1.03±0.07	0.12±0.01	0.10±0.01	0.93±0.04
	10	0.91±0.10	3.47±0.30	0.53±0.03	0.92±0.04	0.83±0.05	0.26±0.04	0.09±0.01	1.06±0.04
	11	0.98±0.07	2.25±0.20	0.42±0.03	0.99±0.04	0.75±0.05	0.17±0.03	0.12±0.02	1.08±0.03
	12	1.34±0.16	4.62±0.60	0.37±0.02	0.84±0.03	1.25±0.09	0.28±0.02	0.08±0.01	0.95±0.04
	13	1.59±0.08	4.12±0.20	0.82±0.06	0.83±0.03	1.27±0.12	0.38±0.04	0.15±0.01	1.11±0.05
	14	1.08±0.05	5.09±0.60	0.56±0.03	0.94±0.04	0.94±0.10	0.21±0.01	0.08±0.01	1.02±0.03
	15	1.66±0.14	3.12±0.20	0.43±0.02	0.96±0.05	1.41±0.16	0.45±0.07	0.10±0.01	1.14±0.04

Discussion

Major Elements Geochemistry

The chemical analyses of the studied granitic rocks indicated that El Shallal two-mica granites are predominantly peraluminous and display

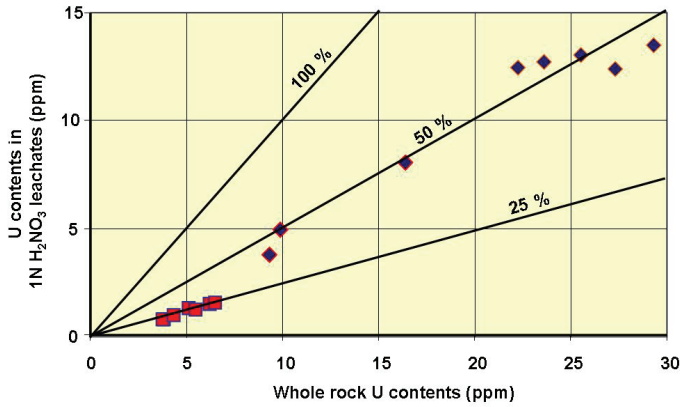

Fig. 5. Around 50% of the uranium in the two-mica granites and only 25% in the hornblende-biotite granites are leachable. Symbols as in Fig. 2.

the characteristics of S-type granite. Conversely, the hornblende-biotite granite is classified as metaluminous and I-type granite. The average Na_2O content of the two-mica granites is 3.79% and that of the hornblende-biotite granites is 4.73%. This is consistent with the fact that the typical S-type granites usually possess low Na_2O contents relative to I-type (Harris *et al.*, 1986). This low value of Na_2O is an indication for the chemical maturity state of the source rocks since Na_2O decreases during alteration of feldspars and subsequent transformation to clay minerals (Stallard, 1988). The two-mica granites of El Shallal area were probably generated by crustal anatexis of metasediments during collision state. This is consistent with the field observations that El Shallal two-mica granites occur associated with mica and chlorite schists and metapelites. Similar two-mica granites were described to be derived from metapelites and schist encountered near the granitic plutons: *e.g.* the Wadi El-Imra district (Furnes *et al.*, 1996) and El Sukkari granite (Dawood *et al.*, 2005). In general, the close chemical and mineralogical similarities of anatectic peraluminous granites and their host metasedimentary rocks led many investigators to propose either dehydration or hydrous partial fusion of the metasedimentary rocks as a reasonable source for these granites (Thompson, 1982; Clemens and Vielzeuf, 1987; Le Fort *et al.*, 1987; Shearer *et al.*, 1987; Barbey *et al.*, 1990 and Dawood *et al.*, 2005).

The I-type metaluminous granites form characteristic constituents of convergent margin settings. Their petrogenesis is commonly attributed to variable interactions of mantle-derived mafic magmas with the

continental crust (Holden *et al.*, 1987 and Altherr *et al.*, 1999) or to simple remagmatization of older crust (Patiño Douce and McCarthy, 1998) with subsequent fractionational crystallization and/or restite crystal fractionation (Chappell, 1996).

The mafic magma source of the hornblende-biotite granites is consistent with their relatively low uranium and thorium contents (Table 2). Conversely, uranium anomalies are widely reported in peraluminous two-mica granites. Generally, the main magmatic differentiation process in the two-mica granites consists of progressive removal of poorly soluble mineral species and a correlated enrichment of incompatible elements such as uranium and thorium. Other uranium enrichments are related to uraninite and pitchblende concentrations in the shear zones associated with mineralogical evidence of an ortho-derived fluid phase rich in F, Zr, Sn, Be and B (Friedrich and Cuney, 1989). Uranium in peraluminous granites is also incorporated in accessory phases such as monazite and zircon (Förster, 1998).

Correlations between uranium and thorium contents in the two granitic types are illustrated in Fig. 6. The positive correlation between uranium and thorium in the hornblende-biotite granites reflects their geochemical coherence during the crystallization of magma, and indicates that both elements are largely accommodated in accessory minerals and were not significantly disturbed by alteration processes. Conversely, lack of positive correlation between uranium and thorium in the two-mica granites indicates post magmatic redistribution of uranium and this could be regarded as a criterion for uranium prospecting in other areas because uranium may have concentrated into deposits within or near the source rock (Osmond *et al.*, 1999 and Dawood *et al.*, 2004).

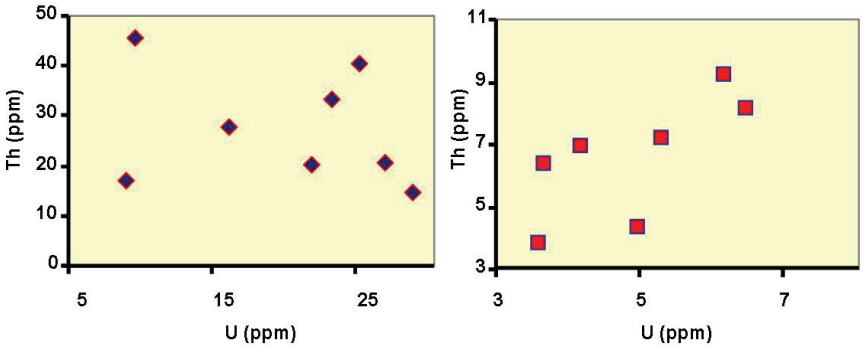

Fig. 6. Bivariant plots of uranium vs. thorium in the two granitic types of El Shallal area. Symbols as in Fig. 2.

Nesbitt and Young (1982) demonstrated that the degree of weathering could be expressed quantitatively by calculation of the Chemical Index of Alteration (CIA) using molecular proportions as follows:

$$CIA = Al_2O_3/(Al_2O_3+Na_2O+ K_2O+CaO^*)$$

where CaO* is the amount of CaO incorporated in the silicate fraction of the rock. A correction is made for CaO content in apatite. In general, weathering and alteration processes tend to remove soluble elements such as Na, K and Ca and cause CIA value to increase. So, highly weathered rocks approach 1.00, the CIA value of kaolinite, whereas for granite and granodiorite, the CIA average of unaltered rocks is 0.5 (Nesbitt and Young, 1982). The calculated CIA values for the collected granitic samples are presented in Table 1 and plotted in Fig. 7. From this figure, the CIA values of the two-mica granites indicate slight alteration effects. These values range between 0.495 and 0.527 with an average of 0.513, slightly higher than the average of unaltered granites (0.5). Conversely, the CIA values of the hornblende-biotite granites range between 0.428 and 0.445 with an average of 0.439 indicative of unaltered rocks.

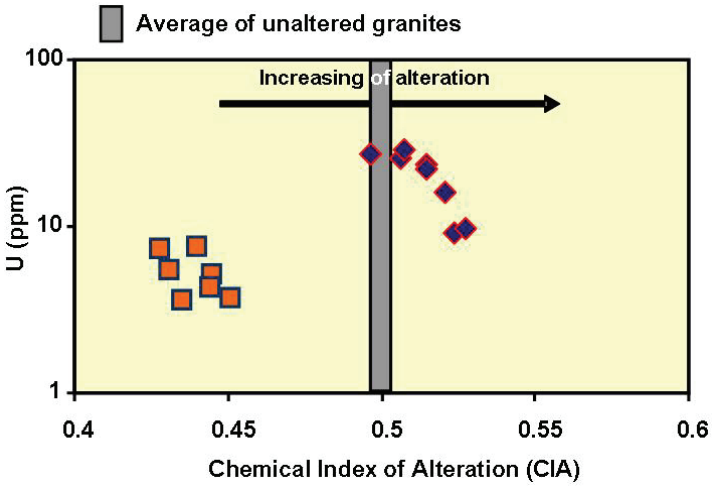

Fig. 7. Relation between uranium contents and the Chemical Index of Alteration (CIA) for the studied granitic rocks. Symbols as in Fig. 2.

U-Th Isotopic Fractionation

The silicic igneous rocks such as granite, rhyolite and pegmatite are considered to be important sources for uranium mobilization because

they have relatively high uranium and thorium contents and contain a significant fraction of labile uranium. Also, they have a consistent spatial association with uranium deposits. Uranium-series disequilibrium can potentially provide an important tool for tracing migration of uranium series radionuclides in different environments. The uranium mobilization is usually associated with fractionation in the uranium isotopes ^{238}U and ^{234}U. This fractionation is the reason for a significant excess of ^{234}U in the hydrosphere (Rosholt, 1982). In most terrestrial water systems, the $^{234}U/^{238}U$ activity ratio is greater than 1 and in some deep aquifers as much as 10 or 20. Moreover, high activity of ^{234}U was reported from groundwater in contact with granitic rocks (Dabous *et al.*, 2002 and Reyes and Marques, 2008). In general, the $^{234}U/^{238}U$ ratios are discussed in terms of the activity ratio even though the isotope ratio is now the measured quantity (Cheng *et al.*, 2000). The removal of uranium from granitic rocks is generally characterized by $^{234}U/^{238}U \leq 1$ and $^{230}Th/^{234}U > 1$ (Latham and Schwarcz, 1987). This is consistent with the activity ratios of the studied Precambrian granitic rocks from El Shallal area (Table 2).The essential cause for these ratios is that groundwater leaches ^{234}U preferentially to ^{238}U and removes both uranium isotopes relative to ^{230}Th. The observed depletion of ^{234}U in these granitic rocks (Table 2) is agreeable with literature results (*e.g.* Rosholt, 1983; Michel, 1984 and Dawood, 2001). For these rocks, physical reactions such as α-recoil that affect ^{234}U**,** must dominate over the chemical reactions**.** Several studies have investigated the potential advantage of α-recoil to isotope fractionation. α-particles and the recoil nuclei can cause the movement of other radionuclides (Johnston *et al.*, 1993 and Leonard, 1995). Based on the CIA index (Fig. 7), the two-mica granites are slightly altered. This slight alteration produced some mineralogical changes and formation of secondary minerals such as chlorite, kaolinite, sericite and epidote. Although, the degree of alteration is insignificant as indicated from Figure 7, uranium was found to be responsive in such conditions. An inverse correlation could be observed between uranium contents and the CIA values in the two-mica granites indicative of significant releasing of uranium with alteration (Fig. 7). In surficial environment, uranium is largely soluble, essentially as uranyl ions (UO_2^{+2}). Other speciations such as uranyl carbonates, phosphates, sulfates and fluoride complexes, when present, greatly enhance the solubility of uranium (Langmuir, 1978 and Dong and Brooks, 2006). Compared to uranium; thorium is rarely soluble in such conditions due to its single oxidation state and incorporation in refractory and non-leachable minerals in the granitic rocks.

The effect of this alteration on the isotopic composition of the ^{238}U series in both the two-mica and hornblende-biotite granites is demonstrated in Table 2 and Fig. 8. The range of the $^{230}Th/^{234}U$ activity ratios in the two-mica granites (1.42-2.15) is higher than the corresponding range in the hornblende-biotite granites (1.03-1.30). This indicates that ^{234}U in the two-mica granites is more mobilized. In this rock type, the activity of ^{234}U decreases significantly with increasing the degree of alteration. Conversely, the corresponding radionuclide in the hornblende-biotite granites shows non-significant relation with alteration (Fig. 8). This outcome is consistent with the previous conclusion that uranium in the hornblende-biotite granites is largely fixed in the non-leachable accessory phases such as zircon. Figure 9 also shows that the mobilization of ^{234}U is accompanied with increasing alteration effects onto the rocks. Compared to the hornblende-biotite granites, the $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ activity ratios of the two-mica granites display values that depart significantly from equilibrium. The sample plots of the two rock types are distributed in a trend reflecting more ^{234}U loss from the two-mica granites (Fig. 9).

The relation between the $^{234}U/^{238}U$ activity ratios for nitric acid leachates are plotted against the corresponding activity ratios of potassium carbonate leachates for the two rock types (Fig. 10). From this figure, the lower activity ratios mean more uranium leaching whereas the higher activity ratios mean less uranium leaching. Obviously, more uranium is obtained from the nitric leachates than the potassium carbonate leachates.

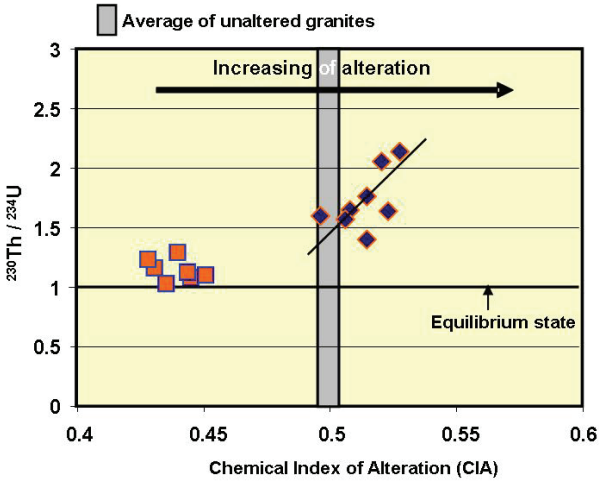

Fig. 8. Variation of $^{230}Th/^{234}U$ activity ratio with the Chemical Index of Alteration in the studied granitic rocks. Symbols as in Fig. 2.

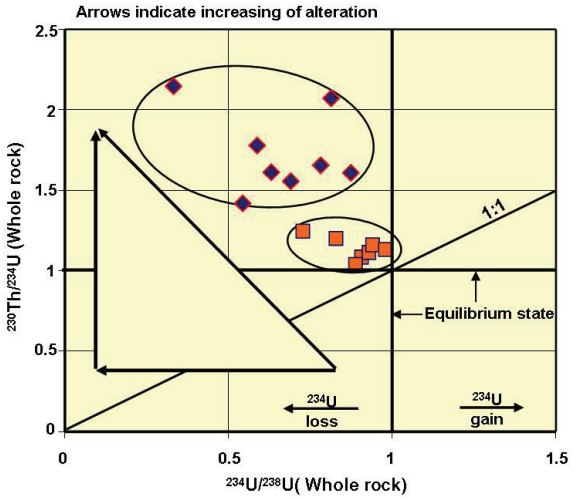

Fig. 9. Evolution of uranium series disequilibrium in the granitic rocks of El Shallal area, the two-mica granites are significantly out of equilibrium compared to the hornblende-biotite granites. The loss of ^{234}U is largely controlling the disequilibrium state. Symbols as in Fig. 2.

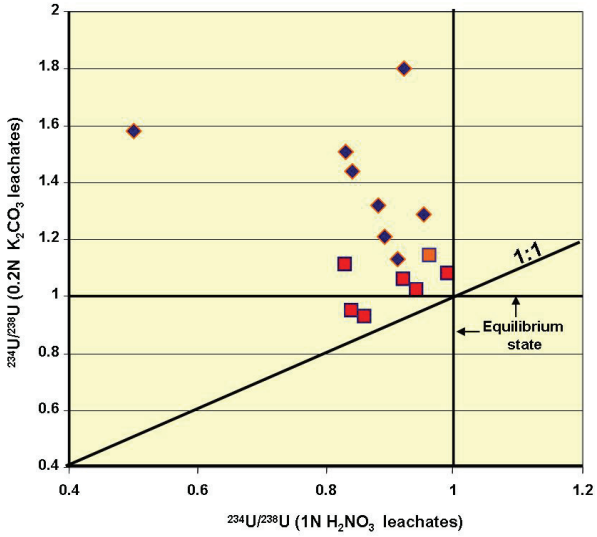

Fig. 10. Plot of the activity ratios of nitric acid leachates vs. the corresponding ratios of potassium carbonate leachates. Symbols as in Fig. 2.

On the other hand, the activity ratios for the potassium carbonate leachates are higher than the activity ratios of the nitric acid leachates. This reflects that potassium carbonate leaches mainly the labile uranium.

Around mineral grain boundaries, the sites of labile uranium, ^{234}U is enriched due to alpha recoil from the uranium bearing minerals. Nitric acid leaches some uranium from the peripheries of these minerals which is relatively enriched with ^{238}U, in addition to leaching of labile uranium. These variations can be understood in terms of α-recoil effects in the mineral grains of the rock. The α-recoil process displaces the daughter nuclei in the uranium decay-series by ~200 to 350 Å (Kigoshi, 1971 and Nasdala *et al.*, 2001). In addition, it triggers off a collision cascade that creates a small structurally damaged volume. The daughter isotope, therefore, may be prone to preferential removal or leaching by fluids. Recoiled daughter isotopes of near surface uranium (incorporated in the crystal lattice or on the grain surface) may be ejected from the grain into the open spaces and thereby become lost (Romer and Rocholl, 2004). In an open system, such a rock would acquire a deficit in ^{230}Th and ^{234}U. Since the solubility of thorium in aqueous fluids is much lower than that of uranium, leaching process would result in lower $^{234}U/^{238}U$ and higher $^{230}Th/^{234}U$ ratios in the mineral. The net effect is depletion in ^{234}U relative to ^{238}U in the surface layer of the grain, and an enrichment of ^{234}U relative to ^{238}U in the area surrounding the grain. Because ^{234}Th is insoluble and particle reactive, ejected ^{234}Th should adsorb to grain surfaces. When it decays to ^{234}Pa and ^{234}U, the recoil from beta-decay is too small to re-implant the generated daughters ^{234}Pa and ^{234}U back into the grains and any implanted ^{234}U is likely to be easily leached (DePaolo *et al.*, 2006) (Fig. 11).

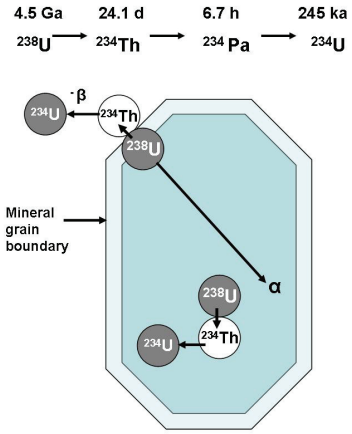

Fig. 11. Effect of alpha-recoil on the isotopic fractionation at the mineral grain boundaries (modified after DePaolo *et al.*, 2006). Recoil ejection of ^{234}Th from a mineral grain as a result of the alpha decay of ^{238}U, followed by beta decay of ^{234}Th to ^{234}U.

Conclusions

The metaluminous granites of Wadi El Shallal area are represented by hornblende-biotite granites of low uranium contents. These granites are of I-type and classified as granodiorite-trondhjemite as indicated from the normative mineral composition. On the other hand, the two-mica granites constitute the S-type peraluminous granites of high uranium potentiality. The two-mica granites are slightly altered compared to the hornblende-biotite granites as confirmed by the Chemical Index of Alteration. The alteration process results in uranium loss in the two-mica granites whereas, the uranium budget of the hornblende-biotite granites is mainly incorporated in refractory and non-leachable minerals.

The removal of uranium from granitic rocks of El Shallal area is generally characterized by $^{234}U/^{238}U<1$ and $^{230}Th/^{234}U>1$. Compared to the hornblende-biotite granites, the $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ activity ratios of the two-mica granites display values that depart significantly from equilibrium. The observed uranium series disequilibrium in the granitic rocks from El Shallal area highlights isotopic fractionation and a recent migration of uranium in the peraluminous granites. Leaching experiments confirmed that the sites around mineral grains are more enriched with daughter ^{234}U relative to the parent ^{238}U where the α-recoil process displaces the daughter nuclei in the uranium series of the grain surface. This daughter is ejected from the grain into the open spaces and therefore, prone to preferential removal or leaching by fluids.

The results of the present study confirmed that uranium mobilization has taken place in the two-mica granites under the oxidizing environment during the past 300 ka to 1 Ma. The meteoric water acting through the structures and cavities increased the rate of oxidation and hence mobilization and leaching of uranium from the aerated parts. The leached uranium was probably precipitated in the nearby Carboniferous Um Bogma Formation where varieties of uranyl minerals have been reported in the middle dolomitic unit of this formation (Abdel Monem *et al.*, 1997). The present work should be regarded as preliminary for more extensive investigations. These investigations should link the uranium-series isotopic compositions of the probable source rocks, groundwater and uranium mineralization of the Um Bogma Formation.

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تجزؤ نظائر سلسلة اليورانيوم كدليل على حركة اليورانيوم فى جرانيتات منطقة الشلال، غرب وسط سيناء، مصر

يحيى حسن داود

كلية علوم الأرض جامعة الملك عبد العزيز، المملكة العربية السعودية

المستخلص. صنفت الصخور الجرانيتية فى منطقة وادى الشلال إلى جرانيتات الهورنبلند والبيوتيت الألومنيومية، وجرانيتات ثنائية الميكا فوق الألومنيومية. دلت التحاليل الكيميائية على أن الجرانيتات ثنائية الميكا تعرضت لعمليات تغاير بسيطة، وأنها تحتوى على نسب عالية نسبيا من اليورانيوم والثوريوم. مقارنة بجرانيتات الهورنبلند والبيوتيت، تبين نسب النشاط الإشعاعى $^{230}Th/^{234}U$ و $^{234}U/^{238}U$ للجرانيتات ثنائية الميكا أنها فى حالة عدم اتزان ذات دلالة. تتموضع عينات كلا النوعين من الصخور فى اتجاه يبين فقدًا للنظير ^{234}U من الجرانيتات ثنائية الميكا. أكدت عمليات الاستخلاص بالإذابة الرشحية أن المواضع حول حبيبات المعادن فى هذه الصخور غنية بشكل عام فى ^{234}U مقارنة بالنظير ^{238}U، حيث يزيح رد فعل خروج جسيم ألفا النظير الوليد عند سطح حُبيبة المعدن. يزاح النظير الوليد ^{234}U إلى المناطق حول حبيبات المعدن، ويصبح أكثر عرضة لعمليات الاستخلاص بواسطة المحاليل. تدل عملية عدم الاتزان فى سلسة اليورانيوم فى صخور الجرانيت بمنطقة الشلال على تجزؤ نظائري وحركة حديثة لليورانيوم فى الجرانيتات ثنائية الميكا. يمكن الاستدلال من تركيزات اليورانيوم العالية والتركيب النظائرى لليورانيوم والثوريوم، أن الجرانيتات ثنائية الميكا ربما تكون المصدر

الصخري لتمعدنات اليورانيوم الثانوية لتكوين أم بجمة القريب من هذه الصخور الجرانيتية. يعكس ذلك تاريخاً من الإذابة والحركة والترسيب بواسطة مياه الأمطار والمياه الجوفية. على الرغم من نتائج هذه الدراسة، يوصى بعمل دراسات تفصيلية لعدم الاتزان فى سلاسل اليورانيوم للمياه الجوفية ومعادن اليورانيوم الثانوية فى تكوين أم بجمة.