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Geology and Mineralogy of the Radioactive Ferruginous Siltstones at Wadi El Seih Area, Southwestern Sinai, Egypt

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Abstract: *The lower member of the Um Bogma Formation at Wadi El Seih area composed mainly of ferruginous siltstones. Most of uranium occurrences located in the ferruginous siltstones. The field observation reveal that the factors affected the localization of U at the study area are topography, structure and lithology. The ferruginous siltstones show high enrichment of Y, Zn, V, Sr, U and Ba. Also, the rare earth elements (REE) analyses reveal that the studied siltstones have high concentrations of Dy, Ce, Nd, Er and Gd.SEM and EDX investigations reveal that iron oxides play an important role in the adsorption and precipitation of trace and rare earth elements. The high radioactivity of the studied siltstones is related to the presence of uranium minerals like uranophane, meta-autunite , sklodowskite and other associated uranium bearing minerals like xenotime and zircon. Gold is detected in the studied ferruginous siltstones of the lower member of Um Bogma Formation (reach up to 1.04 ppm).The presences of iron minerals are playing an important factor in capturing uranium and other elements.*
Key wards: *uranium, gold, Um Bogma*

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I. Introduction

Sinai Peninsula is a famous poly metallogenic province in Egypt, hosts to ferromanganese, gold, copper, uranium and other rare metals mineralization. The metallogenic domains are in close relationship with the Paleozoic sedimentary succession and its surrounding granitic rocks. In the study area, the exposed younger granites are nonconformably overlain by the Paleozoic succession which is capped by Triassic basalt sheet.

Several authors divided the Paleozoic succession in the study area and its vicinities. The most notorious subdivisions are Barron (1907) which includes three major rock units that comprise from base to top: a): Lower Sandstone Series. b): Middle Carbonate Series and c):Upper Sandstone Series. The basal sandstone unit (a) is subdivided into Sarabit El Khadim, Abu Hamata and Adedia formations by Solimanand Abu El Fetouh (1969). They also subdivided the upper sandstone unit (c) into El-Hashash, Magharet El-Maiah and Abu Zarab formations. While, Weissbrod (1980) assigned the name Abu Thora Formation for the upper three formations of Soliman and Abu El Fetouh (1969).Weissbrod (1969) was the first to assign the name Um Bogma Formation for the middle carbonate series of Barron (1907). The unconformity surfaces were recorded between Um Bogma Formation and both of Lower Sandstone Series and Upper Sandstone Series.

Many authors studied the metals content within the Paleozoic sedimentary rocks units in east Abu Zeneima area such as Ag (**Amer, 1993);** U, Mo, As, and V (**Mahdy et al., 1998; Shata and Mira, 2010);** Mn (**El Agami, 1996)**; Al, U-Th, Cu and Zn (**El Aassy et al., 1986; 1997 and 2003; El Agami (1996); Shata and El Bilakassy, 2012)** and REE, U, Cu, V **(Shata, 2013)**.

2. Geological Setting

The study area is located between 33º 21` 09" - 33° 21` 27" E and 28° 53` 08" - 28° 53` 33" N (Fig. 1) and is considered a part of east Abu Zeneima promising area. This area covered mainly with the Paleozoic sediments which have its importance owing to its content from economic ores as coal, copper, manganese, kaolin, glass sands, REEs and uranium. It characterized by moderate to low topography and covered mostly by Paleozoic succession which underlain nonconformably by Precambrian younger granites in some parts (Fig. 2A). The Paleozoic sedimentary rock units in the study area comprise three stratigraphic units arranged from base to top: Lower Sandstone Series(includes Sarabit El Khadim, Abu Hamata and Adedia formations), Um Bogma Formation and the Upper Sandstone Series (includes El Hashash, Magharet El Maiah and Abu Zarab formations which are equivalent to Abu Thora Formation).

Um Bogma Formation is considered the most important rock unit in the study area due to its content from uranium, Mn-Fe ore deposits and secondary copper mineralization.Um Bogma Formation unconformably overlies Adedia Formation (Fig, 2B) and underlies Abu Thora Formation. It comprises three members as follows;

The lower member is rest unconformably on Adedia Formation with undistinguishable contact due to different lithology. It is enriched by manganese and iron ores. This member exhibits three different lithologic facies include a) Mn-Fe ore, ferromanganese siltstone and silty shale facies with black, black brown and reddish brown colors; b) sandy dolomite facies, it is reveals thick bedded, pink color and sometimes shows horizontally laminated; c) black carbonaceous shale, siltstone facies which considered the more important facies for the uranium mineralization and characterized by presence of high radioactive anomalies. It is usually variegated, such as purple, yellow, yellowish green, brown and grey in color within the oxidation zone but the dark gray to black color is dominant in the reduction zone. Uranium mineralization settled by two modes of occurrences. The first, within the black shale, it is found as clusters of crystals within scattered ambient spots. The second mode of occurrence found within facies (a) and (b) it is well developed at the intersection of faults and in the interbedded fracture zone.

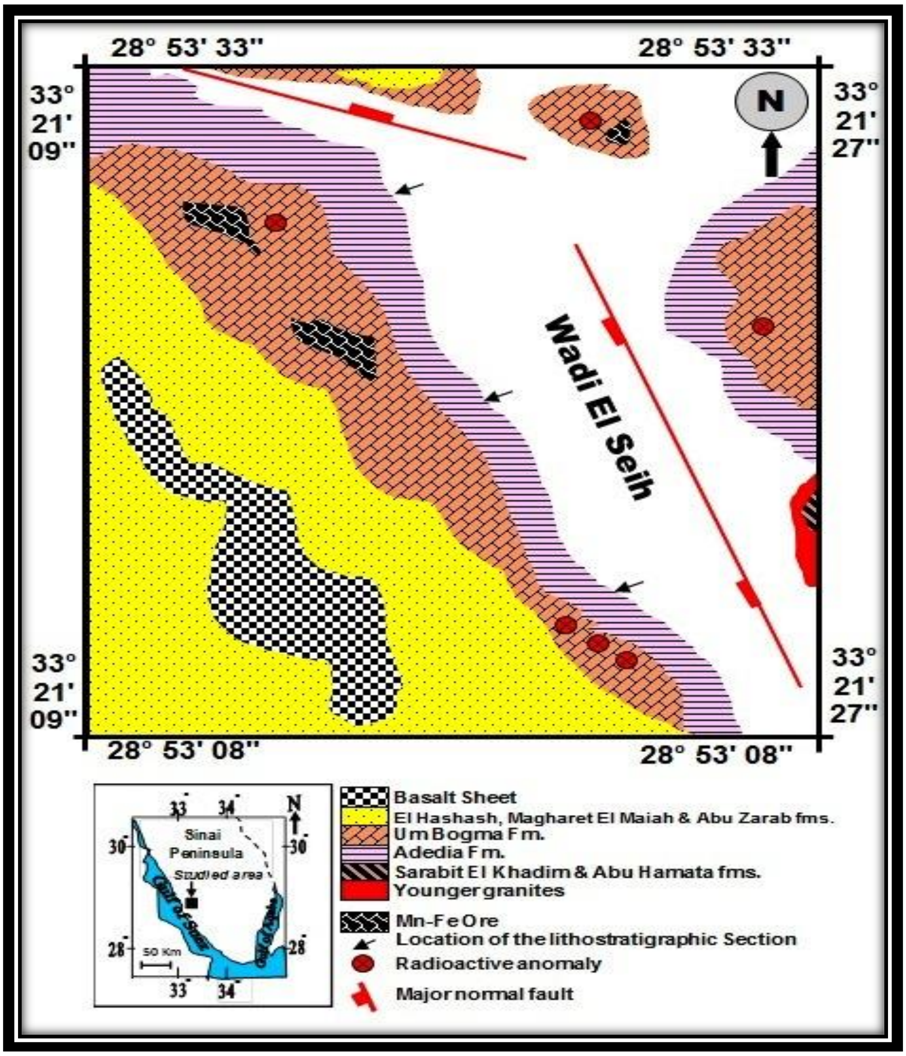


Fig. 1: Geological map of Wadi El Seih area.

The middle member is composed of intercalations of marly dolostone, shale and siltstone with rhythmic variation beds of discontinuous carbonate interbedded with clastics with yellow, earthy black colors and fossils content such as Corals, Crinoids, Brachiopods and Mollusca. The middle member is characterized by presence of high radioactive anomalies in addition to evaporate minerals as gypsum, anhydrite and halite minerals in fibrous and platy habits in the form of parallel veinlets and/or intersection with bedding planes. Also, white and black gibbsite horizon was observed at the boundary between middle and lower members of Um Bogma Formation with high radioactive anomalies.

The upper member is composed of yellow, pink, and grayish crystalline dolomite with little sandstones intercalations. It is overlies conformably the middle member and shows cliffs and steps outcrops.

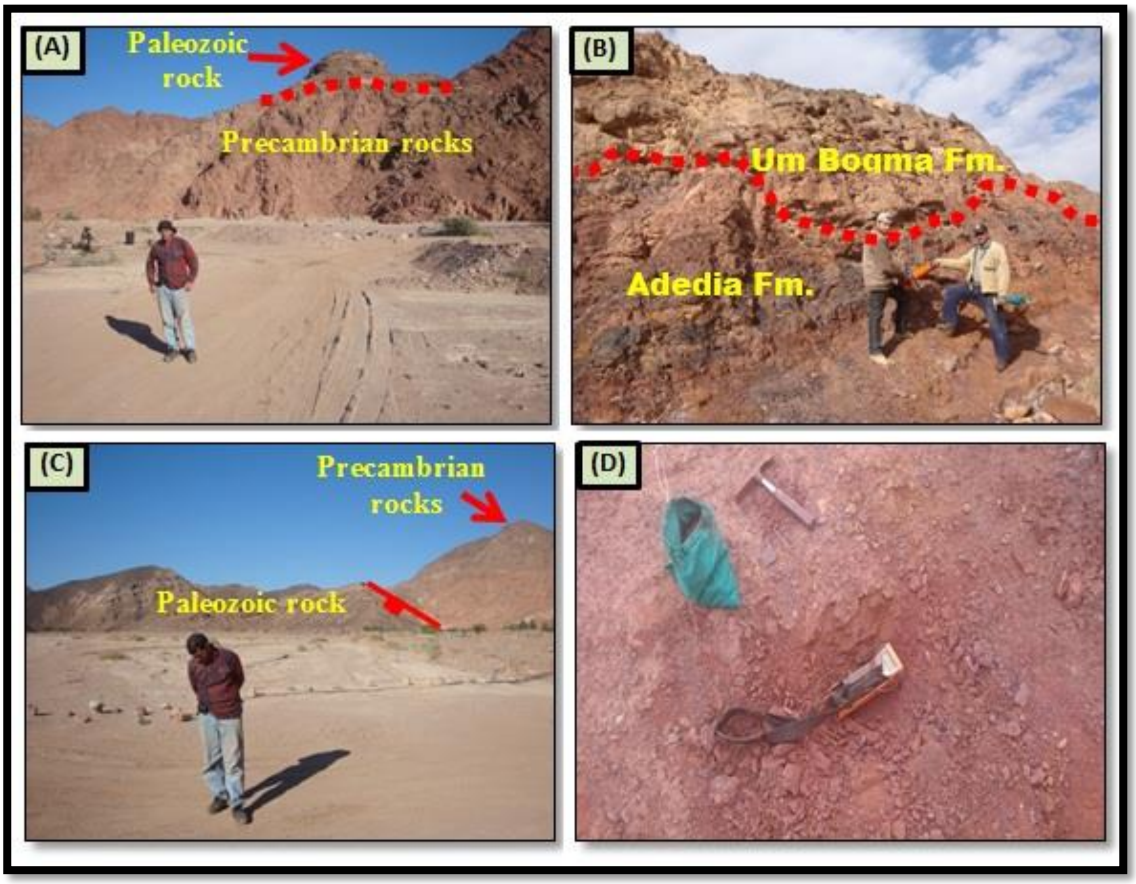


Fig. 2: (A) Precambrian rocks nonconformably overlain by Paleozoic rocks. (B) Um Bogma Formation unconformably overlies Adedia Formation. (C) Normal fault. (D) Radioactive ferruginous siltstones in the lower member of Um Bogma Formation.

The study area were faulted by normal faults (Fig. 2C) with vertical downthrows reaching up to 30m to the north and west directions; the major faults usually control the location of deep wadies as well as the landscape as it causes dipping of Paleozoic succession in the study area toward the west.

Three lithostratigraphic sections were compiled for the study area (Fig. 3) to shed light on the litho stratigraphic succession and correlate the thickness variation along N-S extension for the study area. Adedia Formation is ranging in thickness from 15 to 25 m, which decreasing toward the south direction. Um Bogma Formation is ranging from 5 to 10 m and also decreasing toward the south. On the other hand, the Abu Thora Formation is ranging from 25 to 35 m. finally; the basaltic sheet and sill reach to about 5 m.

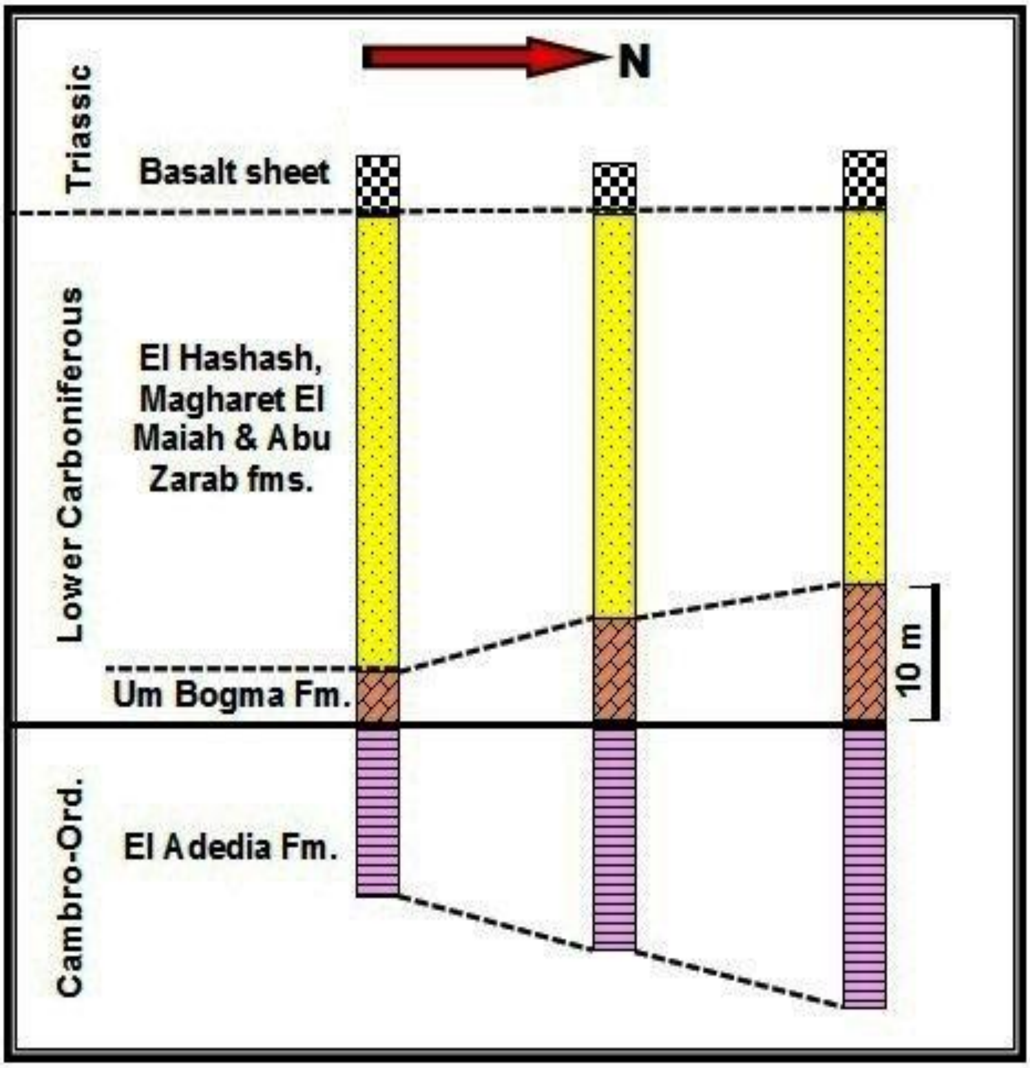


Fig. 3: Lithostratigraphic correlation along Wadi El Seih area.

II. Methodology

The field radiometric measurements of eU (ppm), eTh (ppm) and K% were obtained using a portable differential gamma ray spectrometer model Rs-230 BGO Super-Spec, serial No. 4333, manufactured by Radiation Detection Systems AB, Backehagen 35, SE-79191 FALUN, Sweden and the reading were given directly each 30 second.

The chemical concentrations of the trace and rare earth elements were estimated by ACME Laboratories Canada. 9050 Shaughnessy St Vancouver British Columbia V6P 6E5 Canada.

For measuring gold, fire assay analyses were carried out at the Egyptian Mineral Resources Authority (EMRA), Central Laboratory Sector. Weigh 50 gm of sample. Addition of flux (litharge- Borax –Sodium carbonate –Flour –Silica – silver).Mix sample with flux in ceramic crucible. Melting of (sample + flux) at 1000^0 C for 1.5 hours. Cupellation of (lead + gold +silver) alloy at 900^0 C for 1 hour. Parting of resulting (gold –silver) alloy in Nitric acid and aqua regia heating to get gold solution. Analysis of gold solution by GBC Avanta atomic absorption instrument to get gold concentration with ppm.

The anomalous samples, whether soft or hard, were collected (weighting from 3 to 5kg for each sample). In order to determine their mineralogical constituents; the samples were crushed and screened. The fractions having grain size range between 0.074mm and 0.5mm were used. These size fractions were subjected to systematic mineral separation techniques using bromoform (Sp.G. = 2.85) as a heavy liquid and magnetic fractionation using Frantz Isodynamic Magnetic Separator at side slope of 5^o, forward slope of 20^o and 0.5 A **(Flinter, 1959)**.

The obtained heavy mineral fractions were studied under the Binocular microscope and Environmental Scanning Electron Microscope (XL30-ESEM, Philips) attached with EDAX microanalysis unit developments in high-pressure (low-vacuum). These analyses were carried out in the laboratories of the Nuclear Materials Authority (NMA), Cairo, Egypt. While, the polished sections were studied under the Scanning electron microscopy (Quanta FEG 200, FEI France, Thermo Fisher Scientific, Mérignac, France) while element analysis was obtained using an Oxford Inca 350 EDX microanalyzer (connected to SEM, Oxford Instruments France, Saclay, France)

III. Radioactivity

The radioactivity was recorded in several areas in Sinai. The most notable one were recorded in the Paleozoic rocks, especially in Um Bogma Formation. Most of the radioactive anomalies are concentrated in definite stratigraphic horizon among them, the most significant uranium occurrences located at the study area in the ferruginous siltstones (Fig. 2D) of the lower member of Um Bogma Formation.

The anomalous horizon is located within the down thrown of Wadi El Seih normal fault (structural control) and is very near to the level of Wadi El Seih (low topographic horizon). Uranium mineralization filling the siltstones cavity in the form of bright yellow minerals aggregates. From the field observation, the high eU measurements restricted within specific lower horizon of Um Bogma Formation (strat abound). So, the factors affected the localization of U at the study area are topography, structure and lithology.

According to the field radiometric measurements (Tab. 1) the eU-contents in the anomalous ferruginous siltstones reaches more than 1280 ppm, while the eTh-contents not exceeds 69 ppm.

Table 1: Radiometric and chemical measurements of the anomalous ferruginous siltstones at Wadi Seih area.

Station No.	Member	Fm.	Rock type	eU (ppm)	eTh (ppm)	Uc (ppm)	Th (ppm)	Uc/eU	eU /eTh
1	Lower	Um Bogma	Ferruginous siltstones	230	29	56.2	2.6	0.2	7.9
2				265	37	79.3	3.1	0.3	7.1
3				814	49	180.1	7.6	0.2	16.6
4				965	53	193.2	8.6	0.2	18.2
5				1103	55	194.8	8.3	0.17	20.05
6				1223	62	196.4	8.5	0.16	19.7
7				1287	69	190.2	8.3	0.14	18.65
8				445	38	95.4	5.1	0.21	11.7
			Min	230	29	56.2	2.6	0.14	7.1
			Max	1287	69	196.4	8.6	0.3	20.05
			Average	791.5	49	148.2	6.5	0.19	14.99

The eU/eTh ratio is a very important geochemical index for U migration in or out the studied sediments, it is approximately constant in the same rock type or geologic unit in relatively closed environment. The Clark value for eU/eTh in the sedimentary rocks is equal 1 **(Clark, et al. 1966)**.The eU/eTh ratio of the studied anomalous ferruginous siltstones is very high than Clark value, where the average ratio reaches 14.99.This reveals that the studied anomalous ferruginous siltstones undergo to migration in (leaching) of uranium from the surrounding country rock unites. The presence of iron minerals is playing an important factor in capture of uranium.

D-factor is equal to the ratio of the chemically measured uranium (U)/ radiometrically measured uranium (eU), if this factor was more or less than unity; it indicates disequilibrium state which, could be due to an addition or removal of U **(Hansink, 1976)**. Chemical measurements of uranium contents of the anomalous ferruginous siltstones are less than those of field radiometric measurements. So, the average of D-factor reachesto0.19.This reveals that disequilibrium state characterized the studied horizon due to the removal of uranium.

Generally, the probable origin of the radioactive anomalies recorded in the study area could be attributed to the epigenetic concept, in which, the secondary ascending hydrothermal solutions and circulating meteoric water remobilized and leached U from the surrounding rocks and carry it out to deposit mainly along fractures and faults.

IV. Geochemistry

Four samples of the lower member ferruginous siltstones of Um Bogma Formation at the study area were measured chemically to determine their trace, rare earth elements (REE) and uranium contents (Table 2& 3).

According to the trace elements measurements (Table 2), the ferruginous siltstones show high enrichment of Y, Zn, V, Sr, U and Ba (Fig. 4&5). Um Bogma Formation hosts Mn-Fe ore, Cu, U,REE and gibbsite mineralization **(Mart and Sass, 1972; Segev, 1984; El Agami, 1996; Shata, 2013)**. Also, rare earth elements (REE) analyses reveal that the studied siltstones have high concentrations of Dy, Ce, Nd, Er and Gd(Table 3).

The trace elements measurements in this study normalized to the North American Composite Shale (NASC, Gromet et al. 1984). The studied samples show high enrichment in Y and in all of the compared heavy metals (Fig. 6). Adsorption process attributed to the presence of iron oxides which plays a role in precipitation of trace elements.

The La/Y ratio as geochemical parameter used to interpret the pH conditions of depositional environment of the different facies in the middle member of Um Bogma Formation. Values > 1 and< 1 for La/Y ratio is related to alkalic and acidic environment, respectively (**Crinci and Jurkowic 1990; Maksimovic and Pantó 1996).** Value of La/Y ratio <1 in the studied ferruginous siltstones (Tab. 3) provide reasons to believe that the studied sediments are within alkali epigenetic environment.

Table 3: Trace elements concentrations(ppm) of the ferruginous siltstones at Wadi El Seih area.

	S1	S2	S3	S4
Ba	113	150	129	126
Co	17	17.9	17.5	17.5
Cs	1.1	1.1	1	1.1
Ga	19.62	18.98	18.69	19.6
Hf	2.25	2.45	1.95	2.53
Nb	14.95	14.35	16.63	13.68
Rb	8.5	8.5	7.7	8.1
Sn	1.5	1.5	1.5	1.4
Sr	560	568	512	523
U	193.2	194.6	196.4	190.2
Th	8.6	8.3	8.5	8.3
Ta	0.9	1	1.3	1
Tl	0.179	0.173	0.167	0.173
V	966	962	959	980
W	7.9	8.4	8.4	8.2
Zr	76.7	75.5	67.8	68.9
Mo	23.97	24.26	23.69	24.47
Cu	85.6	85.2	86.4	85.8
Pb				
Zn	1144.8	1181.8	1179.6	1199.6
Ni	132.1	135.5	133.3	130.4
Bi	0.19	0.19	0.18	0.19
As	144.1	140.2	140.7	141.5
Cd	4.37	3.97	3.83	4.24
Sb	9.77	9.74	9.27	9.87
Cr	333	331	328	335
Be	16	18	13	16
Sc	10.2	10.4	9.6	9.7
Li	10.1	10.1	9.3	10.4
In	0.18	0.13	0.15	0.17
Re	0.023	0.019	0.019	0.018
Y	2000	2000	2000	1763

Table 4: Rare earth elements concentrations (ppm) in the ferruginous siltstones atWadiEl Seih area.

	S1	S2	S3	S4
La	130.3	130.8	126.1	126.2
Ce	309.79	312.73	305.54	305.17
Pr	49.9	48.1	47.8	46
Nd	253.4	229.1	230	224.9
Sm	89.3	87.3	84.2	81.1
Eu	20.3	22.2	22.5	19.3
Gd	200.2	208.3	203.6	172.9
Tb	50.6	49.4	50.6	44.6
Dy	340.7	340.6	338.7	303
Ho	78.6	78.8	77.1	70.1
Er	209.1	221.8	214.7	197.4
Tm	30	30.1	30.8	27.7
Yb	148.9	154.3	152.5	135.1
Lu	23.4	22.8	23.3	20.7
∑REE	1934.39	1936.33	1907.44	1774.17
∑LREE	832.59	808.03	793.64	783.37
∑HREE	1101.8	1128.3	1113.8	990.8
LREE/HREE	0.76	0.72	0.71	0.79
La/Y	0.065	0.065	0.063	0.072

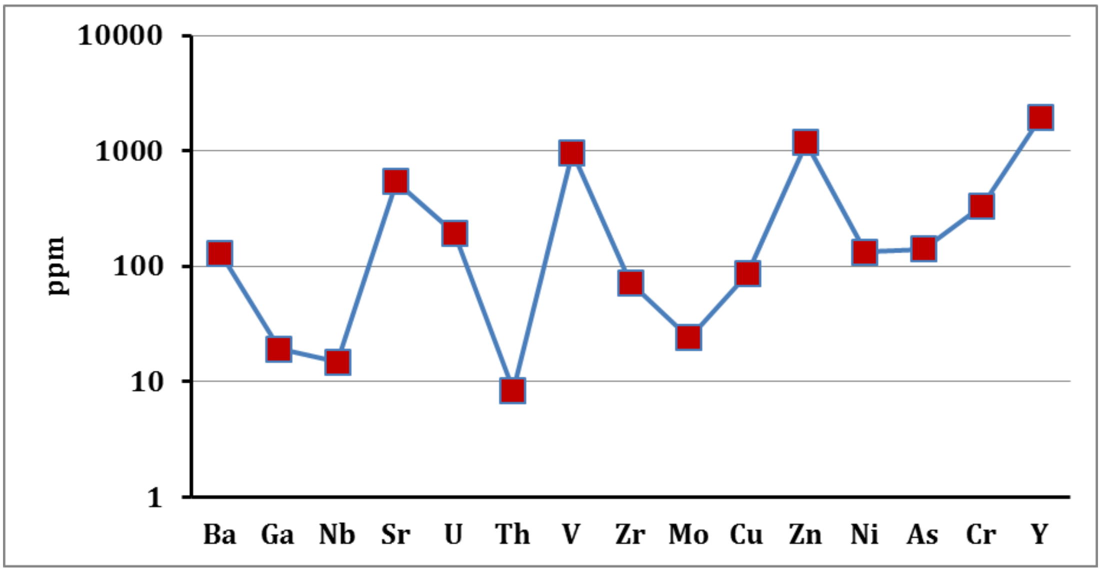


Fig.4: Profile - line diagram showing the average concentrations of the most important trace elements in the ferruginous siltstones.

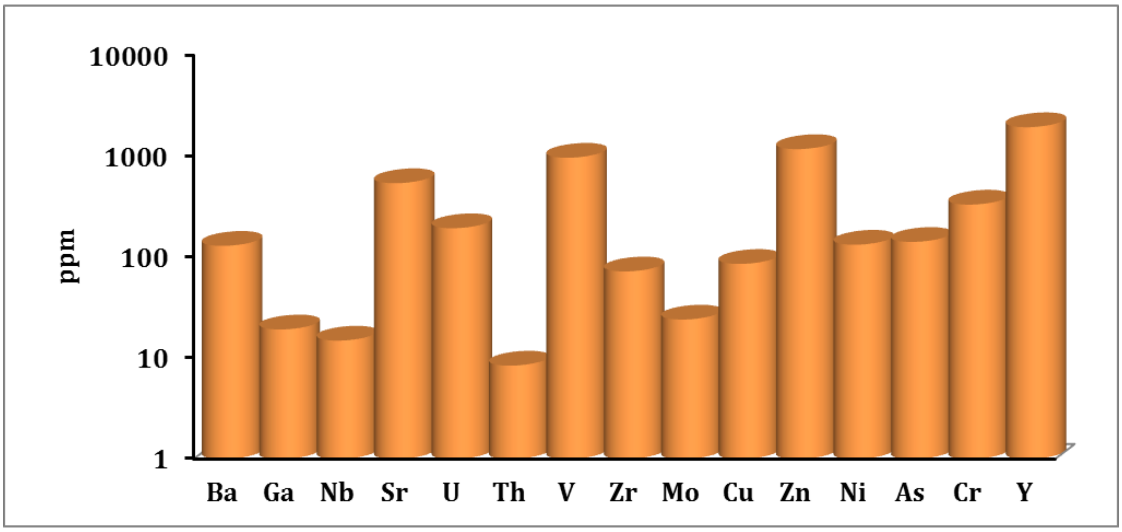


Fig.5: Bar-diagram showing the average concentrations of the most important trace elements in the ferruginous siltstones.

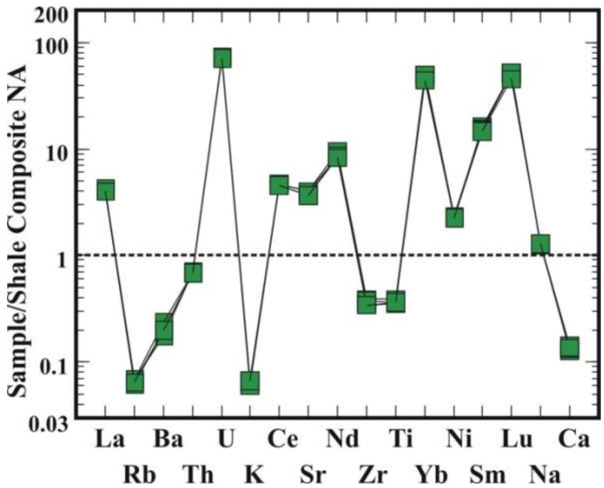


Fig.6: Trace elements pattern normalized to NASC. (Gormet et al., 1984).

The total REEs varies from 1774.17 to 1936.33 ppm, the LREEs ranges from 783.37to 832.59 ppm, while HREEs ranges from 990.8to 1113.8ppm. The LREES/HREES ratio ranges from 0.71 to 0.79 (Table 4) indicating enrichment of HREEs than LREEs. REE patterns of the studied siltstones are demonstrated, as normalized to chondritic abundances **(**Boynton, 1984**)**(Fig.7) and normalized to North American Shale Composite (NASC) abundances **(Haskin et al.,1968;Gormet et el.1984; Taylor and McLennan, 1981)** (Figs.8, 9&10respectively).

According to the normalization patterns, there is enrichment of all rare earth elements (REE) normalized to chondritic and north American Shale Composite (NASC) abundances and there is a positive cerium anomaly (+veCe).

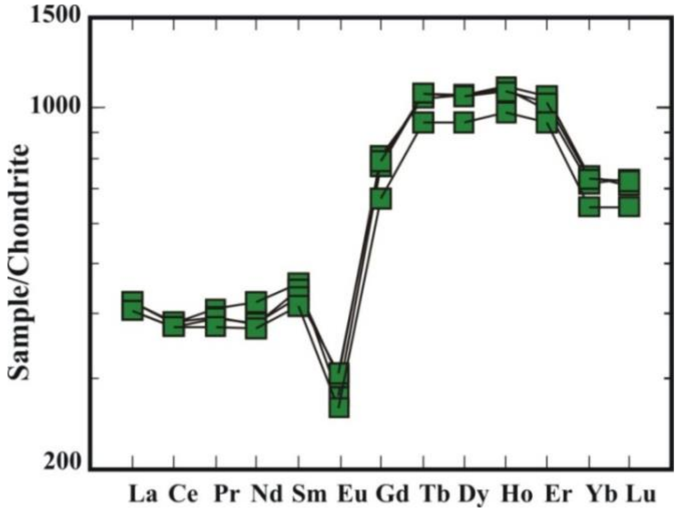


Fig. 7: Rare earth element elements pattern of the studied siltstones normalized to Chondrite (Boynton, 1984).

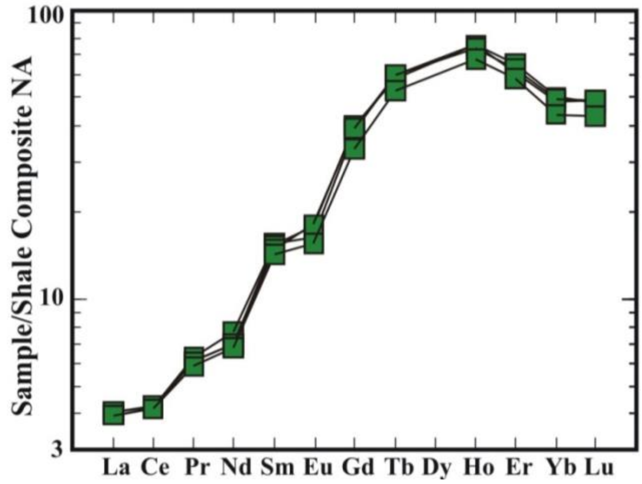


Fig. 8: Rare earth elements pattern normalized to NASC (Haskin et al., 1968).

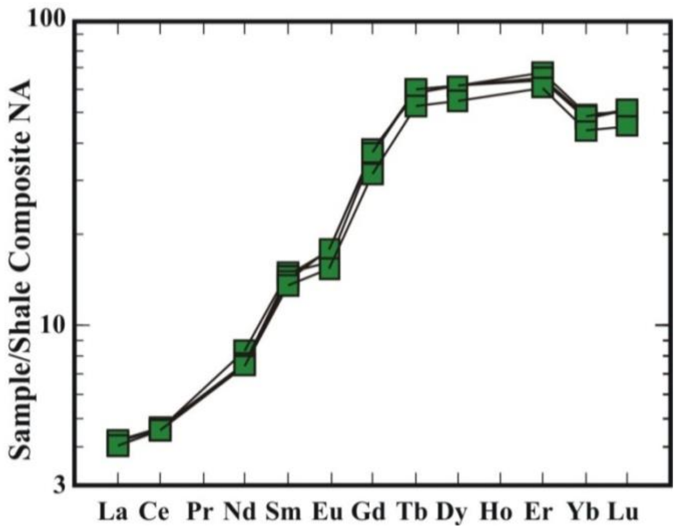


Fig. 9: Rare earth elements pattern normalized with NASC. (**Gromet et al., 1984**)

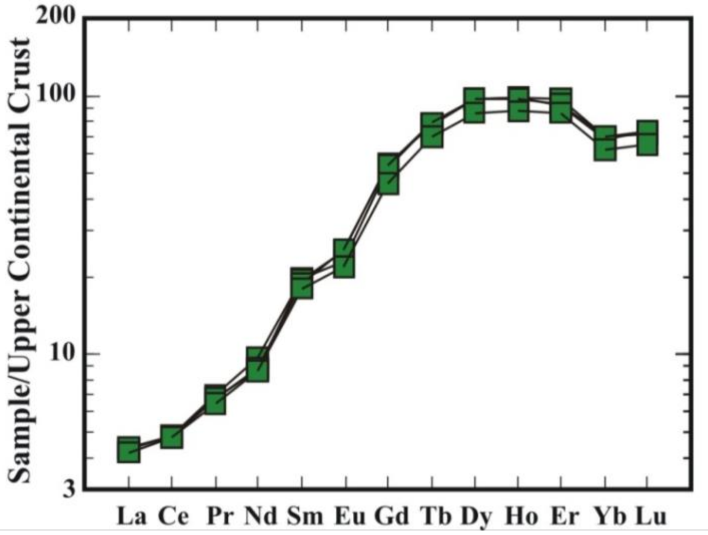


Fig. 10:REEs pattern for the studied siltstones normalized to the Upper Continental Crust (**Taylor and McLennan, 1981)**.

V. Mineralogy

Uranophane [Ca (UO_2) $(SiO_2)_2(OH)_2.5H_2O$]

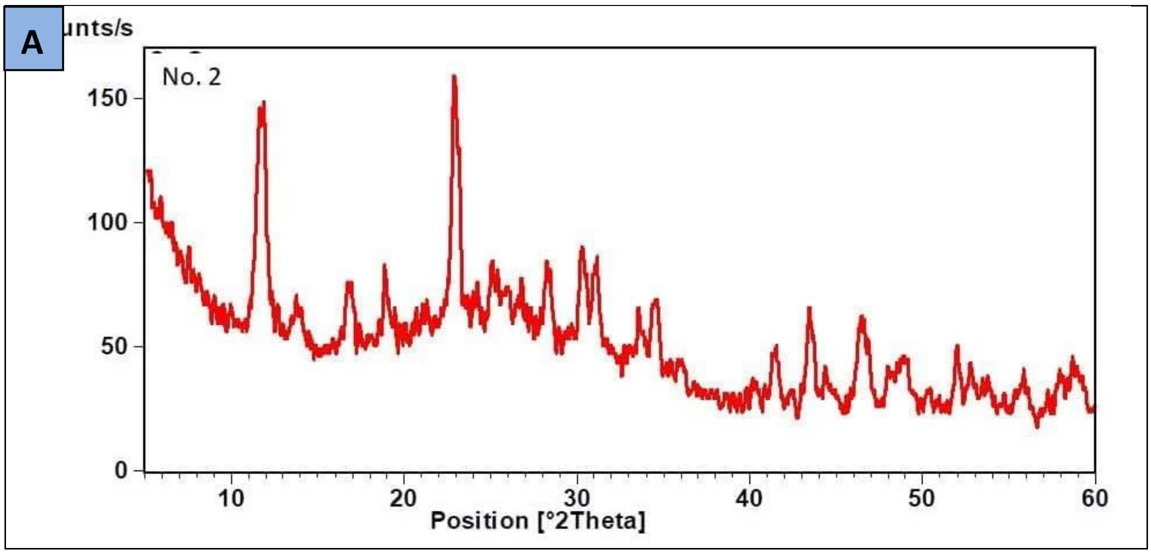
Uranophane is hydrated calcium uranium silicate containing silica in place of the phosphate of autunite. Uranophane is the alteration product of uraninite and the chief constitute of outer silica zone of uraninite alteration. Most uranophane mineral appears to be of supergene origin where it can be noticed in the oxidized parts of deposits. Uranophane is present as small aggregates on quartz surface. The grains are very soft with different grades of yellow to waxy dull color. The EDX analysis (Fig. 11) shows that the semi-quantitative analyses of uranophane grains consist of U (76-79 wt.%), Ca(4.6-3.8)and Si (10-10.2).

Meta-autunite $Ca(UO_2)_2(PO_4)_2 \cdot 6\text{-}8H_2O$

It is greenish yellow to lemon yellow crystals. If the mineral dries out, it can lose its water content and convert to meta-autunite-I, which can turn into meta-autunite-II after heating. It is detected by the XRD analysis (Fig. 12).

Sklodowskite $Mg(UO_2)_2(HSiO_4)_2{\cdot}5H_2O$.

It is a secondary mineral which contains magnesium and is a bright yellow colour. It is detected by the XRD analysis (Fig. 12).



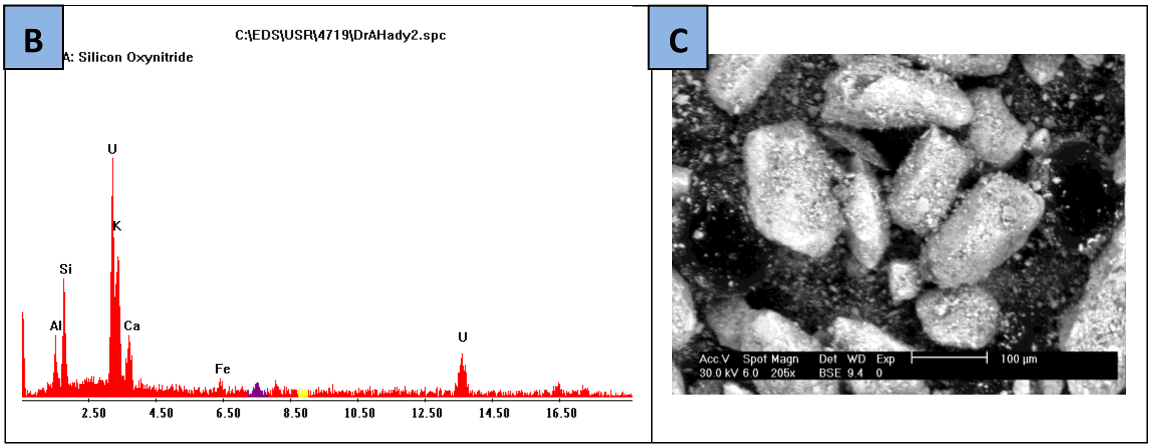


Fig. 11: XRD pattern (A), EDX data (B) and BSE image (C) of uranophane mineral.

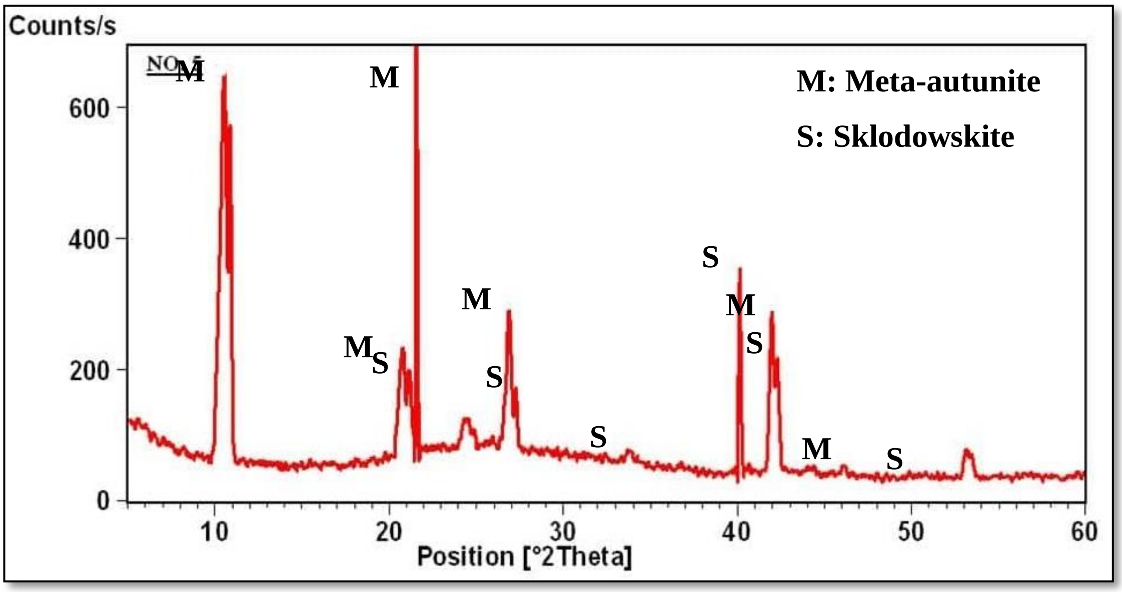


Fig. 12: XRD pattern of Meta-autunite and Sklodowskite minerals.

Xenotime (YPO_4)

Xenotime is a rare earth bearing phosphate mineral, whose major component is yttrium orthophosphate. Xenotime is typically translucent to opaque (rarely transparent) in shades of brown to brownish yellow (most common), but also reddish to greenish brown and gray in color. Xenotime has a variable crystal habit: It may be prismatic (stubby or slender and elongate) with dipyramidal terminations, in radial or granular aggregates, or rosettes. Xenotime was found coating on iron oxides on its surface. The EDX analyses show that P (20.6 wt. %) Y (49 wt. %) and REEs (21.5 wt. %) are the main constituents (Fig. 13).

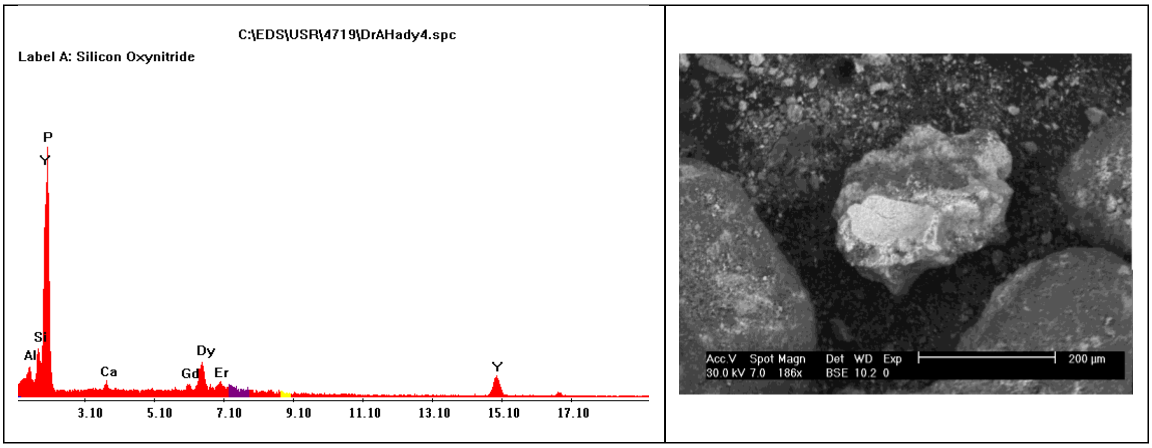


Fig. 13: EDX data and BSE image of xenotime mineral.

Zircon ($ZrSiO_4$)

It is found as subhedral crystals with dark brown colour (Fig. 14). The dark brown color observed in most zircon crystals is caused from iron oxides impurities. The concentration of Y, Th and U in zircon is controlled by magma temperature ***(Li et al., 2014)***.

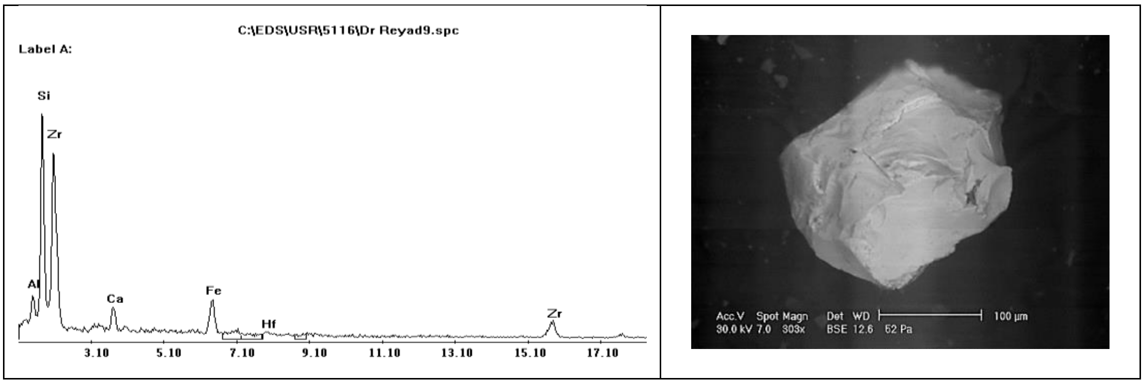


Fig. 14: EDX data and BSE image of zircon mineral.

Gold (Au)

Native Gold occurs in the studied ferruginous siltstones as fine-grained crystals. Gold is present in association with Cu, Fe and Mn(Fig. 15). The gold content was estimated by the fire assay technique, just for preliminary estimation (reach up to 1.04 ppm). **Sallam et al., (2014)** recorded minerals bearing Ag and Au namely uytenbogaardtite and furutobeite in the lower member of Um Bogma Formation at El Sheikh Soliman Area. **Alshami, (2019)** recorded native gold in different facies of Adedia Formation at Um Bogma area.

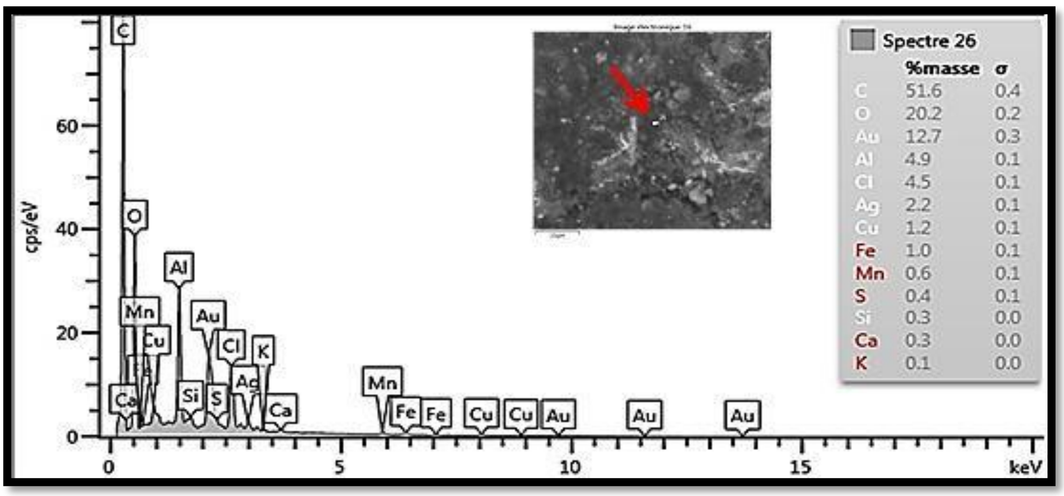


Fig. 15: EDX data and BSE image showing gold with traces of Cu, Fe and Mn.

VI. Conclusions

The Paleozoic sedimentary rock units in the study area comprise three stratigraphic units arranged from base to top: Lower Sandstone Series, Um Bogma Formation and the Upper Sandstone Series. The lower member of the Um Bogma Formation at the study area composed mainly of ferruginous siltstones.

Most of uranium occurrences located in the ferruginous siltstones of the Um Bogma Formation lower member in the study area. From the field observations, the high eU measurements are restricted to specific horizon of the Um Bogma Formation lower part(strata bound). So, the factors affected to localization of U at the study area are topography, structure and lithology.

The eU/eTh ratio of the studied anomalous ferruginous siltstones is very high than Clark value, in which the average ratio reaches 14.99.This reveals that the studied anomalous ferruginous siltstones has migration in (leaching) of uranium from the surrounding country rock unites. The presence of iron oxides minerals is playing an important factor in capturing uranium.

The ferruginous siltstones show high enrichment of Y, Zn, V, Sr, U and Ba. Also, the rare earth elements (REE) analyses reveal that the studied siltstones have high concentrations of Dy, Ce, Nd, Er and Gd. The LREES/HREES ratio ranges from 0.71 to 0.79 indicating enrichment of HREEs than LREEs. .Adsorption process attributed to the presence of iron oxides which play important role in precipitation of trace and rare earth elements.

The high radioactivity of the studied siltstones is attributed to the presence of uranium minerals like uranophane, meta-autunite , sklodowskite and other associated uranium bearing minerals like xenotime and zircon. Gold is detected in the present ferruginous siltstones with significant concentrations reach up to1.04 ppm. The presences of iron minerals are playing an important factor in capturing uranium and other elements.

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