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Research Paper

Recovery of Uranium from the Mineralized Shear Zone of Jabal El Missikat, Eastren Desert, Egypt

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ABSTRACT: *The Nuclear Materials Authority is actively involved in prospection, exploration, reserves evaluation and chemical technological studies for promising conventional uranium ores.A uranium mineralization has actually been recorded in a number of shear zones in both El Missikat and El Erediya Younger granite plutons. They are situated directly south the Qena – Safaga road at sign 85 km. El Missikat granite pluton is just 3 km south of the mentioned sign, while El Erediya is farther south at 25 km from the latter. Both areas are bounded by Lat. 26°15/ to 26°33/N and Long. 33° 15\ to 33° 30\ E.*

The present work studied uranium leaching and extraction from El Missikatmineralized shear zone recently recorded where uranophane mineral is the main uranium one (3030 mg/kg uranium concentration). Through the leaching study, the main factors affecting uranium leaching efficiency were studied such as: grain size, acid type, acid concentration, contact time, temperature and solid / liquid ratio. The obtained optimum uranium leaching conditions are:8 hours contact time, 60 g/l sulfuric acid concentration, at temperature 25 °C, grain size of – 60 mesh size and 1/3 solid/liquid ratio giving 95% leaching efficiency.

Uranium extraction was performed by preparing a stock solution through applying the obtained optimum leaching conditions from which separation ofuranium was achieved using ion exchange resin technique using a dry Chinese anion one (type D263B). After loading and elution, uranium was precipitated using sodium hydroxide solution at pH 7.5where 0.27 gm of sodium diuranate (Yellow Cake) was obtained.

Keywords:*Jabal El Missikat, uranium recovery, recent shear zone anomaly*

I. INTRODUCTION

The nuclear energy sharesnowadays with about 20% in the total international energy production. For example, the nuclear energy contributes by about 87% of the total energy production in France.In this context, during the last 35 years, uranium has become a widely used source for nuclear energy especially for the nuclear electrical generation plants. In the latter, uranium is considered the main fuel. The nuclear fuel activities start with exploration and mining of uranium ore rocks, then milling the uranium ore materials reproduce the base material for the uranium fuel cycle namely the yellow cake (Y.C.).The uranium resources in Egypt can be classified into two main classes namely: the conventional (mined and milled basically as the main product for uranium) and the non-conventional resources (uranium production is a by-product). The Nuclear Materials Authority of Egypt (NMA) has actually discovered several uranium conventional resources (Fig. 1) [1].It is actively involved in prospection, exploration, reserves evaluation and chemical technological studies for promising conventional uranium ores.(as well as for the elements of the nuclear interest), the available assured reserves upon which a full scale industry can be set up are indeed represented by the following two non-conventional uranium (also thorium and rare earths) resources [2], namely:

a) Phosphate ores from which assured reserves in the Egyptian territories are available. They represent an ideal continued and valuable choice as a uranium resource. Such ores are thus considered the first commercial resource for uranium in the Arab Republic of Egypt.

b) Black sands deposits distributed along the northern coast from which assured reserves are also available. They represent the second choice for non-conventional

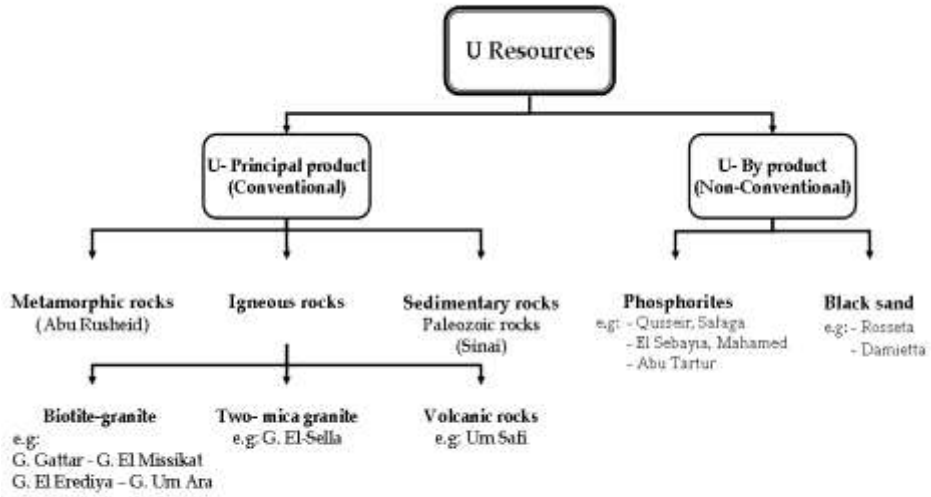


Fig. (1): Uranium conventional resources in Egypt

A uranium mineralization has actually been recorded in a number of shear zones in both El Missikat and El Erediya Younger granite plutons. They are situated directly south the Qena – Safaga road at sign 85 km. El Missikat granite pluton is just 3 km south of the mentioned sign, while El Erediya is farther south at 25 km from the latter. Both areas are bounded by Lat. $26^{o}15^{/}$ to $26^{o}33^{/}$N and Long.33^{0} $15^{\backslash}$ to 33^{0} $30^{\backslash}$ E withthe highest mountainous peak arising up to 1058 m a.s.l. (Fig. 2). On the other hand, it has to be indicated that both these uranium occurrences are indeed strongly associated with silicified shear and fracture zones which extend in the peripheral parts of the two granitic plutons (Fig. 3).

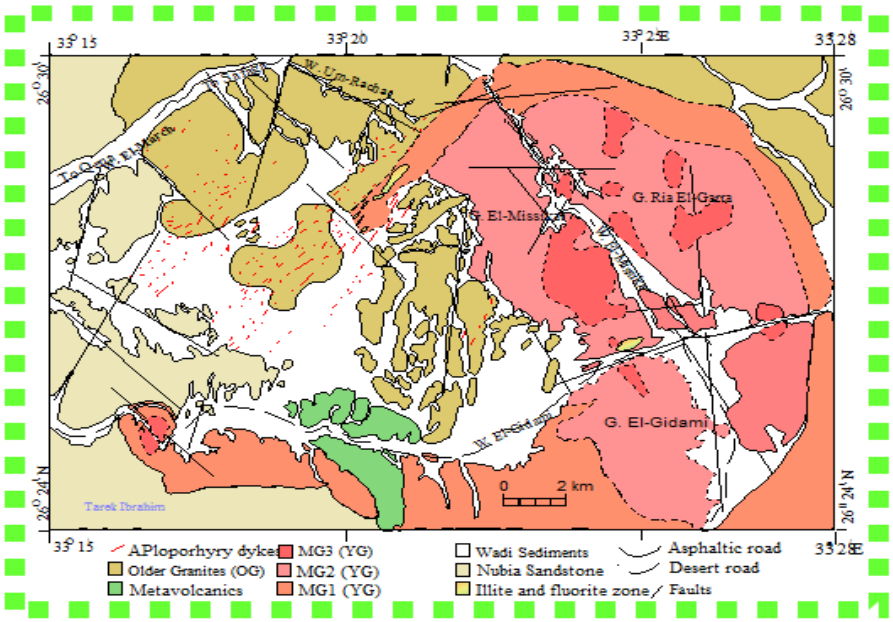


Fig. (2): Location of G. El-Missikat U-Prospect

According toMahdy [3] and to Sayyah,and El Shatoury[4]the black silica veins are occupied by secondary uranium minerals besides fluorite, galena, pyrite, chalcopyrite and molybdenite minerals as well as jasper. In 1995Mohamed[5] has subjected the silica veins to an upgrading procedure followed by analysis of the separated fractions and revealed that theradioactive minerals are: uranophane, uraninite and pitchblende.

Fig. (3): Close view of El Missikaturanium mineralized silica vein

Raslan[6], stated that the uranium mineralizations in El-Missikat occurrence is usually represented by light yellow secondary uranium minerals (mainly uranophane and beta uranophane) (Fig. 4), sparsely disseminated in the shear zones and are usually associated with intense alteration features such as silicification, ferrugination and kaolinization. He detected no uraninite or any primary uranium minerals.

In this regard, it is interesting to refer to the results of acid agitation leaching of El Missikatshear zone achieved by **Mohamed** [5]. She revealed that theoptimized leaching factors involved an acidity of 60 kg H_2SO_4 / t of ore ground to

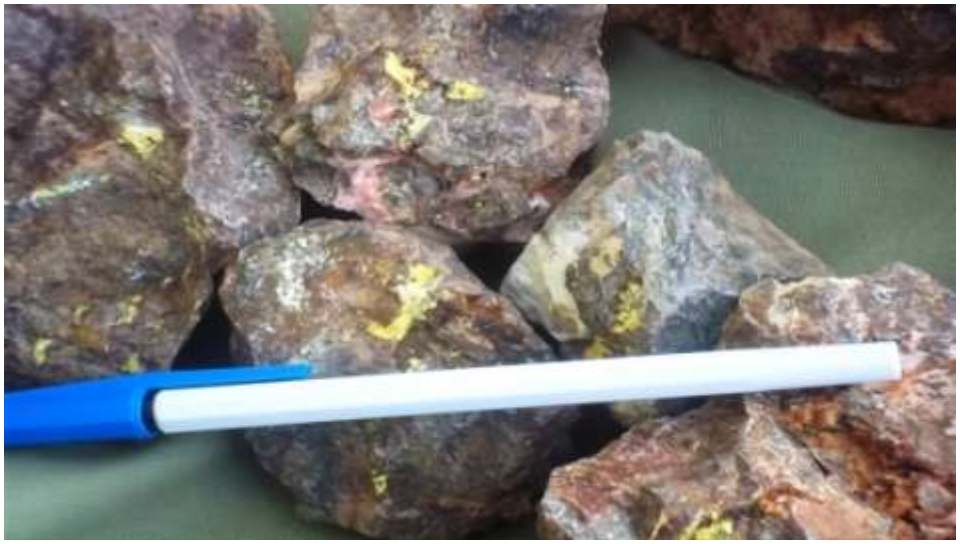


Fig. (4): Visible Uranophane Associated With Black Silica Samples

200 mesh size in a solid/liquid ratio of 1/1 at 60 ^{O}C besides using 20 kg $KClO_3$/t as oxidant for occasional presence of a primary mineral. Under these conditions, the uranium leaching efficiency attained about 91% at 8 hr from El Missikatmineralization. A similar uranium mineralization (uranophane and beta uranophane) in red silica veins has also been found associated with the shear zones of the younger granite of G. Al Aglab to the north of El Missikat occurrence. The agitation leaching parameters of a representative sample for 90% uranium leaching efficiency from Al Aglab occurrence were found to include 30 kg H_2SO_4/t of ore ground to -60 mesh size in a solid/liquid 1/1 for 3 hr agitation time at room temperature **(Awadalla)**[7].

Mousa, et al.,[8], studied the recovery of uranium and REE from El Missikat pluton where they achieved uranium leaching efficiency 95%, REE 70% and iron oxide 14.52% under the optimum parameters: ore particle size 74 µm, sulfuric acid concentration 2.5 M, contact time 240 min., solid/liquid ratio ½, H_2O_2 concentration 0.5 M and agitation rate 600 rpm at 25 ^{o}C temperature.Their obtained data were interpreted with a shrinking core model with diffusion control through a porous product layer.The present work is essentially concerned with uranium recovery from El Missikat mineralization hosted in the recent recorded siliceous vein.The general principle for leaching process, is that the active agents present in a solid matter are extractedand dissolved in a liquid (usually, but not always a solvent), either in nature or through an industrial process. In general, leaching is the process in which inorganic, organic contaminants are released from the solid phase into the water phase under the influence of mineral dissolution[9].

Agitation leaching: Most of the uranium leaching plants applying an atmospheric agitation leaching technique because of its simplicity and its low cost. In this technique, the solid and the leaching acid are agitated by air or a mechanical means for an intimate time at a desired temperature.In the earliest uranium plants, concentration and purification of solutions were achieved by selective adsorption of uranium by an ion exchange resin in fixed-bed columns.

II. EXPERIMENTAL

II.1. Chemical materials and reagents

All chemicals and reagents used were of analytical reagent grade and all solutions were prepared in calibrated flasks and all the solutions and standards used were prepared with de-ionized water.

Uranium determination

Uranium was determined in the pregnant leach solution and the crude uranium concentrate using the oxidimetric titration procedure with a standard solution of NH_4VO_3 till the appearance of a purplish red color represents the end point **(Davies and Gray)**[10]. Uranium concentration in the working sample solution was calculated according to the following equation

U (gm/l) = T.V1.1000/V

T: titration intensity of NH_4VO_3.

2.1.1 Study Sample

A technologicalsample representing a new uranium anomaly of El Missikat uranium occurrence was chosen for this study.It was provided by the field geology team of NMA. They revealed that the secondary

uranium mineral is the main mineral in the study sample.El Missikat mineralized rock sample was firstly subjected to crushing and grinding followed by proper quartering and sieving.

2.2. Leaching Experiments

Studies of acid leaching experiments were conducted to achieve the goal of dissolving maximum uranium constituent under moderate conditions with minimum dissolution of the other undesirable gangues. These experiments were conducted upon El Missikat mineralized rock sample to study the factors affecting uranium leaching efficiency. These factors involved: grain size, acid type, acid concentration, agitation time and leach acid to ore ratio as well as the slurry temperature.

2.2.1. Uranium leaching procedures

In this study the acid agitation leaching technique was chosen as it is the suitable one for the nature of the study sample. The leaching experiments were all studied under the general following conditions: 10 gm sample, 40 gm/l acid concentration, 1/2 S/L ratio, -60 mesh size, at room temperature (25^{o}C) and 2hours contact time which always fixed except the study factor. The relevanteffective uranium leaching factors have been studied namely;the grain size effectwhere the study mineralized sample was first subjected to crushing and grinding followed by proper sieving.The sieved study sample portions were of grain sizes + 18, - 18 + 40, - 40 + 60 and – 60 mesh size.

Effect of acid type: where different types of acids were used in the leaching process under consideration. This factor was studied using 40 gm/l H_2SO_4, HCl and HNO_3 acids. The leaching efficiency percent was calculated according to the following equation:

$$\text{Leaching efficiency,\%} = \frac{\text{Leached metalion conc.}}{\text{Original metal ion conc.}} \times 100$$

- Effect of acid concentration, it was studied to choose the most suitable acid concentration for achieving the maximum leaching of uranium in the studied sample material. Acid concentration of 20, 40, 60 and80gm/l HCl were studied.
- effect of agitation time,agitating time for the leach slurry mixture was studied to choose the most suitable time achieving the maximum leaching of lanthanides. Agitation times of 2, 4, 6, 8 and 10 hours were tested.
- Effect of leaching temperature, this factor was studied using leaching temperature of 25, 60, 70 and 80°C.
- Effect of acid to ore ratiowas studied using ratios of (1:1), (1:2), (1:3), and (1:4).

2.4. Uranium separation

To proceeds the second stage of this work, a stock solution of dissolved uranium was prepared by applying the previously concluded optimum leaching conditions. These conditions were applied on 50 gm of the sample and 250 ml of the study El Missikat mineralized rock sample leach liquor was ready for the next step.Separation ofuranium was achieved using ion exchange resin dry Chinese anion resin (type D263B) which has been specially developed for the extraction of uranium from sulfate solution and have a capacity higher than that of other strong base resins. It has high adsorption rate and its elution is more rapid. About 2 gm of the resin was treated by wetting by 5 ml distilled water and activated by 30 ml of 50 gm/l H_2SO_4.

III. RESULTS AND DISCUSSION

3.1. Chemical characteristics of the sample

The average chemical analysis of Mohamed, 1995 [5] of El Missikat mineralized samplein comparison with that of El Missikat fresh granite is shown in Table 2 while that of the study sample is shown in Table 3.

Table (2): Chemical analysis of El Missikat mineralized silica veins and major fresh granite host (Mohamed, 1995)

Component	El Missikat silica veins, wt%	El Missikat fresh granite, wt%
SiO_2	90.96	75.50
TiO_2	0.13	0.10-
Al_2O_3	2.39	12.50
Fe_2O_3	2.83	1.20
FeO	0.08	-
MnO	0.05	-
MgO	0.19	0.03
CaO	0.7	0.53
Na_2O	0.06	4.01
K_2O	0.27	4.65
P_2O_5	0.01	-
L.O.I.	2.03	-

Table (3): Chemical analysis of the new uranium anomaly of El Missikat mineralized shear zone

Element	Study sample (ppm)	Element	Study sample (ppm)
As	**70**	**V**	**35**
Cd	**5**	**Na**	**2155**
Cs	**10**	**K**	**8420**
Pb	**2075**	**Ca**	**1300**
Se	**135**	**Mg**	**620**
Ag	**-**	**Fe**	**8680**
Sr	**35**	**Ni**	**25**
Zn	**175**	**Mn**	**265**
Cu	**10**	**Ba**	**20**
Cr	**85**	**Co**	**10**
Al	**36840**	**Zr**	**1070**
U	**3030**	**∑REE**	**100**

3.2. Uranium leaching study

3.2.1. Grain size factor

To study the effect of grain size, 4 experiments were performed using 20 gm ore sample portions of grain size ranging from + 18 to – 60 mesh size. Other leaching conditions were fixed at 40 gm/l sulfuric acid concentration, 1/2 solid/liquid ratio for 2 h at room temperature(25 ^{o}C). From the obtained results (Table 4) it is clear that uranium leaching percent has increased from 37 to 75% with decreasing the mesh size from +18 to -60. This can be explained by the fact that by decreasing the grain size, the surface area exposed to the leaching agent would increase and hence the uranium leaching percentage has increased.

Table (4): Effect of grain size upon uranium leaching efficiency

Grain size (mesh)	U leaching efficiency %
+ 18	37
-18 + 40	47
- 40 + 60	53
- 60	75

3.2.2. Effect of acid type

Different acids were studied such as nitric, hydrochloric, and sulfuric acids under experiment conditions of acid concentration 40 gm/l, contact time 2 hours, S/L ratio1:2, at room temperature (25 ^{o}C) and – 60 mesh size. The obtained uranium analysis data revealed that hydrochloric acid (79 %) was the most effective leaching agent but as long as sulfuric acid is economic and available and gave considerable uranium leaching efficiency (75 %), it was thus used in the leaching process (Fig. 5).

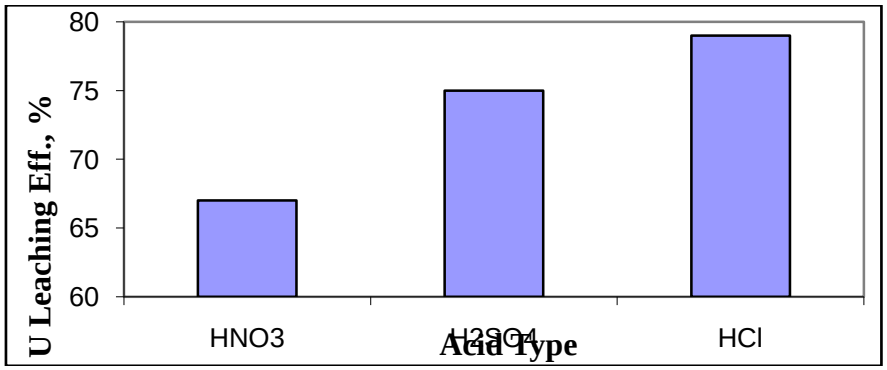


Fig. (5): Effect of acid type upon uranium leaching efficiency from El Missikat mineralized rocks

3.2.3. Effect of acid concentration

Different concentrations of sulfuric acid were used (20, 40, 60and 80 gm/l) as a leaching agent for uranium and the other factors have been fixed at 2 hours leaching time, solid/liquid ratio ½ and25 ^{O}C temperature. The obtained results indicated that a concentration of 100 gm/l gives 80 % uranium leaching efficiency while that of 60 gm/l is 77 % (Fig. 6). It is preferred to continue the following experiments with sulfuric acid concentration with 60 gm/l to avoid element interference at high acid concentration.

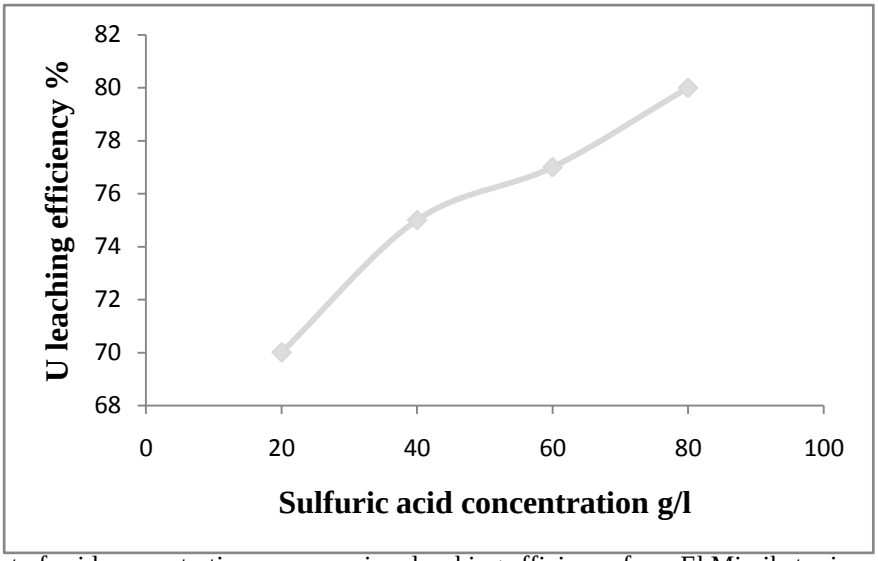


Fig. (6): Effect of acid concentration upon uranium leaching efficiency from El Missikat mineralized rocks

3.2.4. Effect of contact time

Leaching experiments were performed over a range from 2 up to 8 hours. Other variables were fixed at the leaching conditions of 60 gm/l sulfuric acid, 1/2 solid /liquid ratio, at 25 ^{O}C temperature and grain size of – 60 mesh size. The obtained data show that uranium leaching efficiencies increase with increasing contact time achieving its maximum after 8 hours. Increasing time over 6 hourswasfound to be ineffective (Fig. 7).

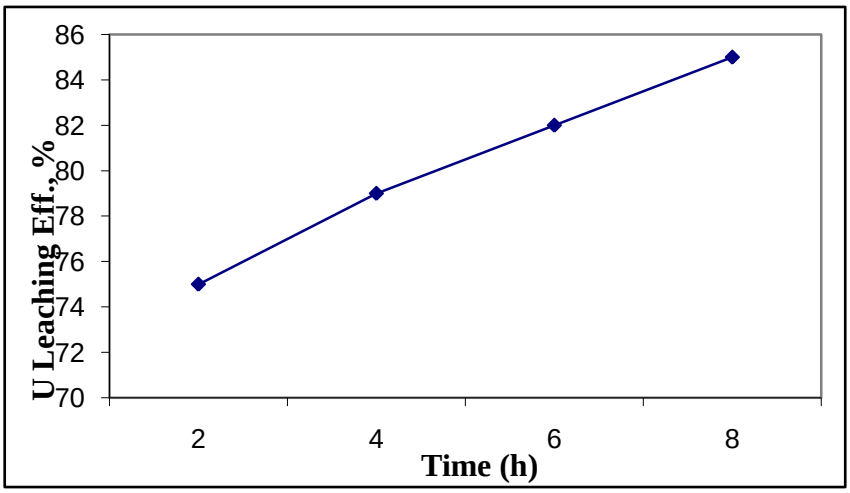


Fig. (7): Effect of contact time upon uranium leaching efficiency from El Missikat mineralized rocks

3.2.5. Effect of temperature

It was necessary to study the effect of temperature of leaching in a series of experiments in which other factors were fixed at 60 gm/l sulfuric acid, 1/2 solid /liquid ratio, 8 h contact time and grain size of – 60 mesh. Effect of temperature covered a range from room up to 80 ^{O}C. From (Fig. 8) uranium leaching efficiency was increased by increasing temperature but from economic point of view temperature 25 ^{O}C was chosen for the next factors.

3.2.6. Effect of Solid / liquid ratio

Although good uranium leaching efficiencies were obtained, it was important to study the effect of solid/liquid ratio upon uranium leaching efficiency.Thus a series of experiments under the following conditions were performed: 8 hours contact time, 60 gm/l sulfuric acid concentration, at temperature 25 ^{O}C and grain size of – 60 mesh. The solid/liquid ratios were varied between 1/1,½, 1/3 and 1/4. Results obtained were plotted in Fig. (9). Henceagitation leaching technique is recommended since the optimum uranium recovery is obtained at 1/4 ratio. From the economic point of view 1/3 S/L ratio was chosen. However, at more dilute pulp a sudden

drop in uranium leaching is noticed, probably because the actual acid concentration is greatly decreased giving the large volume of solution used, in a manner which adversely affected the rate of acid attack.

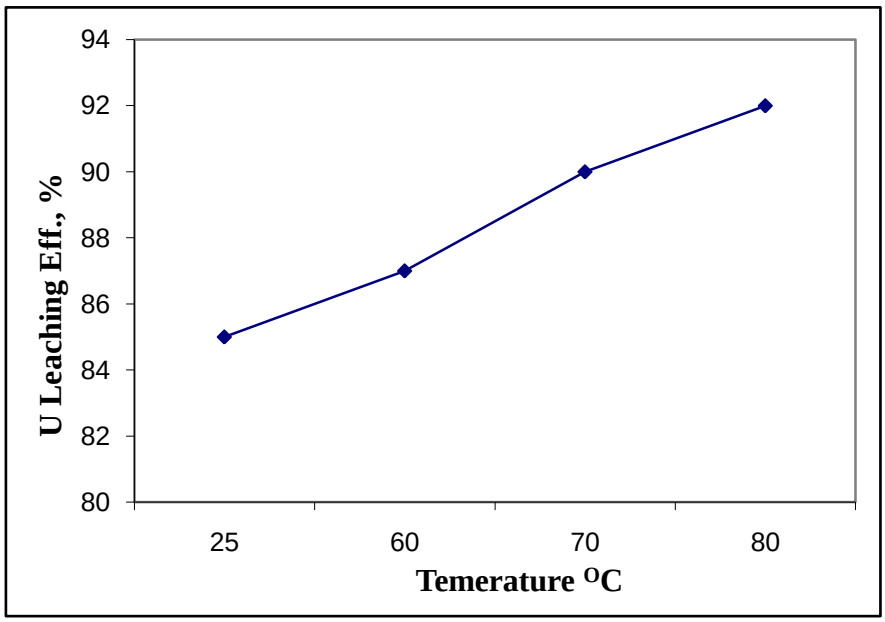


Fig. (8): Effect of temperature upon uranium leaching efficiency from El Missikat mineralized rocks

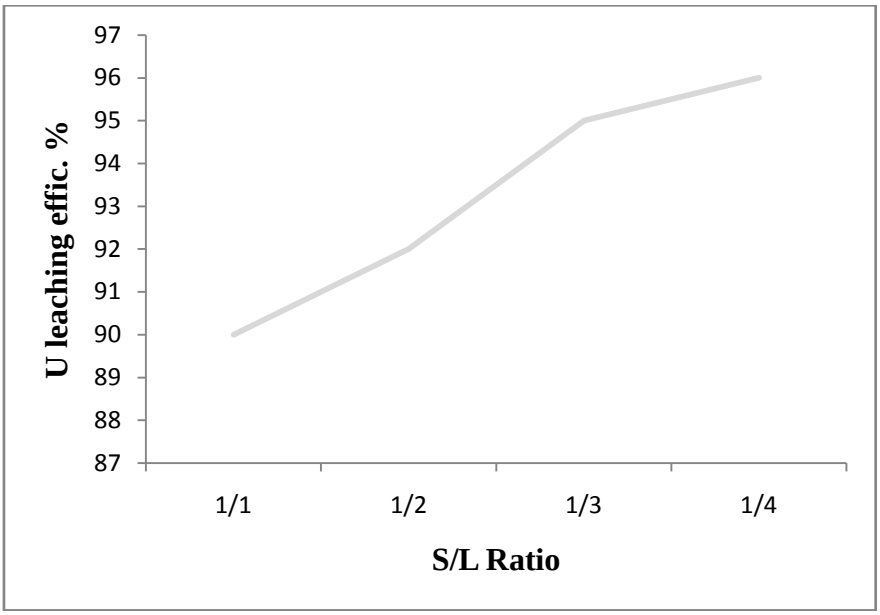


Fig. (9): Effect of solid/liquid ratio upon uranium leaching efficiency from El Missikat mineralized rocks

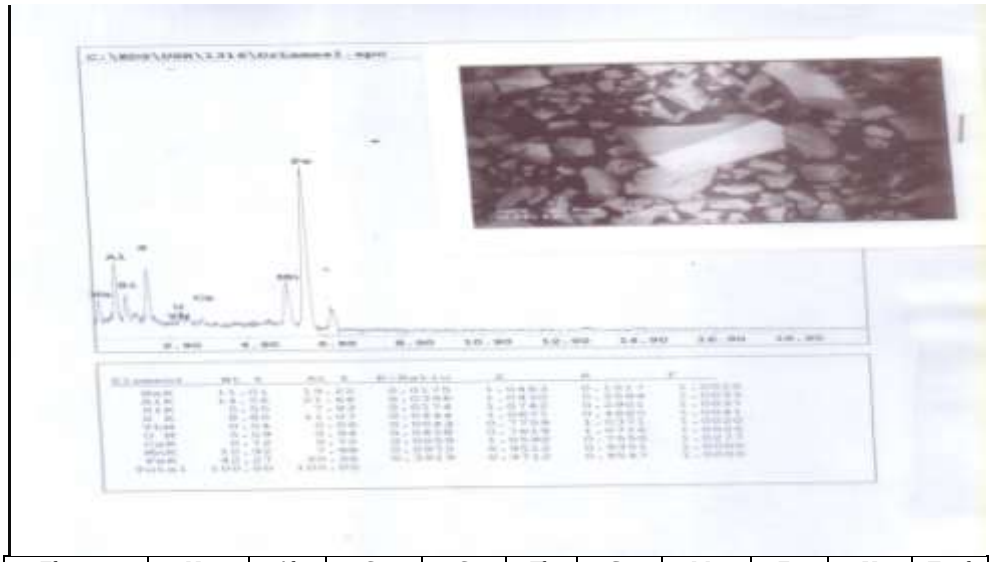
3.3. Uranium separation

A stock solution was prepared for further uranium extraction process by applying the obtained optimum leaching conditions: 8 hours contact time, 60 gm/l sulfuric acid concentration, at temperature 25 ^{O}C, grain size of – 60 mesh and 1/3 solid/liquid ratio (95% leaching efficiency). For this step 250 ml uranium solution was prepared from50 gm sample, this leach liquor was subjected to iron precipitation at pH 3- 3.5 to get rid of undesired element which always associates uranium in the Yellow Cake product (Fig. 10).

Uranium adsorption

Separation ofuranium was achieved using ion exchange resin dry Chinese anion resin (type D263B) which has been specially developed for the extraction of uranium from sulfate solution and have a capacity

higher than that of other strong base resins. It has high adsorption rate and its elution is more rapid. About 2.5gm of the resin was treated by wetting by 5 ml distilled water and activated by 30 ml



Element	Na	Al	Si	S	Th	Ca	Mn	Fe	U	Total
Content wt.%	11.01	14.56	5.55	8.85	0.54	0.72	10.92	42.27	5.59	100

Fig. (10): ESEM micrograph and the corresponding EDX spectrum of the iron precipitate

Of 50 gm/l H_2SO_4. Uranium leach liquor was adjusted to pH 1.7-2 before loading on the resin then about 250 ml (575.7ppm U) of the leach liquor was allowed to pass through the resin under the conditions applied by **Mahmoud et al.,** [11] where 11 bed volumes each 20 ml, loading efficiency 81.5 % were obtained (Fig. 11).

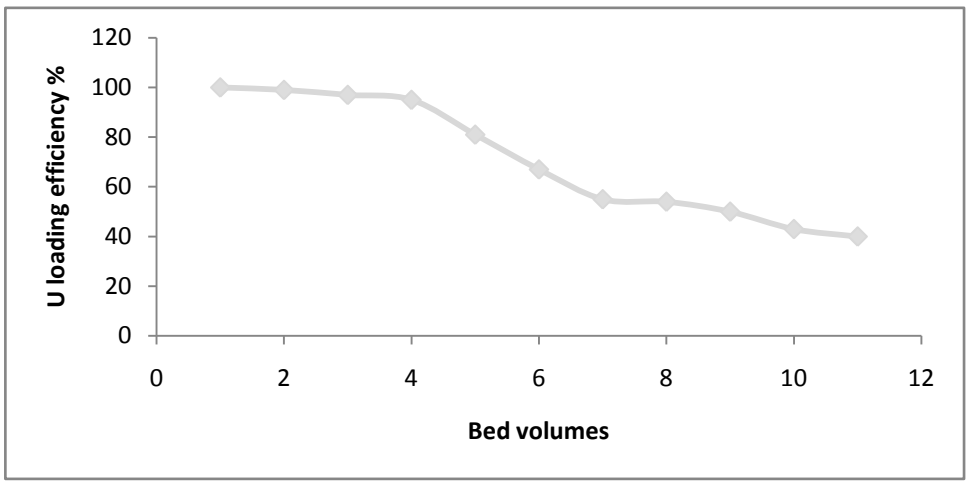


Fig. (11): U loading efficiency present on the ion exchange resin

Uranium elution

Elution of the adsorbed uranium was then performed by an acidified (H_2SO_4)sodium chloride (1 M) and its pH was adjusted to 1. Along 12 bed volumes each 10 ml the eluted solution was obtained and their uranium elution efficiency % was estimated as 89.67% (Fig. 12).

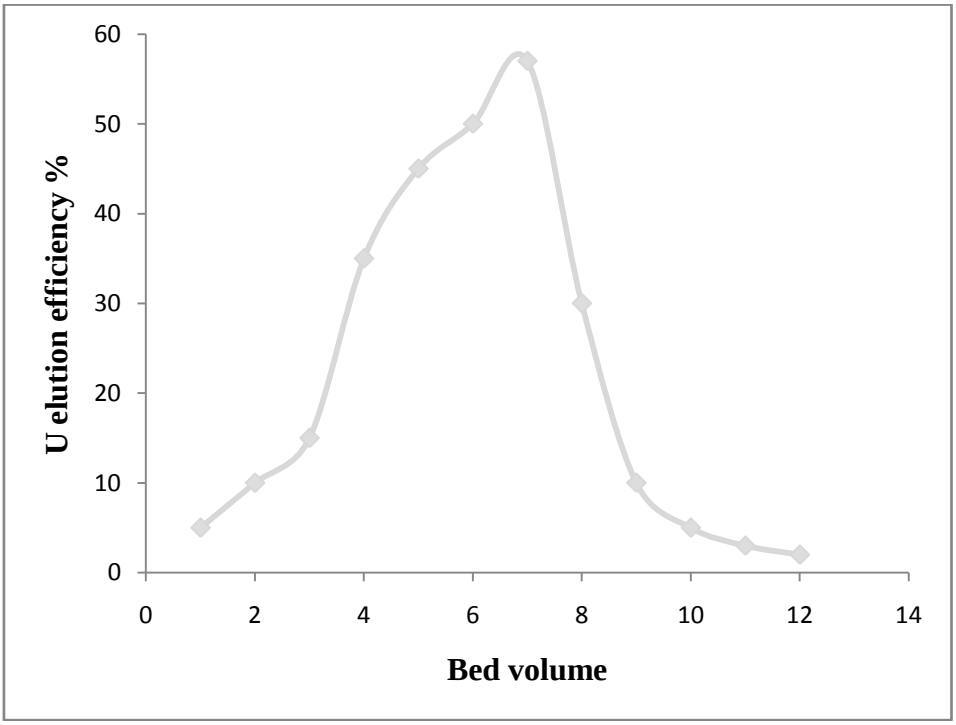
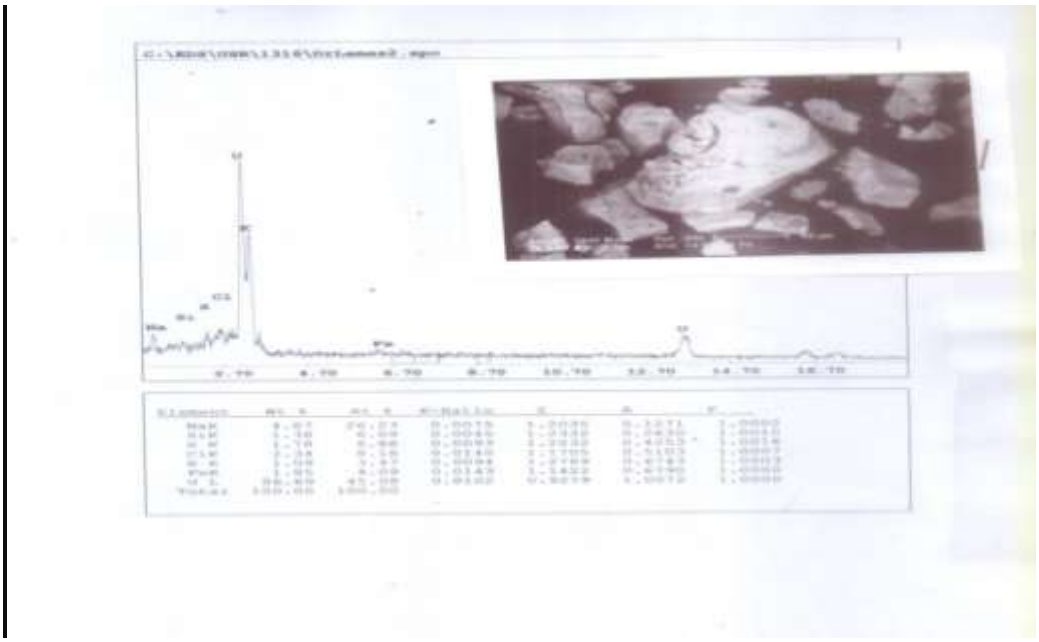


Fig. (12): U elution efficiency present on the ion exchange resin

Uranium precipitation

Precipitation of uranium was achieved at pH 7.5 by adding Na OH solution gradually to the elute solution rich in uranium concentration where 0.27 gmof sodium diuranatecrystals were precipitated (Fig. 13).A technical flowsheet (Fig. 14) was proposed summing up the practical procedures for uranium recovery from El Missikat mineralized shear zone.



Element	Na	Si	S	Cl	K	Fe	U	Total
Contentwt.%	4.87	1.38	1.78	2.34	1.09	1.85	86.69	100

Fig. (13): ESEM micrograph and the corresponding EDX spectrum of the sodium diuranate precipitate

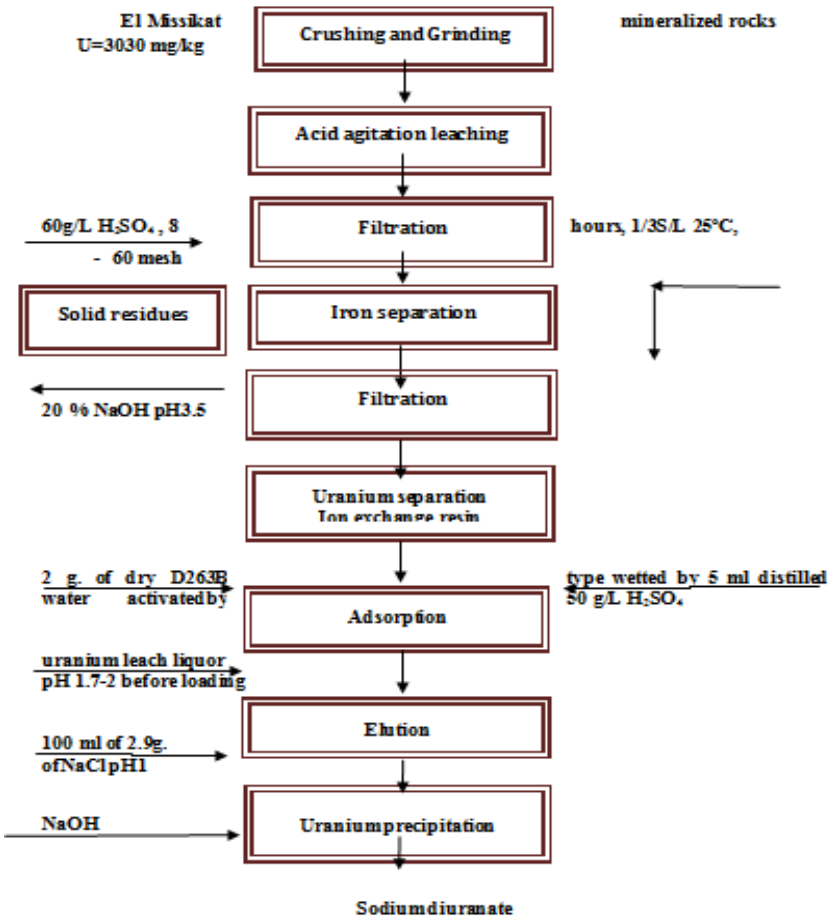


Fig. (14): Flow sheet for uranium recovery from El Missikat mineralized shear zone

IV. CONCLUSION

The present work concluded that recovery of uranium from the recently discovered siliceous shear zone of Jabal El Missikat, Eastern Desert of Egypt is feasible. This study revealed that the optimum leaching factors for uranium are:8 hours contact time, 60 gm/l sulfuric acid concentration, at room temperature (30^{O}C), grain size of – 60 mesh and 1/3 solid/liquid ratio (95% leaching efficiency).

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الملخص العربى

إن نشاط هيئة المواد النووية يتمثل فى التنقيب واستكشاف وتقييم احتياطيات خامات اليورانيوم ، وكذلك الدراسات الكيميائية التكنولوجية على الواعد منها. إن تمعدن اليورانيوم قد سجل فى عديد من نطاقات التكسير فى الجرانيت الحديث لكل من جبلى المسيكات والعرضية، ويقعان جنوب طريق قنا – سفاجا الأسفلتى عند الكيلو85 حيث يقع جبل المسيكات على بعد 3 كيلومتر من علامة الكيلو المذكور. أما جبل العرضية فيبعد 25 كيلومتر عن جبل المسيكات ويحدد الجبلان بخطى عرض N 26°15′ to 26°33′ وطول E 33° 30\ to 33° 15\.

وقد تمت دراسة إذابة واستخلاص اليورانيوم على الدراسة من نطاق التكسير المتمعدن والمسجل حديثا بجبل المسيكات. نتيجة لوجود معدن اليورانيوم الثانوى (يورانوفين) فقد كان تركيز اليورانيوم3030 جزء فى المليون، ومن خلال عملية الإذابة تم دراسة العوامل المؤثرة على كفاءة إذابة اليورانيوم مثل: حجم الحبيبات، نوع الحامض، تركيز الحامض، زمن التقليب، درجة الحرارة و نسبة الصلب للسائل.

وقد دلت النتائج المستخلصة إلى أن العوائل المثلى لإذابة اليورانيوم هى: - 60 مش حجم حبيبات، 60 جرام/لتر تركيز الحامض، 8 ساعات زمن تقليب، درجة حرارة25 مئوية ونسبة صلب إلى سائل3/1، وكانت كفاءة الإذابة95 %.

أما عملية استخلاص اليورانيوم فقد تمت على المحلول المحضر من تطبيق العوامل المثلى المتحصل عليها من عملية الإذابة، وكانت التقنية المستخدمة هى التبادل الأنيونى الراتنجى بالنوع الصينى الجاف (D263B). وبعد عمليتى التحميل والإزاحة لليورانيوم من الراتنج تمت عملية الترسيب من المحلول الناتج برفع الرقم الهيدروجينى 7.5 باستخدام محلول هيدوكسيد صوديوم لنحصل على(gm 0.27) راسب يورانات الصوديوم (الكعكة الصفراء).