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Successive leaching of uranium and associated metal values from Shale ore material of Allouga locality, Um Bogma Formation, SW Sinai, Egypt

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Abstract: The chemical processing of Allouga locality ore material at Um Bogma Formation, occurring at southwestern (SW) Sinai, Egypt, was studied for the recovery of uranium and other associated elements. After the completed determination of optimum leaching conditions of uranium, the spent ore material left behind was subjected to another leaching process of REEs and Cu. Concerning uranium leaching using sodium carbonate as a leaching agent, it was thus possible to achieve leaching efficiency exceeding 95%. This was followed by the second procedure for REEs and copper leaching from the spent left behind using citric acid. The obtained leaching efficiency was about 90% of REEs and 95 % of copper content. The leached metal values namely; U, REEs, and Cu from the study ore material were then recovered as marketable products. These conditions were applied to technological sample assays of 6% CuO; 3500, ppm U, and 2000 ppm REEs.

Keywords: Uranium recovery; Um Bogma Formation; Leaching Efficiency; Rare Earth Elements.

1 Introduction

An exploration program of the Nuclear Materials Authority for uranium led to the discovery of several uranium mineralizations in different areas. Associated valuable elements (e.g. Th, V, Mo, Cu, REEs,…etc) are another concern of the Nuclear Materials Authority. The most important locality of uranium mineralization is Um Bogma Formation at the east Abu Zeneima area, southwestern Sinai between longitudes 33º 20` to 33º 25` E and latitudes 29º 00` to 29º 05 N was found to be irregularly mineralized. The present work was applied at this locality due to its added values of both Cu and U in relatively reasonable values. Declare that the localities of Um Bogma Formation are of good uranium potentiality and presence with some associated elements including Cu, V, and REE besides the famous Fe– Mn deposits in different grades in several localities and rock facies of this formation; such as Allouga, Talet Selim, Abu Thor, etc [1, 2].

An interesting rare metal mineralization was recorded at the Allouga locality of the Abu Zienema area, southwestern Sinai, Egypt. This mineralization is mainly associated with different rock facies that are mainly represented by shale, siltstone, clay, ferruginous sandstone, calcareous sandstone, and feldspathic sandstone [3].

Allouga environs, West Central Sinai, are bounded by lat. 28° 58' – 29° 03' N and long. 33° 21' and 33° 26' E, covering about 100 km^2 (Fig.1). These environs are covered by late Proterozoic basement rocks overlain non-conformably by Paleozoic rock succession. Alloga calcareous shale facies of Um Bogma Formation is considered as one of the most important occurrences of U minerals besides the associated economic elements e.g. REEs, B, V, Co, Ni, Zn, Cu,..etc [1].

The processing of uranium ore in recent times has depended upon the grade and type of the deposits. In the hydrometallurgical methods, leaching is the chief operation, thus many efforts are accomplished to advance extraction percent with reasonably priced facets. Alkaline leaching of uranium ores depends on the uranyl ion which can formed as a stable soluble complex with carbonate species; namely the uranyl tricarbonate $[UO_2(Co_3)_3]^{4-}$.

The advantage of uranium leachability via the alkaline method gives a reasonably pure uranium leach liquor and directly recovers the product without any additional purification. The leach solution could be recycled for further use in the leaching process [4, 5]. The chemical process equations of uranium via alkaline reagents can summarized as follows:

$$UO_3 + 3Na_2CO_3 + H_2O = Na_4UO_2(CO_3)_3 + 2NaOH$$

$$UO_2 + 3CO_3^{2-} + H_2O + 0.5O_2 \rightarrow [UO_2(CO_3)_3]^{4-} + 2OH^-$$

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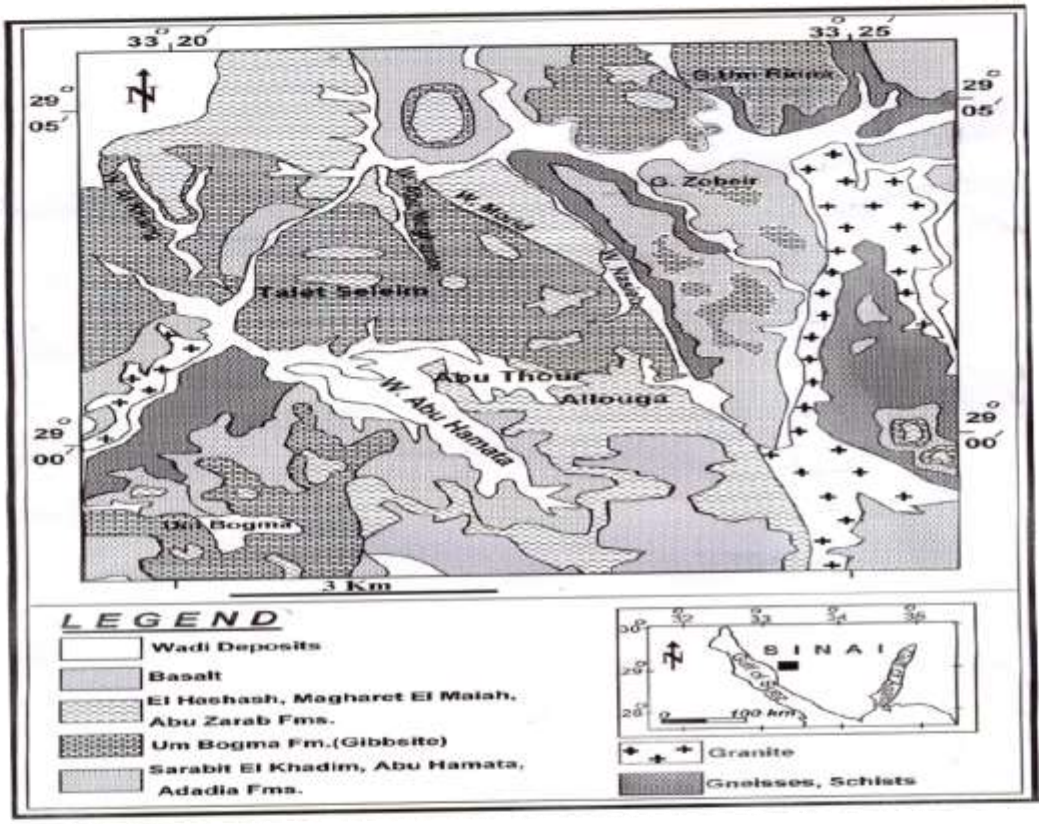

Fig. 1: The geologic map of Um Bogma locality, After [1].

Copper ores could be leached via alkaline reagents such as; carbonate or bicarbonate of sodium or ammonium and/ or ammonium hydroxide. Copper dissolution reactions using ammonia and ammonium reagents as the mathematical following reactions:

$$2CuCO_{3}.Cu(OH)_{2} + 12\,NH_{3} \longrightarrow 3[Cu(NH_{3})_{4}]^{+2} + 2CO_{3}^{-2} + 2OH^{-}$$

Nevertheless, a mixture of ammonium neither carbonate nor hydroxide is used to accelerate the reaction as the following equation:

$$Cu_{2}(OH)_{2}CO_{3} + (NH_{4})_{2}CO_{3} + 6\,NH_{4}OH \longrightarrow 2\,Cu(NH_{3})_{4}CO_{3} + 8H_{2}O$$

From the above reactions, it is obvious that copper dissolution is due to the formation of the Cu ammoniated complex and/ or copper tetra-ammine complex. Nevertheless, in the absence of ammonia, Cu would be dissolute as its bicarbonate.

The citric acid (neutral and deprotonated) may act as a pro-oxidant producing Cu(I) which is one of the important reactants of the Haber-Weiss reactions [6]. Thus, it is important to analyze the following reaction:

$$[\text{Citric-xH}]^{-x} + [Cu(H_{2}O)_{4}]^{2+} \rightarrow [\text{Citric-xH}]^{-x-1} + [Cu(H_{2}O)_{4}]^{1+}$$

It is important to mention herein that, citric acid can react with lanthanide according to [7] to give the following complex $[La(C_6H_5O_7)(H_2O)]n$. The reaction of La_2O_3 and citric acid monohydrate and water at pH 2.2-2.5 led to the formation of the homonuclear and or polymer under hydrothermal solution to get a good yield. This is also due to the stability constant of lanthanide and citrate. Citric acid contains three carboxylic groups, and three hydrogen dissociation constants pKa1 = 3.13, pKa2 = 4.76, and pKa3 = 6.40 at 25 °C. [8].

The main target of this work is to leach valuable elements e.g. uranium and copper from the shale sample of Um Bogma Formation in the Alouga area. In this context, many efforts have been made using mineral acid especially sulfuric acid as a leaching agent and have proven that both metals (U&Cu) are amenable to acid leaching. Conversely, this has been achieved through the high cost of excessive consumption of acid [9, 10, 11 and 12]. This is due to the carbonate content of these ore materials is generally very high up to 40-50%. Some other researchers have produced many efforts to leach both metal values via alkaline leaching [13, 14, and 15].

A representative sample assaying 6% CuO; 3500, ppm U, and 2000 ppm REEs was collected from the shale ore material of the Allouga locality. To avoid high consumption of mineral acid and to separately leach liquor Cu. Carbonate solution for uranium on one hand. Citric acid is an organic acid for REEs and copper leaching process on the other hand upon the residue left behind.

This work is oriented towards the leaching of uranium, REEs, and Cu from the Shale of Allouga via successive leaching. In the first, sodium carbonate was used as a selective agent for uranium leaching. the work then shifted to treat the residue lifted behind by citric acid for leaching of the associated REEs and copper. To realize this goal, a technologically representative sample of Allouga shale was properly collected with assays of 6% CuO; 3500, ppm U, and 2000 ppm REEs.

2 Experimental

2.1. Material characterization

The collected sample from Allouga Cu-U mineralization (about 250 g) was ground. Suitable quartering of the technological sample was achieved after its grinding to less than - 200 mesh size to obtain a typical representative sample. The latter was subjected to comprehensive chemical analysis namely; major and trace elements. The conventional wet chemical technique for the former was practical **[**16**]** where Al_2O_3, SiO_2, P_2O_5, and TiO_2 were determined using spectrophotometric methods while K_2O and Na_2O were examined via flame photometric technique. Total iron as Fe_2O_3, MgO, and CaO were determined using titration methods against dichromate and EDTA solutions respectively. The loss on ignition (L.O.I) was gravimetrically determined at 1000°C for CO_2. The error for these constituents is not more than ±1 %.

The trace elements V, Cr, Ni, Cu, Zn, Ga, Rb, Sr, Y, Zr, Nb, Pb, and Ba were also analyzed using the X-ray fluorescence technique (XRF) Philips Unique II unit fitted with an automatic sample changer PW 1510 (30 position), connected to a computer system using X-40 program for spectrometry. The detection limit of the measured elements by the XRF technique was estimated to be 5 ppm.

2.2. Leaching procedures

Many alkaline dissolution experiments uranium have been done using altered reagents such as; carbonate and bicarbonate of ammonium and sodium or else in a manner to enhance the applicable leaching factors. In these experiments, a suitable weight of the working U ore ground to -200 mesh size was mixed with the accurate volume of the reagent solutions of numerous concentrations. The concerned mixture was then agitated at a suitable temperature for a fixed time. The treated slurry was subjected to a filter process and the residue left behind was carefully washed with distilled water and filtrate and washings were made up to volume before analysis**.**

The spent sample free from uranium was subjected to another leaching process for both REEs and Cu content by citric acid. Several series of experiments were carried out to study the effects of citric acid concentration, S/l ratio, contact time, and dissolution temp.

2.3. Control analysis

To follow the dissolution and recovery efficiencies of the concerned experiments, the various stream solutions were subjected to U and Cu analysis. For U the oxidimetric method titration against ammonium meta-vanadate was used after the reduction of uranium **[**17**].**

For the copper, an atomic absorption spectrophotometer (AAS) Unicam 969, England was used. A spectrophotometric method was applied where REEs form a stable colored complex with Arsenazo III in weakly acidic media. In this method, the pH is adjusted at about 2.6 and the absorbance is measured at wavelength 650 nm against a reagent blank solution **[**18**]** using a double-beam UV-visible Shimadzu (model 160A) spectrometer.

3 Results and Discussion

3.1. Material characteristics

The essential chemical analysis of the major content of the concerning technological ore sample is tabulated in Table (1). From the mentioned table, it is evident that besides a loss on ignition of up to 6.47 % indicating a mild carbonate content, the ore material assays 51.51% SiO_2, 6.02 % Fe_2O_3, and 9.78% Al_2O_3. On the other side, another part of the technological sample was investigated by X-ray fluorescence for the analysis of some trace elements (Table 2). The latter shows that U attains 3500 ppm besides interesting values of Cu, Ni, Y, and Ce; namely, 6068, 126, 869, and 1090 ppm respectively.

Table 1: The chemical analyses of the working Allouga Shale ore sample.

Component	wt.%	**Component**	wt.%
SiO_2	51.51	K_2O	1.12
TiO_2	0.24	P_2O_5	0.24
Al_2O_3	9.78	Na_2O	0.42
Fe_2O_3	6.02	L.O.I .(1000^0C)*	6.74
MgO	2.28		
CaO	2.9	Total	80.83

*L.O.I. = loss on ignition

Table 2: XRF analytical results of trace elements of the Allouga Shale ore sample.

Trace element	Ppm	Trace element	ppm
Ba	714	Pb	410
CuO	6068	Cr	54
Ni	126	Sr	274
Nb	31	Ga	17
Zn	714	Zr	321
Ce	1090	U*	3500
Y	869		

*Chemically analyzed.

3.2. Results of uranium carbonate leaching

3.2.1. Effect of different reagents

The effect of different alkaline leaching reagents, urea, and citric acid upon uranium, copper, and REEs dissolution efficiencies has possibly been studied. During these experiments, the other leaching factors were applied at -200 mesh ore size, 180 min. contact time, 70℃ leaching temperature, and 1/3 solid-liquid (S/L) ratio as well as the leaching agents concentration is 150 g/L. The obtained leaching efficiencies of different metals value are tabulated in Table (3) and have shown that the best chemical reagent is Na_2CO_3 with a concentration of 150 g/L.

Table 3: Effect of different reagents upon metal values leaching dissolution of the working Allouga technological ore sample.

Reagents	**Leaching efficiency,%**		
	U	**Cu**	**REEs**
Na_2CO_3	95	33	nil
Na HCO_3	80	63	nil
Citric	75	70	nil
Urea	32	85	nil

From Table (3) it can be indicated that using Na_2CO_3 reagents at 70℃, a leaching efficiency of 95 and 33 % for uranium and copper was respectively realized. The latter has thus been used for studying the other applicable parameters that have been examined in this study; namely the concentration of the carbonate reagent, the leaching temperature, time, and the solid-liquid ratio.

3.2.2 Effect of Na2CO3 concentration

The effect of Na_2CO_3 concentration upon U and Cu leaching or dissolution efficiencies was examined between 100 and 250 g/L , while the other leaching conditions were carried out -200 mesh size, 180 min. agitation or contact time, 70℃ leaching temperature and 1/3 solid-liquid (S/L) ratio. The leaching efficiencies of uranium which are tabulated in table (4), indicate that the best conc. is 150 g/L for uranium leaching. Under these conditions, the leaching efficiency of copper has attained 33% within uranium dissolution efficiency achieved of 95%.

Table 4: Effect of Na_2CO_3 concentration upon Cu and U leaching efficiencies of the working Allouga technological ore sample.

Na_2CO_3 concentration, g/L	**Leaching efficiency,%**	
	U	**Cu**
100	80	22
150	95	33
250	98	39
270	100	46

3.2.3. Effect of temperature

The alkaline leaching experiments by Na_2Co_3 was used to examine the effect of temperature upon uranium and copper leaching efficiencies from room (about 25 ℃) up to 90 ℃. In these mentioned experiments, the leaching conditions were stable at -200 mesh size ore material, Na_2CO_3 of 150 g/L concentration for 180 min. contact time and using S/L ratio of 1/3. The resulted data are shown in table (5), in comparison to that results at 70℃ expose actually the adequate of temperature to attain a high leaching efficiency of uranium. At room temperature within fixed conditions as above did not leach more than 27 % of Cu while that of U to only13 %. Increasing the leaching temperature to 50℃ has led to increase the dissolution efficiency of Cu and U to 25 and 66 % respectively. Also, its additional increase to 70℃ increased the Copper leaching efficiency to 33% while that leaching of U increased to 95% and after that there has decrease leaching efficiencies at 80 and 90 ℃. Thus optimum leaching temperature of uranium under the above mentioned conditions would be 70℃.

Table 5: Effect of leaching temperature upon U and Cu leaching efficiencies of the working Allouga technological ore sample.

Temperature, ^{o} C	**Leaching efficiency,%**	
	U	**Cu**
25	27	13
50	66	25
70	95	33

80	80	45
90	75	56

3.2. 4. Effect of agitation time

The effect of leaching or dissolution time on Copper and uranium leaching efficiencies was investigated in the range from (60 to 180) min. the other leaching parameter was fixed at 150 g /l concentration of Na_2CO_3, -200 mesh size fitness, a leaching temperature of 70 ^{0}C, and using a solid/liquid ratio of (1/3). The obtained data given in Table (6) shows that a high uranium leaching efficiency of 95% occurs within the experiment of 120 and 180 min. they are associated with about 12 and 33 % copper leaching efficiency respectively. Thus, it can be decided that 120 minutes is suitable for leaching 95% of uranium and only 12% of copper.

Table 6: Effect of leaching time upon Cu and U leaching efficiencies of the working Allouga technological ore sample.

Time/minutes	**Leaching efficiency,%**	
	U	**Cu**
60	70	6
90	82	8
120	95	12
180	95	33

3.2. 5. Effect of the solid/liquid ratio

To study the concerned effect of the S/L ratio on uranium and copper dissolution efficiencies was studied using 1:2, 1:3, and 1:4 solid/liquid ratios, while the other testing at 150 g /l concentration of Na_2CO_3, -200 mesh size fins, a leaching temperature of 70 ^{o}C. Data in Table (7) emphasized that beyond the 1/3 S/L ratio, only a slight increase in the leaching efficiencies of uranium has been achieved. Accordingly, a solid/liquid ratio of 1/3 would be considered the optimal ratio at which the leachability of uranium attained 95 % and only 12% of copper.

Table 7: Effect of leaching time upon Cu and U leaching efficiencies of the working Allouga technological ore sample.

S/L ratio	**Leaching efficiency,%**	
	U	**Cu**
1:2	70	6
1:3	95	12
1:4	98	24

The obtained data of leaching factors of the working sample concluded that the optimum leaching conditions for dissolving about 95% of U and only 12 % of copper would be summarized as follows:

Grain size: -200 mesh.
Na_2CO_3 concentration: 150 g/L
Leaching time: 120 minutes
Leaching temperature: 70 ℃.
Solid/liquid ratio: 1/3

3.2.6 Results of copper and REEs leaching from the residual lifted behind

Previous works have successfully dissolved U from the shale of the Allouga locality by applying alkaline leaching using sodium carbonate. From the latter process, the remaining two metal values; 88% of copper and all REEs were left behind in the spent residue. The latter represents the feed material of the present work for Cu (5339 ppm) and REEs (2000 ppm). The Lns (REEs) and Cu left behind were leached by using citric acid.

3.3. Acidic leaching of REEs and copper

3.3.1. Effect of citric acid concentration

The effect of citric acid concentration on the leaching dissolution of REEs and Cu from spent ore residue nearly free from uranium was investigated between 50 and 200 g/L citric acid while fixing the other dissolution conditions at a contact time of 2.5 h and S / L ratio 1/3 at ambit temperature. The results shown in Table (8) show that the best citric acid concentration was 150 g/L for leaching about 85 % of REEs and 80 % copper content. At the high acid concentration of 200 g/L, REE dissolution efficiency was decreased to about 70 and 75 % for copper and REEs respectively.

Table 8: Effect of citric acid concentration upon REEs and Cu leaching efficiencies, %.

Citric concentration, g/L	**Leaching efficiency,%**	
	Cu	**REEs**
50	25	9
100	65	60
150	80	85
200	70	75

3.3.2. Effect of stirring time:

A series of experiments has been done to study the effect of stirring time or contact time upon REEs and copper leaching efficiencies, these leaching experiments have been accomplished by changing the stirring time between 60 and 180 min. The other associated leaching conditions were stable at acid conc. of 150 g/L citric acid , solid / liquid ratio of 1:3, at ambit temperature. The REEs and copper leaching efficiencies are represented in Table

(9). From the obtained results, it is noticed that by increasing the stirring time, the leaching efficiency of REEs and copper was increased and reached its maximum value at 180 min achieving 90 and 95 % for REEs and copper respectively. Increasing the leaching time to 240 min., there is no marked increase in the leaching efficiency of REEs or copper.

Table 9: Effect of stirring time upon REEs and Cu leaching efficiencies, %.

Effect of stirring time/min	Leaching efficiency,%	
	Cu	REEs
60	45	40
120	70	81
180	95	90
240	97	92

3.3.3. Effect of the solid/liquid ratio

To study the effect of solid/liquid ratio (S/L ratio) on leaching efficiencies of REEs and copper several experiments were done at the solid/liquid ratio (S/L) ranges from 1:3, 1:4 to1:5 at the stable conditions of 150 g/L citric acid, stirring time of 3 hours at room temp., the attained results (Table 10) indicated that, at the S/L ratio of 1/3 is the optimum solid/ liquid ratio which achieved 90 and 95 % for REEs and copper respectively, only slight increase in the leaching efficiencies of REEs takes place by increase solid/ liquid ratio to ¼ and 1/5.

Table 10: Effect of S/l ratio upon REEs and Cu leaching efficiencies, %.

Solid /liquid ratio	Leaching efficiency,%	
	Cu	REEs
1:1	39	34
1:2	84	81
1:3	95	90
1:4	97	92
1:5	98	93

Finally, from the foregoing acidic leaching study, it can be concluded that the optimum leaching conditions for dissolving 90% of REEs and 95 of copper content would be summarized as follows:

Citric acid concentration:	150 g/L
Stirring time:	180 min.
Temp.:	room
S/L:	1/3

4 Conclusions

Appropriate uranium selective leaching using the Na_2CO_3 agitation process was applied for the Allouga ore sample. The obtained leaching conditions for leaching about 95% U and 12% Cu were 150g/L Na_2CO_3 concentration, 120 min as leaching time at 70 °C and 1/3 S/L ratio. In this context, the remaining two metal values; 88% of copper and all REEs were left behind in the spent residue and successfully leached using 150 g/L citric acid, 180 min. at room temperature and S/L =3/1 with leaching efficiencies of 90% of REEs and 95 of copper.

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