Techno-economic study on reagents consumption during uranium leaching, a case study: Gattar pilot plant, Egypt

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Research Article

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Abstract

The techno-economical parameter must be considered during the decision-making stage of project development. In Gattar, results of column leaching indicate that 21.2 and 32g of acid are consumed using an acid concentration of 20 and 40g/l respectively to have a 1% increase in the uranium leachability. Also, 1.0 and 1.8 l solutions using 0.1:1 and 0.2:1 sprinkling intensities are consumed respectively to have a 1% increase in the uranium leachability during the column leaching period. The vat leaching indicates that the 4.1m³ solution and 88kg acid are consumed to have a 1% increase in the uranium leachability after 90 days.

Introduction

Several authors have investigated the geology of the uranium occurrences in Gabal Gattar [1-5]. One of the most common uranium occurrences in the Gattar region (G-V uranium occurrence), which corresponds to perigranitic vein-type deposits in terminology, is found in the Hammamat sedimentary rocks. The other uranium occurrences are associated with granite, whereas the G-II uranium occurrence (Fig.1) is the first in magnitude for exploration and mining. Secondary uranium minerals are found in alkali feldspar granite as stains or filling micro-fractures. Metasomatic alterations are developed in some cases synchronously with the deposition of uranium mineralization. They represented by hematitization, kaolinitization, silicification, episyenitization of the granite, carbonatization and manganese oxides. The uranium deposits are ascribed to the uranium-thorium economic type and also have an economic significance as sources of rare-earth elements.

Mineralogically, many researchers working on the mineralogy of the uranium deposits in G-Gattar area have concluded that the primary uranium minerals are represented by pitchblende (Fig.2a) and uraninite. While the secondary uranium minerals are mainly represented by the two dimorphous minerals uranophane and beta uranophane [CaO(UO₂)₂(SiO₃)₂·5H₂O] (Fig.2b). Both minerals of the same chemical composition are of quite similar physical properties [6], while mostly results from the alteration of uranite occurs predominantly in the form of massive and plate like soft grains with different grades of the yellow color and which turns to brownish yellow when it is stained by hematite. In addition to, minor amounts of the secondary uranium minerals have been identified including clarkiet, zippiete, soddyite, and kasolite beside some trace amounts of primary mineral uraninite [7, 8].

Several countries have turned to heap leaching technology for low-grade uranium mineralization due to the decline of high-grade uranium mineralization [10-13]. Taylor 2009 describes how uranium heap leaching is becoming more common, with plans to use alkaline leaching on low-grade calcium carbonate-rich materials in the future [14].

Since many developing countries have large reserves of economic minerals, mining and extraction processing are the primary source of national income in those countries, piles leaching technology is becoming more common. As a result, finding cost-effective ways to extract these minerals, especially
uranium, was critical. Because of its low capital cost and technological simplicity, heap dissolving is a good option. Many counters, like Peru, Mexico, and Zambia are in this situation [15, 16],

There are various advantages and economic benefits to heap leaching. Low capital and operating costs [17], the absence of a milling step for crushing and agglomeration, the simplicity of leaching equipment’s, the possibility of processing low grade ores, wastes, and considering the absence of a liquid-solid separation step for counter-current operations [18].

The flow sheet for the leaching stage is very simple, in which solutions of high uranium concentration suitable for the ion exchange extraction process are directed to PLS tanks, and liquids of low concentration of uranium are directed to (ILS) tanks for recycling over the piles [19]. The solutions (effluents) from the ion exchange (adsorption) extraction process are recycled and irrigated with fresh solutions on the surface of the ore pile.

An intermittent liquid addition technique is preferred to improve the efficiency of uranium dissolution by leaching [20]. Continuous addition is punctuated by a pause, accompanied by the addition of a new carrier solution or liquid in periodic leaching. According to several studies on mineral and soil treatment [21-23], this contributes to an improvement in leaching quality. Intermittent leaching is appropriate for large particle size leaching, and a long pause and dry period are expected to result in more effective leaching [21]. Furthermore, as opposed to continuous liquid addition operations in industrial leaching scale, the cost of intermittent leaching operations can be significantly lower [24].

The study on hydrometallurgy must begin with laboratory tests to provide optimum conditions for selection and determination of ore treatment method and for the preliminary evaluations of the economic feasibility of the provided technological solution. After that, the process flow sheet needs to be checked, improved, and refined through testing on a larger scale.

Acid leaching of uranium from uraniferous granite sample assaying 150mg/l of G-II has been studied for the recovery of uranium by the agitation leaching technique.. Under the optimum conditions, it was possible to realize a dissolution efficiency of about 94 % for uranium [25]. Uranium was recovered by acid leaching from the waste tailing of uranium mineralization in (G-II) after re-crashing the uranium mineralized ore to –10 mm, using 20 g/l sulphuric acid solution. The leachability of uranium in the column under acid leaching conditions reached a maximum value of 83.9[26].

Extensive laboratory studies have been carried out on uranium dissolution from different uranium ore mineralization resources from the Eastern Desert of Egypt using sulfuric acid vial laboratory tests. The small column also investigated after applying optimum conditions, the obtained results show that column leaching of GII uranium ore mineralization attained about 78.3% using 34 kg/ton at 44 days leaching period [27]. In this paper, the sprinkling intensity and acid consumed during column and vat leaching are investigated. For this purpose, four column and vat leaching tests have been done at a condition shown in Table (1) for studying the reagents consumed during leaching of uranium from G-II uranium mineralization.
Materials And Methods

2.1 Chemical characteristics of GII mineralized sample

As a matter of the fact, the nature and content of major elements are generally quite important in selecting the leaching technique. Presence of acid consuming minerals makes the application of an alkaline method for uranium leaching more economic, and vice versa. Acid leaching is generally preferred because it gives higher result for uranium dissolution, unless the ore contains minerals that would reduce the acid consumption prohibitively excessive. The chemical composition of GII mineralized sample was found sufficiently simple and well situated for acid leaching since its assay is low in CaO (1.7%) and MgO (0.5%). In addition, the silicic gangue minerals (feldspars and quartz) equivalent to 73.3% SiO$_2$ are frequently predominating and which are practically inert to acid attack. The other major oxides, namely Al$_2$O$_3$, Na$_2$O and K$_2$O assaying 12.3, 3.5 and 3.4% respectively are mainly present in feldspar minerals except a relatively small Al$_2$O$_3$ content in the clay minerals. The latter are indeed of minor importance as indicated from the low loss on ignition (1.6%).

2.2 Leach experiments

2.2.1 Column leaching tests

The percolation leaching via column leaching tests was done using a representative sample from the uranium mineralization in Gattar area. The leaching experiments were performed using four PVC columns 100 mm in diameter and 1.5 m in height. A total of 15.0 kg of uranium mineralization sample was loaded into each column. The effect of the physical and chemical parameters were assessed and optimized using preliminary metallurgical experiments as column leaching using the leaching conditions shown Table 1. Using three particle size fractions of -40 mm, -20 mm, and -10 mm, the effect of particle size on the efficiency of uranium leaching was investigated [28]. For this study, an ore sample with a size of -10 mm was chosen.

Table1: Column leaching experimental conditions

| Column No. | Ore type | Ore grade, ppm | Ore weight, kg | Grain size, mm | Acid concentration g/L | Sprinkling intensity, L/S |
|------------|---------|----------------|----------------|----------------|------------------------|--------------------------|
| GII-1      | GII     | 330            | 15             | -10           | 40                     | 0.1:1                    |
| GII-2      |         | 15             | -10           | 20            | 0.1:1                  |
| GII-3      |         | 15             | -10           | 20            | 0.2:1                  |
| GII-4      |         | 15             | -10           | 20            | 0.05:1                 |

2.2.2 Vat leaching experiment
The vat (box) leaching having a general slope of about 1% from front to back to allow solution collection. Its dimension, (5m width *12m length *5 height) the total ore amount about 350 ton. To prevent seepage of pregnant liquor, the ground surface of the pad covered with polyethylene sheets, and about 10cm sand filter layer for the further filtration process. Driblet irrigation pipe is adopted to distribute lixiviant over the ore surface, to minimize the rate of evaporation. Mixing of concentrated H$_2$SO$_4$ and water or barren solution (lixiviant), and the flow rate of a lixiviant into the heap is adjusted.

2.3 Uranium determination

The uranium contents of the ore-samples, intermediate pregnant solution, and pregnant leaching solutions were determined according to the method described by Davies and Gray and modified by Nuclear Materials Authority laboratories, Egypt [29].

Results And Discussion

3.1 Results of columns leaching of GII uranium mineralization

To investigate the acid leaching of uranium mineralization in the course of heap leaching and evaluate the reagent consumption per unit mass of uranium dissolution. In this paper, the sprinkling intensity and acid consumed during column leaching are investigated. In the leaching experiments, lixiviant was added until the dissolved uranium value decreased to a sufficiently almost steady low level. For this purpose, four column percolation leaching experiments have been performed under the conditions given in Table (1), for studying the reagents consumed during dissolving of uranium from G-II uranium mineralization.

3.1.1 Acid consumption

The most important factor in the cost of chemical reagent treatment of uranium mineralization has been the amount of consumed sulfuric acid which produce a satisfactory recovery in the leaching stage. The characteristics of the feed uranium ore mineralization and the leaching conditions influence the acid concentration used in leaching. Excessive acid can speed up recovery that lead to increas acid consumption. Furthermore, the high acid concentration can cause an excessive amount of associated gangue minerals to dissolve in the pregnant solution. To avoid the precipitation of dissolved uranium, the free acidity (pH) in the leaching liquor should be maintained at a certain value.

As shown in Fig. 3, column 1 with the acid concentration (40g/l) had a higher uranium dissolution with a maximum recovery of 77.6 percent compared to 74.5 percent recovery in column 2 with the acid concentration (20g/l) for the same 42-day leaching period. Extending the leaching period in column 2 to 56 days increased total uranium recovery by 4.8 percent and increased the recovery rate to 79.3 %.

3.1.2 Interpretation of acid consumption from techno-economic view
The required reagent consumption per percentage increase in uranium recovery within the ore sample for different acid concentrations is known as the leaching index (β) \[\text{[30]}\]. As seen in the following equation: **see equation 1 in the supplementary files.**

Where, (β) is the leaching index, V total volume of the consumed liquid (L), M, total mass of the consumed acid (g), E, uranium dissolution (%).

The leaching index (β), as shown in Table 2 and Fig 3, was calculated for column 2 (20g/l) during the extension of leaching time from day 42 to day 56. According to the leaching index (β), 87.5g acid is required to achieve a 1% increase in uranium recovery in column 2 with an acid concentration of 20g/l after day 42, while the maximum amount of acid required up to day 42 requirement per unit of recovery was around 18 g. After day 42, the leaching index for column 1(40g/l) indicates that 400g acid was consumed (if extended). While, as shown in Table 3, results show that 21.2 and 32 g of acid are consumed over the duration of the dissolution process, with acid concentrations of 20, and 40 g/l, respectively, to achieve a 1% increase in uranium recovery.

Table 2: Summary of the values for m, maximum uranium extraction, E and (β) index

| Acid concentration,g | Acid consumption (g/day) | Leaching time (Days) | M=Total amount of the consumed acid (g) | E=Uranium dissolution (%) | (β)= (ΔM/ΔE), g |
|----------------------|-------------------------|----------------------|---------------------------------------|--------------------------|------------------|
| 20                   | 30                      | 56                   | Up to 42 = 1260                       | Up to 42 = 74.5%        | 1680 - 1260      |
|                      |                         |                      | Up to 56 = 1680                       | Up to 56 = 79.3%        | = 87.5g          |
|                      |                         |                      |                                       |                          | 79.3 - 74.5      |
| 40                   | 60                      | 42                   | 2520                                  | 77.6                     | 2520 - 1260      |
|                      |                         |                      |                                       |                          | = 400            |
|                      |                         |                      |                                       |                          | 77.6 - 74.5      |

Table 3: Total acid consumed during leaching period per percentage increase in uranium recovery
3.1.3 Accumulative liquid / solid ratio

During column leaching, three different sprinkling intensities were investigated 0.2:1, 0.1:1, and 0.05:1. As shown in Fig. 4, the leaching efficiency reached 78, 79.3, and 73% for 0.2:1, 0.1:1, and 0.05:1 respectively. The time period for process and solution consumption are two key techno-economic factors and play a key role in the process design and decision-making steps. From this standpoint, optimum sprinkling intensity 0.1:1 is preferable to 0.2:1.0 sprinkling intensity, the leaching rate is almost the same, but, when using 0.2:1 sprinkling intensity it will introduce solution reagent in the rate higher than in 0.1:1 and hence, will decrease the PLS residence time, concentration, and correspondingly more of the lixiviant will be percolated through the column with little or no interaction with ore particles. As in Fig.4, extending the leaching time in column 2 (0.1:1) up to 56 days increased the overall uranium recovery by about 4 % and the recovery reached 79.3%.while the lowest uranium recovery was obtained at sprinkling intensity of 0.05:1 during 62 days. The lixiviant's percolation rate should be slow enough to allow the lixiviant to have enough contact time with the ore particles to achieve a high rate of ore dissolution.

In Fig. 4 more than 50% of uranium dissolution during the first week of leaching in all sprinkling intensities, this is believed to be due to all fast-dissolution of uranium near the surface, and after that, the leach reagent has to migrate from the particle surface into the particle to recover further uranium. These results are consistent with what Ogbonna found in 2006 [31].

During the leaching experiment, a higher sprinkling rate dilutes the uranium content in the pregnant solution of the column, as shown in Fig. 5. The sharp and abrupt decrease in uranium concentration in PLS at various times and sprinkling intensities is due to the addition of fresh lixiviant on these days and the recycling of intermediate pregnant solution (IPS). The addition of fresh solution and recycling into the leaching process has an immediate positive impact on the uranium extraction rate at various sprinkling intensities in all time periods. The addition of fresh solution is more successful during the time periods of (14-21), (24-30), and (37-41) days, as shown in Fig 5.

### Table: Acid concentration, Acid consumption (g/day), Leaching time, days, M = Total amount of the consumed acid (g), E = Uranium dissolution (%), $\beta = \frac{M}{E}$, g

| Acid concentration (g/day) | Acid consumption (g/day) | Leaching time, days | M = Total amount of the consumed acid (g) | E = Uranium dissolution (%) | $\beta = \frac{M}{E}$, g |
|---------------------------|-------------------------|--------------------|-------------------------------------------|----------------------------|--------------------------|
| 20                        | 30                      | 56                 | Up to 56 = 1680                           | Up to 56 = 79.3            | 21.2                     |
| 40                        | 60                      | 42                 | Up to 42 = 2520                           | Up to 42 = 77.6            | 32.4                     |

3.1.4 Interpretation of reagent consumption from techno-economic view
As given in Table 4, the leaching index \((\beta)\) has been calculated for column 2 during the extension of leaching time from day 47, up to day 56. The results show that after day 47, 3.1 and 23 L solution was needed to achieve a 1% increase in uranium recovery in column 2 and 3 with a sprinkling strength of 0.1:1 and 0.2:1 respectively,

| Sprinkling intensity | Reagent consumption (L/day) | Leaching time, days | V= Total volume of consumed leaching solutions, (L) | E=Uranium extraction (%) | \(\beta\) = \(\frac{\Delta V}{\Delta E}\), (L) |
|----------------------|----------------------------|--------------------|-------------------------------------------------|--------------------------|----------------------------------|
| 0.1:1                | 1.5                        | 56                 | Up to 47 = 70.5                                  | Up to 45 = 75%          | 3.1 L                            |
|                      |                            |                    | Up to 56 = 84                                   | Up to 56 = 79.3         |                                  |
| 0.2:1                | 3.0                        | 47                 | 141                                              | 78                       |                                  |

Figure 6 shows the average values of the leaching index \((\beta)\) vs. leaching time for different sprinkling intensities in column 2 (0.1:1) and column 3 (0.2:1). There is no linear effect of having a high irrigation rate in the column leaching. In fact, the process of dissolving uranium from the two columns has several stages. After the surface has been depleted of quick uranium dissolution, leaching would most likely continue to dissolve uranium mineralization values for slower-leaching minerals near the surface, rather than the faster-leaching minerals closer to the center of large ore particles [32]. During the leaching process, the different sprinkling intensities would have techno-economic benefits to the process.

3.2 Vat leaching process in GII

Gattar uranium ore assaying 0.035% U was subjected to crushing to <50 mm grain size, then vat leached with diluted sulfuric acid (50~20 g/l) at optimized conditions, a leaching efficiency of about 70.7% was obtained. The obtained pregnant solution assayed 570 mg/l contains complex of mixture cations and anions, the composition of such solution is influenced by both the mineralogy of ore and the leaching parameters employed. Pregnant solution was analyzed for determination of uranium and other elements associated it.

3.2.1 Evaluation of uranium dissolution stages in vat leaching with leaching period

The obtained leach liquor might sometimes need to be recycled in the same vat or another new vat in order to obtain relatively enriched uranium concentration in pregnant liquor before being directed to the
extraction operation using the ion exchange resin in a fixed bed columnar system.

In Fig 7, uranium leaching from G-II takes place in three stages. The first stage (lag stage) during the first ten days has large quantities of reagents consumed in uranium and gangue minerals dissolution. The resulting solutions are also recycled to adjust the pH of the uranium-bearing solutions. The second stage, is the exponential stage, which obtain most of the pregnant leaching solutions (PLS). The third stage, is the stationary stage, which the concentration of pregnant leaching solutions decreases less than 80 ppm and is recycled on the vat to increase the concentration of uranium and continues until the uranium concentration decreases to less than 40ppm at this point, leaching stage was stoped because the economic return is not feasible.

### 3.2.2 Interpretation of reagent consumption during vat leaching as a techno-economic view

#### 3.3.2.1 Lixivient consumed

From the obtained data of vat leaching that extended for 90 days, it was found that a total volume of sprinkling lixivant attaining about 290 m$^3$, in the meantime, an overall leaching efficiency of about 70.7% has been obtained and the accumulative liquid to solid ratio attain about 0.83, as shown in Fig. 8.

#### 3.2.2.2 Recycling solutions

The obtained leach liquor was recycled in the same vat (350 ton) in order to obtain relatively enriched pregnant liquor. In Fig. 9, the recycling period at (1-10), (28-32), (48-52), and (63-66), the uranium in pregnant solution increased from (250-600), (360-500), (150-350), and (120-210) ppm respectively. Another reason for using the recycling process is to adjust the pH of pregnant uranium solution (in the first 10 days of leaching). The most important reason for the recycling process is to save reagent consumption, as one-third of the consumed solutions are saved during the recycling process.

#### 3.2.2.3 Lixiviant (barren) regeneration

Leach solution management is an important part of heap leach operation, both to maximize metal recovery and to prevent the loss of reagent bearing solution to the environment. The resulting acidic barren solution from ion exchange (IX) process is recycled back into the vat via the barren pond. Taking into account that, the first 10 bed volumes of the adsorption cycle are wasted to tailing in order to minimize chloride ions that obtained from elution process. The resulting PLS often includes different types of the desired metal, especially the types of decomposed impurities that were not recovered in the IX stage. The increased element concentrations increase the density, impurities and viscosity of the process solutions, and form insoluble secondary products that may be precipitated through the solution channels and/or coated resin surfaces.

The resulted effluent solution from the adsorption process in G-II was analyzed for determining the associated elements which are, Mn, Zn, Na, Mg, Fe and Si. These ions are less adsorbed on resin during uranium adsorption since their effluent /leach percent being (95.3, 76, 81.1, 76.5, 90 and 74.6%).
respectively. The barren solution was recycled to leaching for a total of 4 cycles. A laboratory test verified that uranium was dissolved in the recycled solution in spite of the sulfuric acid concentration (15g/l). In addition, the solution contains a certain concentration of ferric ion (1.9g/l) which speeds up the process of dissolving uranium dissolution.

From the leaching index (β), the result indicates that 4.1m$^3$ solutions are consumed to have a 1 % increase in the uranium recovery of vat leaching with the sprinkling intensity of 0.1:1 after 90 day, as shown in Table 5.

**Table 5: Solutions consumption during leaching period, and (β) index**

| Sprinkling intensity | Leaching time, days | V=Total volume of the consumed lixiviant, m$^3$ | E=Uranium dissolution, (%) | (β) = (V/E), m$^3$ |
|----------------------|---------------------|-----------------------------------------------|-----------------------------|-------------------|
| 0.1:1                | 90                  | 290                                           | 70.7                        | 290/70.7=4.1       |

**3.2.2.4 Acid consumption during vat leaching**

Acid consumption is the major economic factor in uranium extraction from uranium ores by vat leaching technique. As shown in Fig. 10, the increase in the uranium dissolution with a maximum recovery of 70.7% was obtained after the leaching period of 90 days. At the same time, the overall consumed acid after this period was 17.6kg/ton of ore.

From the leaching index (β) in Table 6, the result indicates that 88 kg acid is consumed to have a 1 % increase in the uranium recovery of vat leaching with the acid concentration between 50-10g/l after 90 days. Finally, a proposed flow sheet for the leaching of uranium mineralization is shown in Fig. 11.

**Table 6 Acid consumption during vat leaching period and (β) index**

| Acid concentration,g/l | Leaching time, days | M=Total amount of the consumed acid (kg) | E=Uranium extraction (%) | (β)= (M/E), (kg) |
|------------------------|---------------------|------------------------------------------|--------------------------|-----------------|
| 50-10                  | 90                  | 6220                                     | Up to 90 day=70.7        | 6220/70.7=88    |

**Conclusion**

Circulation of the leach (ILS) and barren solutions have a notable effect on the capital cost of the heap leaching system. The most important reason for the recycling process is to save reagent consumption, as one-third of the consumed solutions are saved during the recycling process. Results indicate that in
column leaching 21.2 and 32g of acid are consumed using an acid concentration of 20, and 40g/l respectively to have a 1 % increase in the uranium recovery. Also, 1.0 and 1.8 L solutions using 0.1:1 and 0.2:1 sprinkling intensities are consumed respectively to have a 1 % increase in the uranium recovery. The results of vat leaching indicate that 4.1m$^3$ solution and 88 kg acid are consumed to have a 1 % increase in the uranium recovery of vat leaching with the sprinkling intensity of 0.1:1 after 90days.

**Declarations**

**Conflict of Interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figures
Figure 1

Geologic map of Gabal Gattar uranium occurrences area, North Eastern Desert of Egypt

Figure 2

EDS analyses and ESEM images showing the mineralogy of the uranium mineralizations (a) pitchblende (b) uranophane [9].
Figure 3

Effect of acid concentration during column leaching of GII uranium mineralization

Figure 4
Cumulative uranium extraction against time for different sprinkling ratio

Figure 5

The variation in the uranium concentration against time from PLS of columns at different sprinkling ratio

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Figure 11

Hydrometallurgy process flow sheet based on vat leaching

Supplementary Files

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