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Environmental Protection by Leaching of the Remaining Radioactive Element from Granitic Wastes Obtained from the Pilot Plant of Gabal Gattar, Eastern Desert, Egypt

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Abstract

The leaching of uranium from granitic wastes of Gabal Gattar pilot plant, eastern desert, Egypt using several leaching agents such as acidic and alkaline reagents for optimization the extraction and recovery of uranium has been studied in order to protect the environment from the serious damage caused by this element especially when it leaks into groundwater in the areas surrounding the granite waste. The effect of different parameters such as different acidic leaching agent, different alkaline leaching agent, sulfuric acid concentration, grain size, solid/liquid ratio, leaching time, and leaching temperature were investigated. Uranium dissolution efficiency of 90.5% was obtained after 60 minutes contact time leaching using 4M H_2SO_4 and solid/liquid ratio 1/30 at leaching temperature 80°C and 0.8 mm grain size and 420 rpm mechanical stirring speed without any oxidant addition.

Keywords: Environmental Protection; uranium leaching; Gabal Gattar; acid leaching.

Introduction

The growth of the uranium mining and ore processing industries are exceptional. Several years ago, the uranium mining was grown from nothing to a major hydrometallurgical industry, and so other ore-processing has developed quickly. The industry has not only grown rapidly, but it also becomes the leader in developing hydrometallurgical operations such as leaching, solvent extraction, solid-liquid separation, and ion-exchange **(Seidel, 1980)**.

The release of a radionuclide into the environment as a waste by an unused radioactive substance can result in major hazardous problems. To address this issue, retain liquid waste in lead tanks and solid waste in lead chambers until their radioactivity level falls below an allowed level for disposal into the environment. This procedure is costly, time-consuming, and far from practical in terms of preventing the negative effects of radiation. Furthermore, because the tanks or chambers cover enormous areas, this technology cannot provide complete shielding for radioactive activity. Finally, the diluted radionuclide is released into the environment, where its activity is decreased to a safe level, because radionuclides transform to stable metal ions in their steady state, it causes heavy metal pollution and poisoning in addition to radioactive contamination.

Approximately 13% of global electricity is generated from nuclear energy, according to the World Nuclear Association **(Mahmoud, 2021).** A number of nations, including India, the United States, Russia, Japan and China **(IEA/OECD, 2011),** have declared nuclear energy targets. Four nations Kazakhstan, Canada,Namibia,- Australia, and the United States produce more than half (53%) of the world's uranium production with high uranium concentration **(Yan et al., 2011),** and 43% of the world's uranium production is accounted for by the top five uranium mines **(Mahmoud, 2021)**.

Different methods were applied for leaching uranium from its ore, many reagents such as alkalis and acids are usually used. Usually it is preferred to use acid leaching, because it gives higher results than alkali leaching for uranium dissolution, unless the minerals in the ore cause significant acid consumption **(Marvin et al., 1956).**

The kind of uranium mineralization and impurities minerals in the ore being studied determine whether an acidic or alkaline leaching method is used, for example, the primary ores associated with pegmatites, which contain uranium chemically related with various refractory oxides require strong acid concentrations to break down, and be incapable of alkaline leaching. Other primary ores such as pitchblende and all of the secondary ores are in fact impervious to leaching by both acid and alkaline processes **(Forward and Halpern, 1955).**

Since it is readily available and inexpensive, sulfuric acid is used as the leaching agent in all commercial uranium mills that use acid leaching [(The Palabora mining operations uranium recovery using nitric acid leaching of a uranothoriante concentrate made by gravity concentration is the sole exception) **(IAEA,1980)**].

One of the first hydrometallurgical processes is heap leaching. In Portugal in the 1950s, uranium was first reportedly used **(Cameron, 1980; Audsley and Doborn, 1963).**

Using acid, alkaline, and salt as lixiviants, **Mahdy and El-Hazek (1996)** investigated the agitation leaching characteristics of uranium from sediments in Wadi Belih Hammamat. They found that the leachability of uranium was 95%, 91%, and 88%, respectively.

The recovery of uranium from the El-Sela mineralization using sulfuric acid was tested in a column percolation leaching study, and the effectiveness of uranium leaching reached about 81.1%. Percolation leaching of uranium has been studied on the mineralization of the Eastern desert in Egypt **(Nagar et al., 2016).**

Laboratory tests using batch and column-pack methods demonstrate that uranium may be produced quickly from such refractory ore using alkaline bicarbonate solutions containing 0.1 to 5.0 weight percent NaOCl. In column-pack tests, ultimate recoveries of 90% or higher were achieved. NaOCl seems to be a potent enough oxidant to penetrate carbonaceous materials and reveal uranium mineral species that have been imprisoned **(Thomas et al., 1985).**

Several experiments have been successful in removing U and related elements from Gabal Gattar. Researchers explored acid and alkaline agitation leaching for the leaching of uranium and molybdenum from Gabal Gattar deposit. By employing 50g L^{-1} H_2SO_4 and a solid/liquid ratio of 1/2 at ambient temperature for 12 hours, acid leaching has shown to completely dissolve U/Mo. At 60°C for 8 hours, 50g L^{-1} Na_2CO_3 or $NaHCO_3$ was used to achieve an efficiency of 95.1% for uranium leaching **(Kamal, 1995).**

Both Gattar-II and Gattar-V mineralized samples were examined for uranium leaching. The latter was leached using H_2SO_4 at ambient temperature for 24 hours at 0.2 solid / liquid ratio utilizing just 30 and 40 kg ton^{-1} sulfuric acid, respectively, with 99% leaching efficiency **(Mahmoud, 2000).**

Agitation leaching of low-grade uraniferous granite from Gabal Gattar-II using sulfuric acid (150 ppm), as well as the factors impacting the leachability, such as acid concentration, grain size, agitation speed, temperature, solid/liquid ratio and period of leaching. The findings suggest that leaching effectiveness is significantly influenced by particle size. In terms of GII, the leaching efficiency of -10 mm sample is 76.9%, but -40 mm sample has a leaching efficiency of 47.4% **(Mahmoud et al., 2001).**

Material and Methods

1. Sample location

Gabal Gattar is located in the northern eastern desert, approximately 45 km southwest of Hurghada, between latitudes 27° 02' and 27° 08' N and longitudes 33° 15' and 33° 25' E (Figure 1). It represents the northern parts of the Gattar batholith, a large pink granite batholith with a high potential for usable uranium resources. This location exhibited more than 20 U-occurrences with apparent yellow secondary U-minerals. On the map, they are labelled as Gattar-I, Gattar-II, Gattar-III, Gattar-IV, Gattar-V, Gattar-VI and so on (Figure 2). The primary focus of this research is the incidence of Gattar-II of granitic wastes from pilot plant.

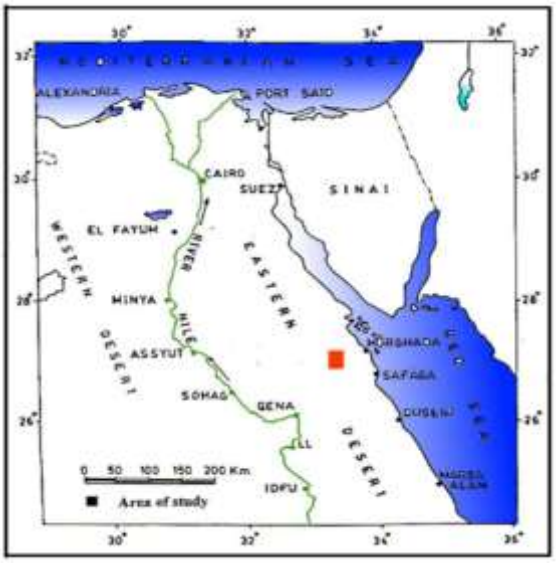

Figure 1: Location map of Gabal Gattar area **(salman et al., 2005)**.

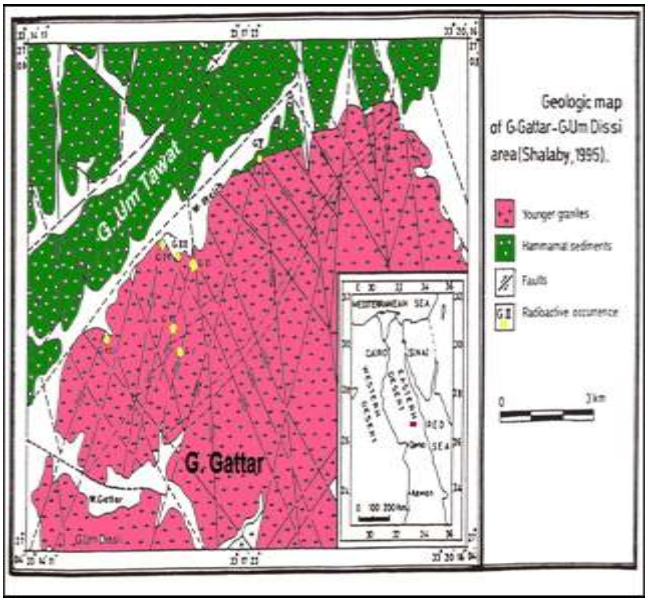

Figure 2: Geologic map of Gabal Gattar **(shalaby, 1995)**

Methods of Analysis

2.1. Chemical analysis

The collected sample from granitic wastes of Gabal Gattar pilot plant ore was ground to mesh size of 0.8 mm. Proper quartering of the sample was performed after grinding to less than 0.8 mm size to obtain a representative sample which was then thoroughly chemically analyzed to determine the amount of both major and trace content. In order to measure SiO_2, Al_2O_3, TiO_2, and P_2O_5, their corresponding spectrophotometic techniques were examined **(Shapiro and Brannock, 1962),** whilst the flame photometric approach was used to measure Na_2O and K_2O **(Jackson, 1967).** Titration techniques were used to measure total Fe as Fe_2O_3, MgO, and CaO against EDTA solutions, respectively. Gravimetric analysis was used to evaluate the loss of ignition (L.O.I.) for H_2O, humidity, and organic matter as CO_2 at 110°C, 550°C, and 1000°C for lattice water. For these important components, the estimated error is no greater than ±1%. Oximetric titration has been used to determine the control analysis of uranium in the various aqueous stream solutions following its reduction using a standard solution of ammonium metavanadate **(Mathew et al., 2009).** Following a suitable uranium reduction step using ammonium ferrous sulfate, this was made achievable. When di-phenyl sulphonate is utilized as an indicator in this process, its color changes to a violet red, Next, the uranium recovery percentage was computed using the formula (R=Cc/Ff*100) in which R is the uranium recovery percentage, C is the uranium weight in the pregnant solution, c is the uranium prenatal solution assay, F is the uranium weight in the ore sample, and f is the uranium assay in the ore sample. The uranium content of each size fraction was then ascertained chemically using the oxidimetric titration method.

2.2.2. Reagent

Every reagent was created using chemicals of analytical quality. H_2SO_4, HCl, NaOH, $CaCl_2$, K_2SO_4, HNO_3, Urea, Ammonium sulfate, Ammonium nitrate Arsenazo III, Phosphoric acid, Bromine water, ferrous sulphate.anhy, ammonium vanadate and Diphenylamine sulphonic acid sod. Salt supplied by Riedel-deHaen, USA. Ascorbic acid and Super phosphate from El-Nasr pharmaceutical chemicals, Egypt, All reactions were performed utilizing flame-dried glassware.

2.2.3. Apparatus

Using a digital pH meter modal (Schott, Germany), the acidity and alkalinity of solutions were determined. An analytical balance (Shimadzu model AY220, Japan) was used to weigh all samples, chemicals, and reagents. Using the arsenazo (III) indicator and a 650 nm wavelength, a double beam spectrophotometer (UNICAM, England) was used to perform a quantitative measurement of U(VI) in comparison to a suitable standard solution **(Abd El-Rahem et al., 2021).** A hot plat and magnetic stirrer (Model US152, UK) is used to stir and heat samples.

2.2.4. Agitation leaching

Using a magnetic stirrer, each leaching experiment in the leaching method involved mixing a weighed quantity of the sample of ground (1-5 gm) with a given volume of acid at a particular concentration at a particular solid/liquid ratio for a predetermined amount of time at the necessary temperature. The metal levels of the resulting leach liquors were then examined in order to quantify their dissolving efficiency. The primary pertinent factors for acid leaching that affect uranium recovery are solid/liquid ratio, temperature, acid concentration, leaching time and grain size.

Results and Discussion

3.1. Material properties.

Chemical examination of the main components of the working sample was examined and the results were tabulated in table 1.

Table 1: Chemical analysis of the major elements for the working raw sample

Constituent	Fe_2O_3	CaO	MgO	TiO_2	SiO_2	K_2O	Na_2O	Al_2O_3	P_2O_5	L.O.I	Uppm
Oxide, %	4.32	2.80	5.77	0.11	68.7	3.06	4.21	7.26	1.32	1.91	210

3.2. Factors Affecting Uranium Leachability

3.2.1. Effect of different acidic agent

The collected sample was subjected for dissolution using different acids, several experiments were studied for uranium leaching with fixing acid concentration at 2 M, at room temperature, 1 hour. leaching time and 1/50 solid/liquid ratio. The results obtained were plotted in (figure 3), from the results, it was found that H_2SO_4 was chosen as the best acidic reagent for uranium leaching from the working selected sample, which gave the highest leaching efficiency.

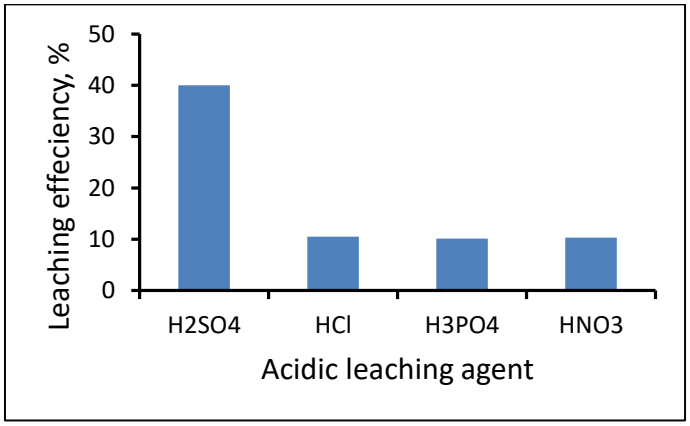

Figure 3: Effect of acidic reagents upon uranium leaching efficiency.

3.2.2. Effect of various alkaline leaching agent

The effect of various alkaline leaching reagent whether single or in combination on uranium leaching efficiency has been studied. Several experiments were examined with fixing other leaching factors at room temperature, 60 minutes. leaching time and the concentration and 1/50 solid/liquid (S/L) ratio of leaching agents was (0.1M). The obtained leaching efficiencies of uranium were represented in (figure 4). These results showed that NH_4NO_3+H_2SO_4 and $(NH_4)_2SO_4$+H_2SO_4 were chosen as the best alkaline leaching reagents for uranium leaching. The previous results showed H_2SO_4 was chosen as the best choice for uranium (VI) leaching from the working sample.

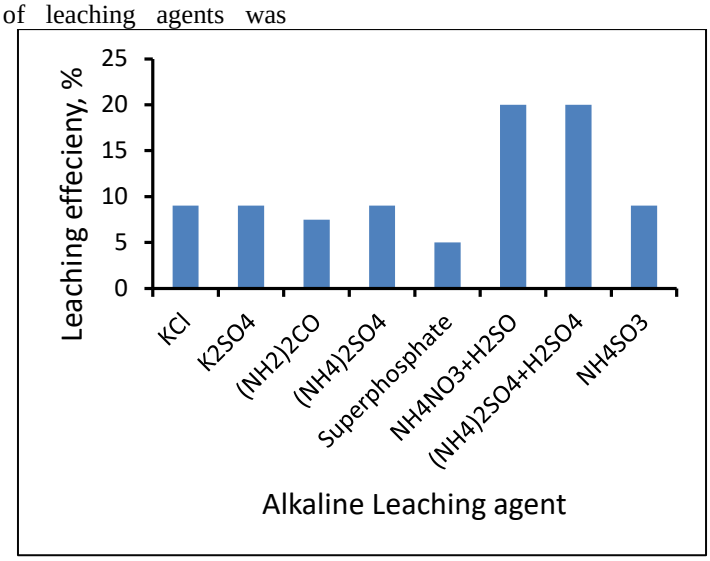

Figure 4: Effect of alkaline reagents upon uranium leaching efficiency

3.2.3. Effect of different H_2SO_4 concentration.

The concentration of H_2SO_4 is very important factor for leaching of uranium (VI) from the working sample, so several experiments were investigated for different H_2SO_4 concentration ranging from 0.5 to 8 M with fixing other factors at room temperature, 60 min. mixing time and 1/50 S/L ratio. The results obtained were plotted in (figure 5), these results indicated that leaching efficiency of uranium was increased gradually with increasing H_2SO_4 concentration from 0.5 to 4 M, above 4 M H_2SO_4 the leaching efficiency of uranium was decreased, hence 4 M H_2SO_4 was chosen as the best one for uranium leaching from the concerned sample.

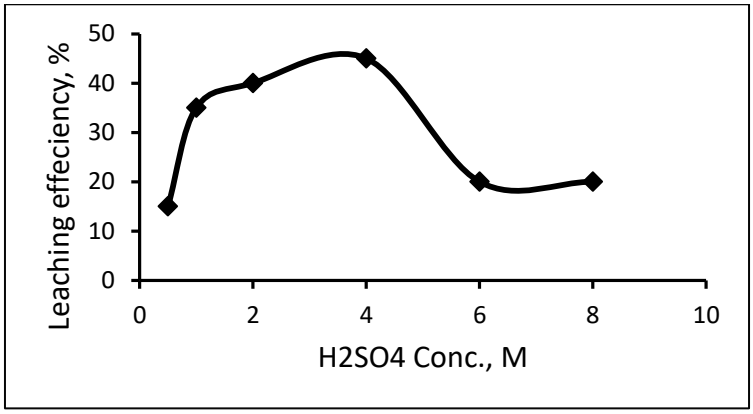

Figure 5: Effect of different H_2SO_4 concentration upon uranium leaching efficiency.

3.2.4. Effect of mixing time.

The effect of leaching time upon uranium (VI) leaching efficiency from the collected sample was studied in the range from 30 minutes to 4 hrs, while the other leaching parameters were fixed at 4 M H_2SO_4, room temperature and 1/50 solid/liquid ratio. The obtained data were shown in (figure 6). From the obtained data, it was clear that leaching percent of uranium from working sample was increased by increasing time from 30 minutes to 60 minutes and over this time the leaching efficiency was slightly decreased, so that, 60 minutes agitation time was chosen as the best choice.

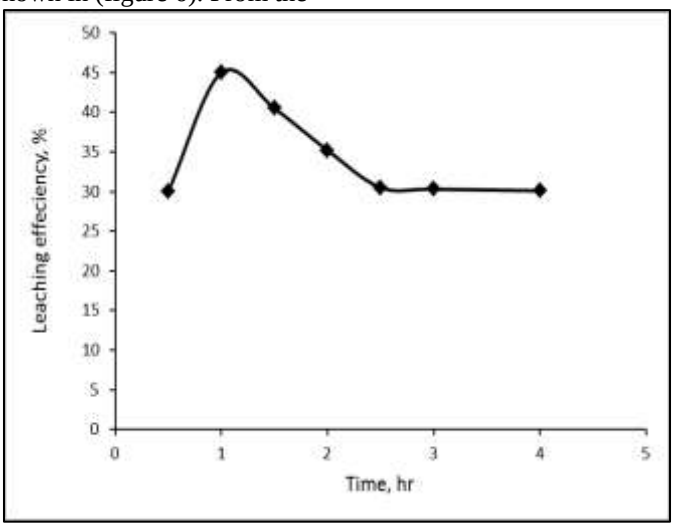

Figure 6: Effect of agitation time upon uranium leaching efficiency.

3.2.5. Effect of grain size.

Several experiments were performed to study the effect of different grain size of the collected sample which gives the highest uranium leaching efficiency. Different grain size were examined with fixing other conditions at room temperature, 4 M H_2SO_4 leaching reagent, 60 minutes agitation time and at 1/50 solid/liquid ratio. The results founded were plotted in (figure 7) and the results revealed that 0.8 mm was chosen in this work to give the best leaching efficiency of uranium.

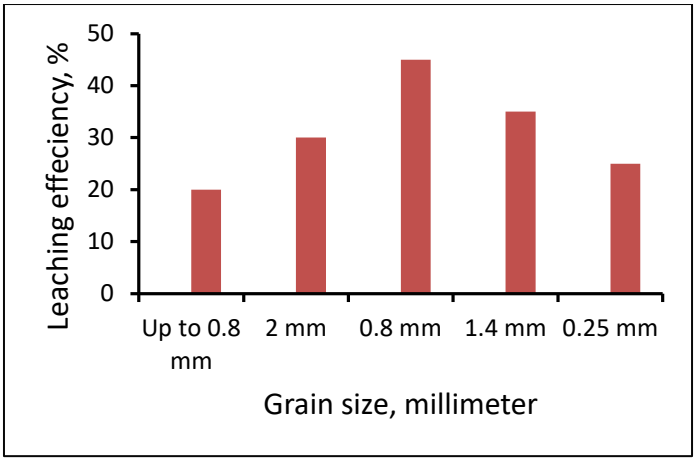

Figure 7: Effect of grain size upon uranium leaching efficiency.

3.2.6. Effect of solid/liquid ratio.

Several experiments were conducted to investigate the effect of different solid/liquid ratio of the collected sample which gives the highest uranium leaching efficiency. Different solid/liquid ratios were examined with fixing other conditions at room temperature, 4 M H_2SO_4 leaching reagent, 60 minutes agitation time and at 0.8 mm grain size. The results founded were plotted in (figure 8) and the results revealed that 1/30 solid/liquid ratio was chosen in this work to give the highest leaching efficiency of uranium.

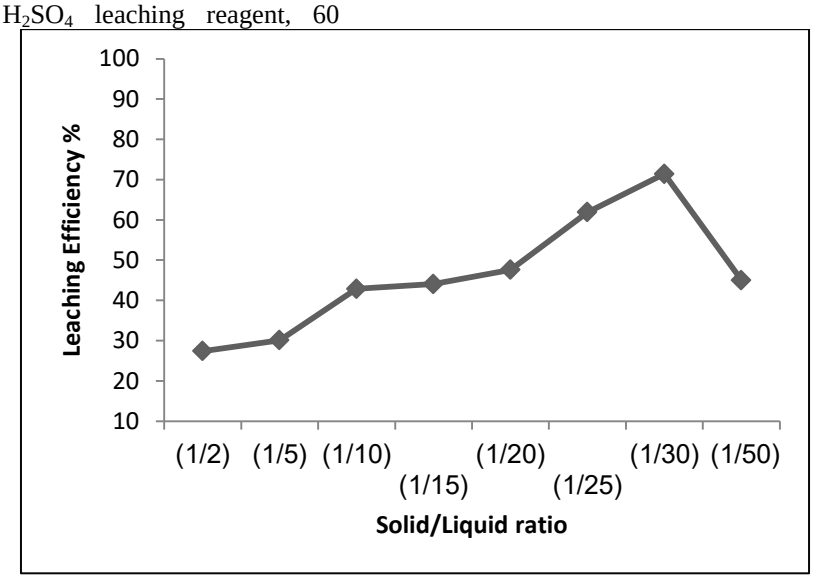

Figure 8: Effect of different solid/liquid ratio upon uranium leaching efficiency.

3.2.7. Effect of different temperature.

The effect of temperature upon uranium (VI) leaching efficiency from the collected sample was studied in the range from ambient temperature to 100 ^oC. while the other leaching parameters were fixed at 4 M H_2SO_4 leaching reagent, 60 minutes agitation time and at 0.8 mm grain size and 1/30 solid/liquid ratio. The obtained data were shown in (figure 9). From the obtained data, it was clear that leaching percent of uranium from working sample was increased by increasing temperature from room temperature to 80 ^oC, and then it was fixed.

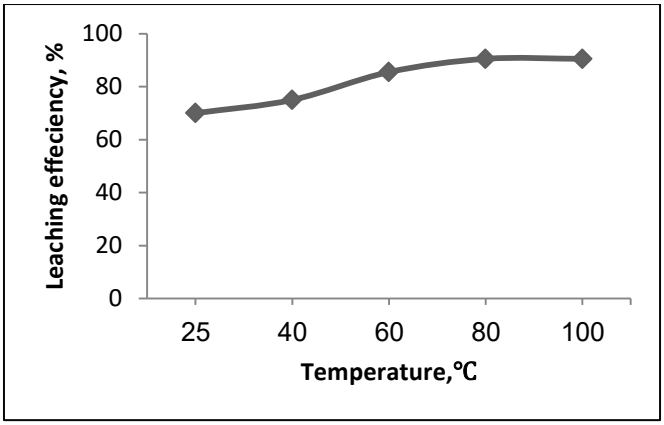

Figure 9: Effect of temperature upon uranium leaching efficiency

Conclusion

From the above previous leaching parameters, it was concluded that uranium can be leached from the collected sample from Gattar wastes, Eastern Desert, Egypt with high leaching efficiency using acidic leaching reagent at 4 M H_2SO_4, 80 ℃ , 1/30 solid/liquid for 60 minutes time period and 0.8 mm grain size. We recommend not using both urea and ammonium sulphate fertilizers in lands containing a percentage of uranium, as they are efficient in dissolving it. Superphosphate fertilizer is also less efficient in dissolving uranium.

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حماية البيئة من خلال إذابة العناصر المشعة المتبقية من النفايات الجرانيتية المستخرجة من محطة جبل جتار التجريبية بالصحراء الشرقية بمصر

نادر زكريا محمد , حسن سعيد الجندي , أحمد سعيد أبوزيد , عثمان عبد النبي دسوقي , محمد علي عبد السلام

يهدف هذا العمل إلي أذابة النفايات الصلبة الناتجة من الوحدة التجريبية بجبل جتار وإستخلاص اليورانيوم المتواجد بها لتقليل الأضرار البيئية وحماية البيئة من الأثار السلبية والخطيرة لعنصر اليورانيوم المشع والعمل علي تقليل نسبتة. تمت دراسة أذابة اليورانيوم من النفايات الجرانيتية لمصنع جبل جتار التجريبي بالصحراء الشرقية بمصر باستخدام العديد من عوامل الأذابة مثل الكواشف الحمضية والقلوية لتحسين أذابة اليورانيوم . تم التحقيق في تأثير المعاملات المختلفة مثل عامل الاستخلاص الحمضي المختلف وعامل الأذابة القلوي المختلف وتركيز حامض الكبريتيك ووقت الأذابة وحجم الحبيبات ونسبة الصلب إلى السائل ودرجة حرارة الأذابة. تم الحصول على كفاءة إذابة اليورانيوم بنسبة 90.5٪ بعد 60 دقيقة من وقت الأذابة باستخدام حامض كبريتيك (4 مولر) ونسبة الصلب إلى السائل 30/1 عند درجة حرارة الأذابة 80 درجة مئوية وحجم الحبيبات 0.8 مم وسرعة التقليب الميكانيكية 420 دورة في الدقيقة دون إضافة أي مؤكسد.