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Extraction of uranium from carbonate leach liquor by using dynamic ion exchange technique

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ABSTRACT

In this study uranium extraction was carried out by agitation in a separate batch operation. After equilibrium, the solution and resin mixture was filtered and the remaining uranium was analyzed in the solution. A carbonate leach liquor with 300 ppm uranium was used against Amberlite IRA 402 (Cl^-). The extraction efficiency reached to 90% in one stage (pH of the leach liquor equal 8, 20 min agitation time, resin to solution ratio is 1/20 at room temperature). Elution step was achieved by 1M sodium chloride with resin to liquid ratio of 1/10 and 20 min agitation time at room temperature. These conditions led to nearly complete elution of the adsorbed uranium on the used resin.

KEYWORDS

Anionic resin;
Carbonate leach;
Uranium.

INTRODUCTION

The extractive metallurgy of uranium is well established industry and some of the processes in use utilize ion exchange resin[1]. Ion exchange operation in its simplest terms involving the redistribution of the counter ions with those present in the liquid phase[2]. Historically, operation were performed statistically through fixed beds of ion exchange resin placed in columns. Some technical advantages were expected through dynamic ion exchange. Acceleration of ion exchange reaction by agitation is a logic imagination. Dynamic ion exchange in a separate batch was applied[3]. In carbonate medium, uranium forms a definite uranyl carbonate complex:

$$UO_2^{2+} + 3CO_3^{2-} \longleftrightarrow UO_2(CO_3)_3^{-4} \quad (4)$$

The ion exchange reaction means selectively adsorption of uranyl carbonate anion as follow:

$$4\,R^+X^- + UO_2(CO_3)_3^{-4} \longleftrightarrow (R)_4\,UO_2(CO_3)_3 + 4X^-$$

Where R^+ represents the fixed ion exchange sites of the resin, $X^- = NO_3^-$ or Cl^-. The above reactions are reversible, and the uranium may be desorbed or eluted from the resin by shifting the reactions to the left[5,6]. This is usually accomplished with the solutions containing either nitrate or chloride salts[7].

In Nuclear Material Authority, uranium mineralized granite sample was finely grounded and subjected to leaching using mixture of sodium carbonate and sodium bicarbonate solution. Uranium concentration in the produced leach liquor was 300 ppm uranium. Ion exchange experiments were hold in a glass beaker with suitable volume and agitation was performed by electrical motor with a suitable power supplied with turbine mixture adjusted at 300 rpm. To determine the uranium concentration, known aliquot was taken from the leach liquor after extraction. The uranium extraction percentages were performed spectrophotometrically using Arsenazo III method as follow[8]:

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$$\% \text{ U Uptake} = (C_{\text{initial}} - C_{\text{remain}}) \text{ X } 100 / C_{\text{initial}}$$

Where $C_{initial}$ and C_{remain} are the initial and remain concentration of uranium in aqueous solution respectively.

In the elution expermints, uranium recovery percentage was calculated as follow:

$$\% \text{ U Recovery} = (C_{load} - C_{remain}) \text{ X}100 / C_{load}$$

Where C_{load} and C_{remain} are the concentration of uranium loaded and remain onto the resin respectively[9,10].

EXPERMITAL

Uranium extraction experiments were carried out by using 1gram of the resin against suitable volume of leach liquor. The relevant factors of uranium extraction were studied.

Effect of pH

To test the effect of pH of the leach liquor on the extraction of uranium, 1gram of resin was taken against 20 ml of carbonate leach liquor at room temperature and pH intervals from 2 to 12 for 20 minute agitation.

Effect of agitation time

By fixing the pH of the liquor at a suitable value, the uranium extraction was carried out to test the effect of agitation time in time interval 2 to 100 minute with resin to liquid ratio of 1/120 at room temperature.

Effect of resin to solution ratio

This factor was investigated under pre-determined pH and agitation time, then interval ratios varied from 1/5 to 1/80.

Effect of Temperature

By fixing the pre-tested factors, the temperature effect was investigated in temperature intervals varied from room temperature to 80°C.

RESULTS AND DISCUSSION

Analysis takes place for the carbonate leach solution after precipitation using sodium hydroxide. The result indicate that the main elements present in the solution are 63.34% uranium (300 ppm) and some elements present in small quantities like S 0.1%, Al 4.37%, K 6.21%, Ca 2.6%, and Si 16.54%.

The influence of certain factors affecting the extraction included pH, shaking time, temperature, resin to solution ratio (R/S), and effect of adding carbonate ions.

Effect of pH

An uptake measurement under controlled pH was done using 1M NaOH & 1M HNO_3 by taken solutions of different pH intervals from 1.3 to 9.8 by placing 1 g of resin in a round flask containing 20 ml of the metal ion solution under investigation then shaking for 20 min. at room temperature 28±1 °C on a Vibromatic-384 shaker at 300 rpm. Then solution was taken for analysis. From the data obtained in Figure (1) it was found that the maximum extraction take place at pH between 8.8 - 9.8.

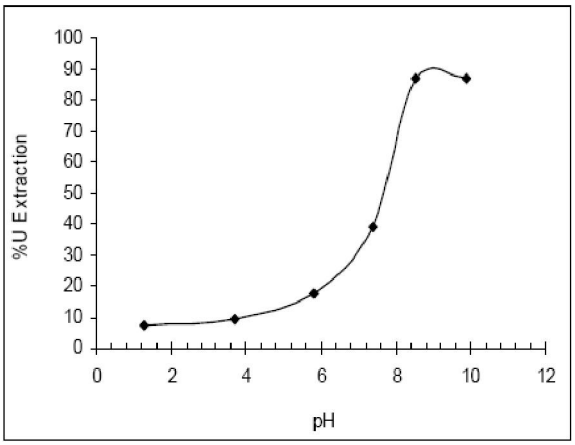

Figure 1 : Effect of pH on the extraction of uranium

Effect of shaking time

Measurements to define the equilibrium time was done at pH = 9.8 by shaking 20 ml of metal ion solution from 2 min. to 120 min., at room temp. 28±1 °C. The results given in Figure (2) show that the extraction increase as the shaking time increase then the extraction process become nearly stable after 20 min, which mean that the equilibrium time take place after 20min

Effect of temperature

The effect of temperature on the adsorption of metal ions was carried out at different temperature 28, 40, 50, 60 and 70 °C by placing 1g of resin in a series of flasks containing 20 ml of metal ion solutions at desired pH and shaking time. From the data in Figure (3) it was found that extraction decrease with increase in temp. therefore, the optimum temperature is 28 °C.

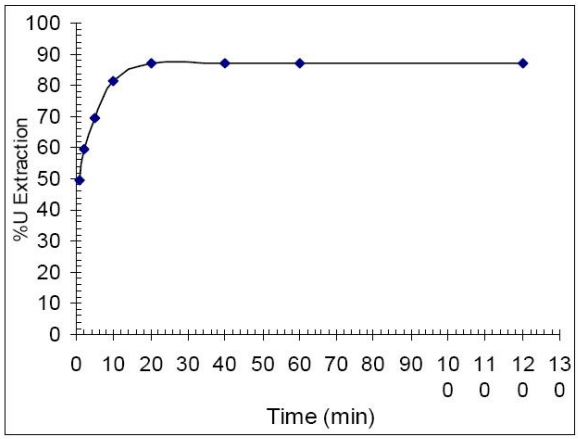

Figure 2 : Effect of shaking time on the extraction of uranium

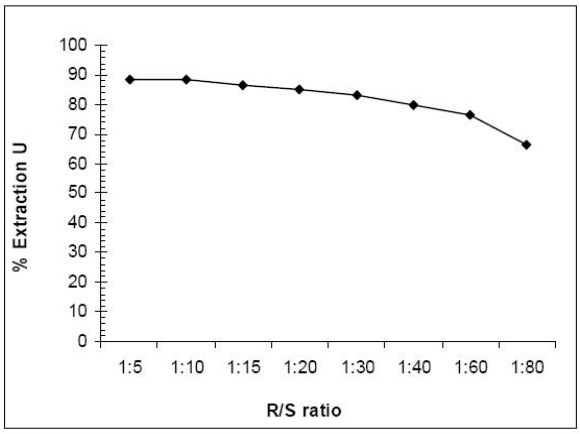

Figure 4 : Effect of resin to solution (R/S) on uranium extraction

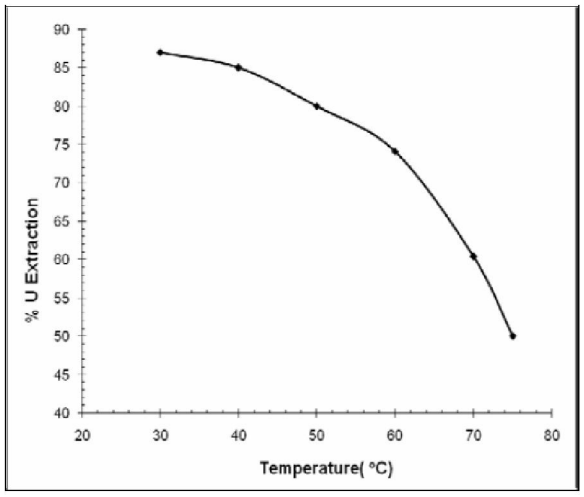

Figure 3 : Effect of temperature on uranium extraction

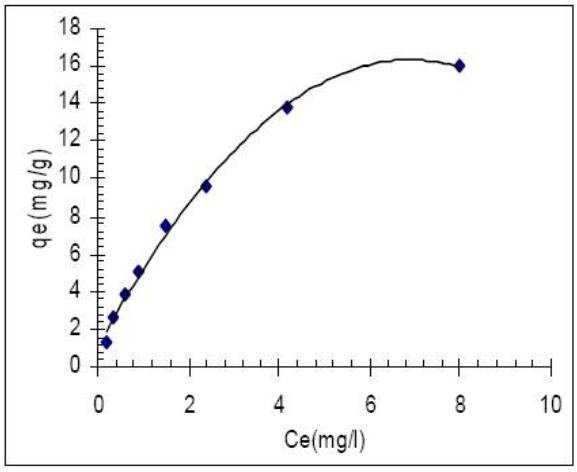

Figure 5 : General adsorption isotherm

Effect of R/S ratio

The effect of resin to solution ratio on the extraction of metal ions carried out at different ratios 1:5, 1:10, 1:20, 1:30, 1:40, 1:60, 1:80 by placing 1g of resin in a series of flasks containing different volumes (ml) of metal ion solutions at fixed pH, shaking time and temperature. The data is plotted in Figure (4) which show that as the volume of the leach solution increase the extraction decreases because the concentration of metal ion increased, with fixed resin sites. The maximum extraction was 88.3% at 1:5 ratio, but we choose the ratio 1:30 which give 83.3% to be suitable for pilot scale.

Extraction isotherms

The results obtained for the extraction isotherms of uranium from carbonate media reflect the efficiency of anionic resin (Amb. IRA402) for the extraction of uranium. The general adsorption isotherms calculated and represented in Figure (5). The distribution of each metal ion between the solid liquid interface at equilibrium has been applied to different isotherms.

Langmuir equation can be written as[11]:

$$C_e / q_e = 1/b\ Q_o + Ce / Q_o$$

Where b is Langmuir constant, Q^o ($mg_{metal}/g_{sorbent}$) is the maximum amount of metal taken up. From the slope and the intercept of the linear relation, Q^o for U(VI) was found to be 8.12 ($mg_{metal}/g_{sorbent}$), and b for U(VI) is 2.55

Elution process

To recover uranium from the loaded resin using different eluents, certain affected factors were studied. These factors are the concentration of eluent solution, shaking time, temperature, and resin to solution ratio (R/S ratio). Uranium was loaded from the carbonate leach liquor on the resin to make the resin contain 270 mg/g resin.

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Effect of using different eluents

Recovery of uranium from the loaded resin (Amb. 402) takes place using different reagents. Measurement was done using different concentrations of NaCl, Na_2SO_4, Na_2CO_3, and KNO_3. By placing 1 g of the loaded resin in a round flask containing 20 ml of the prepared solutions then shaking for 20 min. at room temperature 28± 1 °C. From the data obtained from Figure (6) it was found that the maximum recovery take place by 1.25M NaCl & 1M KNO_3 with values of 60.3% and 67.6%, respectively. Although the recovery in case of nitrate is much more effective than chloride but we preferred sodium chloride from the economic point of view.

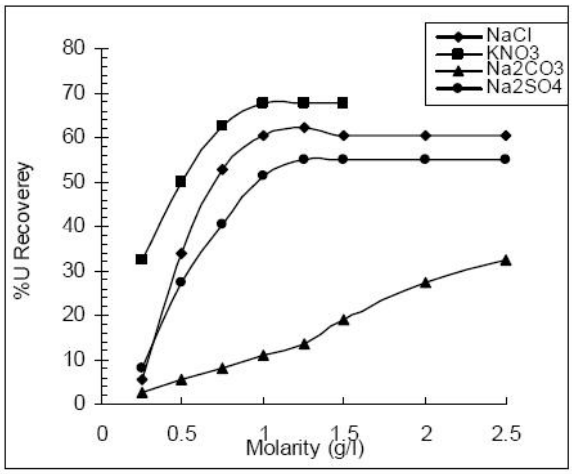

Figure 6 : Effect of different conc. of different eluents on uranium recovery from the loaded resin, shaking time = 20 min., R/S ratio = 1:20, temperature = 28±1°C.

Effect of shaking time

Effect of shaking time on uranium recovery from the loaded resin has been investigated. The shaking time was varied from 1 to 120 minutes. The obtained results represented in Figure (7) indicate that the recovery of uranium increased as the time increase then the extraction process become nearly stable after 5 min, which mean that the equilibrium take place after 5min.

Effect of temperature

Recovery measurements take place by taking 20 ml of NaCl solution with 1g of the loaded resin at optimum conc. of NaCl, and time and temperature varies from 30 to 80°C. Obtained results represented in Figure (8) indicate that the recovery of uranium doesn't change with the change in temperature.

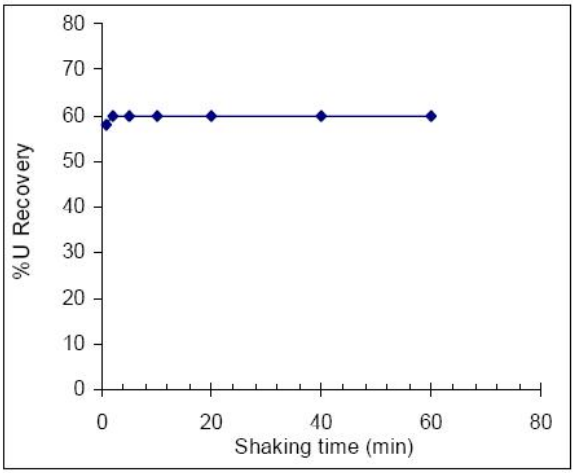

Figure 7 : Effect of shaking time on uranium recovery from the loaded resin, NaCl concentration 1.25M, R/S ratio = 1:20, temperature = 28±1°C.

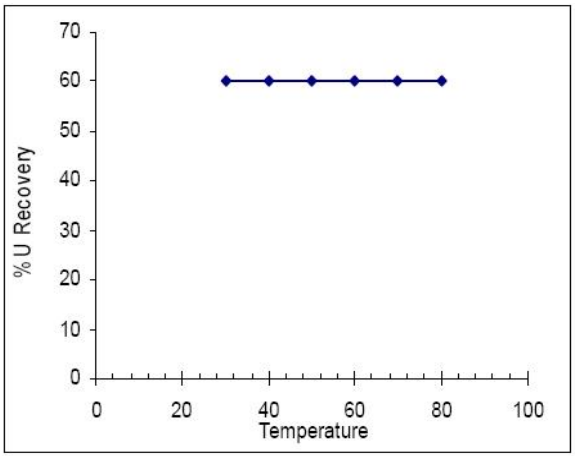

Figure 8 : Effect of temperature on uranium recovery from the loaded resin, shaking time = 5 min., NaCl concentration 1.25M, R/S ratio = 1:20.

Effect of resin to solution (R/S) ratio

Recovering of uranium at different R/S ratios from 1:5 to 1:160 has been investigated. The obtained results are represented graphically in Figure (9). It was found that the recovery of uranium from the loaded resin decrease gradually as the volume of the solution increase which indicate that the resin sites has high affinity toward the chloride ions rather than uranium complexes. The maximum recovery was 99.3% at 1:5 ratios which give the maximum recovery with the minimum volume (volume reduction).

Analysis of recovered uranium

The recovered uranium by sod. chloride was precipitated using sodium hydroxide. EDAX analysis of the precipitated uranium indicated that it is present

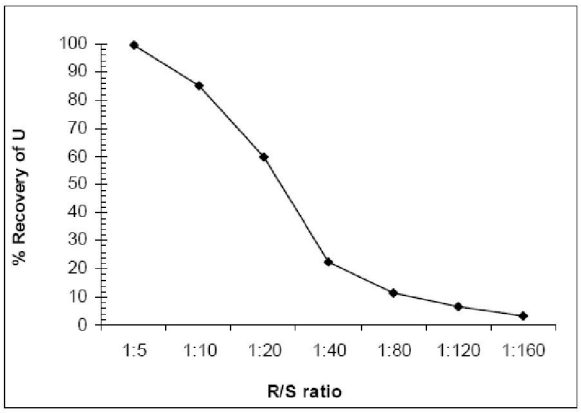

Figure 9 : Effect of resin to solution ratio (R/S) on uranium recovery from the loaded resin, shaking time = 5 min., NaCl concentration 1.25M, temperature 28±1 °C.

as major constituent of 90 %, and the residue contains minor amounts of Al, Na, K, Si, and P Figure (10).

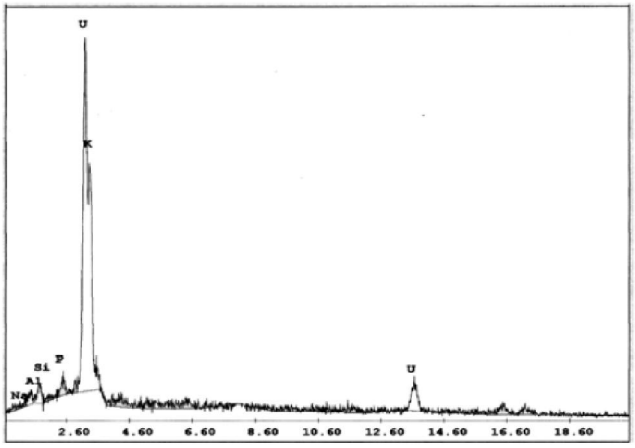

Figure 10 : EDX analysis of the uranium solid product obtained from ion-exchange extraction approach using NaCl as recovering agent.

CONCLUSION

From this study we can conclude the following advantage of agitation of resin in separate batches:

- Agitation accelerates ion exchange reaction.
- Highly shortening in operation time comparing with classical fixed beds technique.
- Maximum utilization of resin capacity.
- Excluding classical parameters such as porosity and retention time.
- Agitation ion exchange is very closer to solvent extraction technique but without possibility of third phase formation.

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