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A novel procedure for processing of the xenotime mineral concentrate of southwestern Sinai

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Abstract−A novel procedure for processing the xenotime mineral concentrate of southwestern Sinai (55.4% RE_2O_3 and 0.9% U_3O_8) has been studied. This procedure is based on leaching of uranium and rare earths from the xenotime concentrate using ferric sulfate solution by hydrolytic leaching with ferric ion, wherein ferric iron is precipitated to generate acid for the leaching step. The obtained slurry is washed by water and sulfuric acid. The relevant conditions of the ferric ion leaching of the concentrate and the washing step have been optimized. From the obtained sulfate liquor, rare earths are directly precipitated as oxalates. Uranium is extracted from the REE-free sulfate solution using ion exchange resin Amberlite IRA-400. Highly pure oxide products of the two interesting metal values, REEs and U, have properly been prepared.

Keywords: Xenotime, Ferric Sulfate, Sulfuric Acid, Uranium, Rare Earths, Ion Exchange Resin

INTRODUCTION

Rare earth elements have unique physicochemical properties that make them essential elements in many high-tech components such as hybrid cars, wind turbines, compact florescent lights, flat screen televisions, mobile phones, disc drives and defense technologies [1]. Xenotime is one of the three most commonly and commercially important mineral sources of rare earth elements (REEs) together with monazite and bastnasite [2]. Xenotime is an yttrium phosphate containing about 67% rare earth oxides, mostly of the heavier elements in addition to xenotime may or may not contain uranium or thorium [3,4].

Rare earth minerals are usually beneficiated by flotation or gravity or magnetic processes to produce concentrates [1]. A xenotime mineral concentrate is prepared by flotation from Sarabit El Khadem ferruginous sandstone of southwestern Sinai (55.4% RE_2O_3 and 0.9% U_3O_8) [5]. According to Gupta and Krishnamurthy (1992), xenotime mineral concentrate is generally treated using three breakdown processes: a digestion sulfuric acid route at 300 °C, fusion by sodium hydroxide route at 400 °C, and roasting by sodium carbonate route at 900 °C [6-10].

Most uranium is extracted from its ores by leaching in sulfuric acid [11]. The latter is made more effective by strong oxidizing agents, particularly the refractory ores. The most commonly used oxidants, sodium chlorate and pyrolusite, act indirectly by converting ferrous sulfate, formed by the dissolution of iron minerals in the ore or iron introduced during grinding, into ferric sulfate [12]. The refractory ores involve primary uranium minerals, typically uraninite and brannerite and acid consuming minerals, typically phosphate and carbonate minerals [13-19]. The added acid must be sufficient to prevent formation of jarosite, $MFe_3(OH)_6(SO_4)_2$, goethite, FeO(OH) and other refractory iron minerals which precipitate from iron solution due to oxidation as follows and represented by Eqs. (1)-(4):

$$Fe_2O_3+3\ H_2SO_4 \rightarrow Fe_2(SO_4)_3+3\ H_2O \tag{1}$$

$$Fe_2(SO_4)_3+4\ H_2O \rightarrow 2\ \mathbf{FeO(OH)}+3\ H_2SO_4 \tag{2}$$

Lepidocrocite or geothite

$$Fe_2(SO_4)_3+(2+2x)\ H_2O \rightarrow 2\ Fe(OH)SO_4.x\ H_2O+H_2SO_4 \tag{3}$$

$$Fe_2(SO_4)_3+M_2SO_4+12\ H_2O \rightarrow 2\ \mathbf{MFe_3(OH)_6(SO_4)_2}+6\ H_2SO_4 \tag{4}$$

Jarosite

where M represents any of the ions Na^+, K^+, Al, H_3O^+

In the meantime, and in much the same manner as U and REE adsorption by already existing iron minerals, iron when present in a soluble form (ferric oxide hydrates) can adsorb uranyl and lanthanide ions and thus removes most of the uranium and lanthanides from solution down to a concentration of 100-200 ppm [20-24]. According to Sperr et al., 1981, Abaa, 1991 and Dutrizac et al., 2006, adsorption on iron oxy-hydroxides is effective in pH ranging between 0.2 and 1.6 but decreases at pH's above 1.6 [21,25-27].

The present work is concerned with a novel procedure for processing of the xenotime mineral concentrate that has been physically upgraded through froth flotation from its ferruginous sandstone in Sarabit El Khadem area of southwestern Sinai. The latter was found to assay up to 82.8% RE phosphates and 0.9% U_3O_8. The agitation leaching with ferric sulfate procedure has thus been found adequately convenient for the working xenotime concentrate to economically recover its constituents by hydrolytic leaching with ferric ion, wherein ferric iron is precipitated to generate acid for the leaching step. The relevant factors of the ferric ion leaching step have first been studied and after studying the washing step of the ob-

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Table 1. Chemical composition of the working xenotime concentrate

Component	Weight percent, %
RE_2O_3	55.4
P_2O_5	27.7
Fe_2O_3	7.4
SiO_2	6.7
U_3O_8	0.9
Al_2O_3	1.8
Total	99.9

tained slurry by water and different concentrations of H_2SO_4, both the REE and U values have been presented in the sulfate liquor. From the latter, the REEs have been directly precipitated as their oxalates, while recovery of the U value left behind has properly been studied via ion exchange resin technique using Amberlite IRA-400.

EXPERIMENTAL

1. Material Characterization

The xenotime concentrate used as raw material for the experimental purpose was obtained from the ferruginous sandstone of Sarabit El Khadem area, southwestern Sinai through froth flotation as mentioned above. The chemical composition of xenotime, given in Table 1, was found to contain 27.7% phosphate as P_2O_5 and 55.4% RE_2O_3 (xenotime constituents). On the other hand, the associated U content was found to assay 0.9% as U_3O_8 while Fe_2O_3, SiO_2 and Al_2O_3 were found to assay 7.4, 6.7 and 1.8% respectively [9].

2. Experimental Procedure

2-1. Breakdown (Leaching) of Xenotime Concentrate

2-1-1. Ferric Sulfate Leaching of the Xenotime Concentrate

Several leaching experiments were performed upon the working xenotime concentrate using $Fe_2(SO_4)_3$. In these experiments, different concentrations of ferric ion were agitated with fixed weight sample portions (2 gm) of the xenotime concentrate at different solid/liquid ratios for different time periods and at different temperatures. A hot plate with magnetic stirrer was used in these experiments. The obtained slurry was cooled, filtered, washed with distilled water and made up to volume for REEs and U analysis and from which the xenotime dissolution efficiency was calculated. The leaching efficiency percent of REEs and U is calculated according the equation:

$$\%\ \text{Leaching efficiency} = \frac{\text{Metal content in the leach liquor}}{\text{Original metal content in the ore}} \times 100$$

Relevant factors of each of these experiments were studied and included mainly temperature, reagent amount and conc. as well as the leaching time.

2-1-2. The Washing Step of the Slurry

Different concentrations of sulfuric acid were used for washing of the obtained slurry produced via the optimum leaching conditions of the xenotime concentrate with ferric sulfate in stead of dist. water.

2-2. Recovery of REEs and U

To study the recovery of both REEs and U, a 25 gm sample portion of the working xenotime concentrate was subjected to ferric ion leaching under the optimum conditions of ferric sulfate followed by the filtration and proper washing of the obtained slurry by sulfuric acid.

2-2-1. Recovery of REEs

REEs were relatively recovered from the obtained sulfate leach liquor by their direct precipitation as oxalates using oxalic acid while U was left behind in the solution.

2-2-2. Recovery of U

To obtain an adequately pure uranium concentrate, it was necessary to study the U extraction from the REE-free sulfate leach liquor by anion exchange resin technique using Amberlite IRA-400 followed by its eluation with acidified NaCl solution. The obtained U eluate was then subjected to its precipitation as sodium di-uranate ($Na_2U_2O_7 \cdot 2H_2O$) via pH adjustment using 10% NaOH solution until pH 7.5.

3. Control Analysis

For the control analysis of all the performed experiments in laboratories of Faculty of Science, Zagazig University and Egyptian Nuclear Materials Authority, REEs were analyzed in all the working stream solutions using a spectrophotometer to measure their concentration with Arsenazo-III as the chromogenic reagent [28], while uranium was analyzed volumetrically by the oxidimetric titration procedure using ammonium metavanadate in the presence of diphenyl sulfonate indicator until its color end point changes to slightly violet red color [29]. On the other hand, the two highly pure products of both the REEs and U were analyzed in the labs of the Chemical Warfare by SEM qualitative analysis using EWAR Model FEI Inspect S, (Hollanda-USA).

RESULTS AND DISCUSSION

1. Processing of Xenotime Concentrate

As previously mentioned, the agitation leaching using ferric sulfate has been used for the breakdown of the xenotime mineral and the relevant factors of this procedure have been optimized; namely the ferric ion concentration, solid/liquid ratio and agitation time as well as the leaching temperature. After filtration and washing of the obtained slurry with different concentrations of sulfuric acid (in the range of 10 to 70 g/*l*) and water, the dissolved rare earths and uranium were taken as a measure of the mineral breakdown followed by precipitation of the first using oxalic acid and the latter using ion exchange resin (Amberlite IRA-400).

1-1. Breakdown (Leaching) of Xenotime Concentrate

To optimize extraction of elements, the residue from the ferric leaching step, following separation from pregnant solution, was washed in a mild acid solution. Such a process may be described as a two-step process.

1-1-1. Optimization of the Relevant Factors of Ferric Sulfate Leaching Step

With this object in view, the present study provides a process of leaching the xenotime concentrate comprising the steps of forming an acidic solution containing ferric ion; and contacting the acidic solution containing ferric ion with the xenotime concentrate to extract elements by hydrolytic leaching with ferric sulfate wherein ferric iron is precipitated to generate acid for the leaching step. In such process, ferric sulfate has a dual role acting as a leaching reagent, and providing acid by precipitation to supplement an external acid addition to the leaching step.

The chemistry of ferric iron precipitation and formation sulfuric acid proceeds as follows and represented by Eq. (5):

$$Fe_2(SO_4)_3+6H_2O \rightleftharpoons 2Fe(OH)_3+3H_2SO_4 \quad (5)$$

The chemistry of ferric sulfate formation proceeds as follows by Eq. (6):

$$Fe_2O_3+3H_2SO_4 \longrightarrow Fe_2(SO_4)_3+3H_2O \quad (6)$$

(i) Effect of Solid/Liquid Ratio (S/L)

The effect of S/L ratio (xenotime concentrate/$Fe_2(SO_4)_3$ conc. ratio) upon the breakdown of the working xenotime concentrate was studied with S/L ratios ranging from 1/5 to 1/25. The other variables were fixed at 4 g/*l* concentration of ferric sulfate and an agitation time for 6 h at temperature of 35 °C. The obtained leaching efficiencies of U and REEs are plotted in Fig. 1.

From the obtained data, under the applied leaching conditions, the leaching efficiency of U and REEs increased from 5.5 to 22.8% and 8.8 to 28.5%, respectively by increasing the S/L ratio from 1/5 to 1/20. Further, increase in the S/L ratio at 1/25 decreased the leaching efficiency of U and REEs. This decrease may be due to formation of jarosite and goethite mineral forms and their other assembly minerals. Therefore, it can be concluded that the S/L ratio of 1/20 would be considered as an optimum S/L ratio for leaching of uranium and the rare earths.

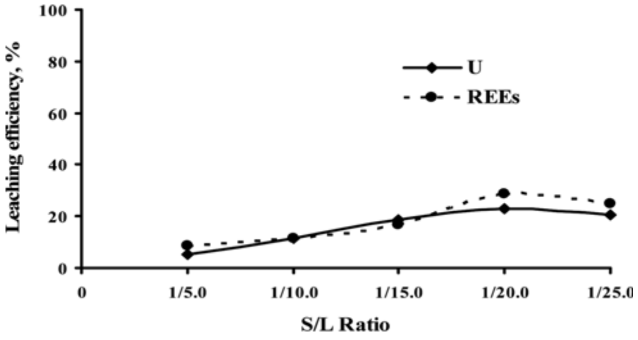


Fig. 1. Effect of S/L ratio upon the breakdown of the working xenotime concentrate.

(ii) Effect of Agitation Time

To study the effect of agitation time upon the leaching efficiency of the working xenotime concentrate, a set of leaching experiments was performed at different time periods ranging from 6 to 36 h. The other leaching conditions were fixed at a ferric ion conc. of 4 g/*l* and 1/20 S/L ratio at temperature of 35 °C. The rare earths and uranium leaching efficiencies are plotted in Fig. 2, indicating that the breakdown of the working xenotime concentrate required long leaching periods, where the leaching efficiency of U and REEs increased from 22.8 to 37.0% and 28.5 to 40.9%, respectively, by increasing the leaching time from 6 to 30 h. Further, increase in the time at 36 h did not improve the leaching efficiency of U and REEs thereafter. Therefore, it can be concluded that an agitation time of 30 h would be considered as an optimum time for leaching of U and REEs from the working xenotime concentrate.

(iii) Effect of Leaching Temperature

The effect of leaching temperature upon the leaching efficiency of uranium and rare earths elements from the working xenotime concentrate was investigated in the range of 35 to 75 °C under the fixed experimental conditions of 4 g/*l* ferric sulfate conc. and S/L ratio of 1/20 at a leaching time of 30 h. The obtained data are illustrated in Fig. 3.

From the obtained results, the dissolution efficiencies of metals increased gradually by increasing leaching temperature from 35 to 65 °C where they attained 54.7% for U with 58.5% for REEs. Extending leaching temp. up to 75 °C has an adverse effect on the dissolution efficiencies of U and REEs where the dissolution efficiencies decreased to 50.2% for U and 52.9% for REEs. This decrease may be due to formation of some complexes of polysulfates, which might have led to U and REE re-precipitation. Therefore, it can be concluded that a leaching temperature of 65 °C would be considered as an optimum temperature for leaching of U and REEs from the working xenotime concentrate.

(iv) Effect of Ferric Sulfate Concentration

To study the effect of $Fe_2(SO_4)_3$ conc. upon the breakdown of the working xenotime concentrate, a set of leaching experiments was performed using different ferric salt concentrations ranging from 4 to 12 g/*l*. The other leaching conditions were fixed at temperature of 65 °C, S/L ratio of 1/20 and leaching time of 30 h. The obtained

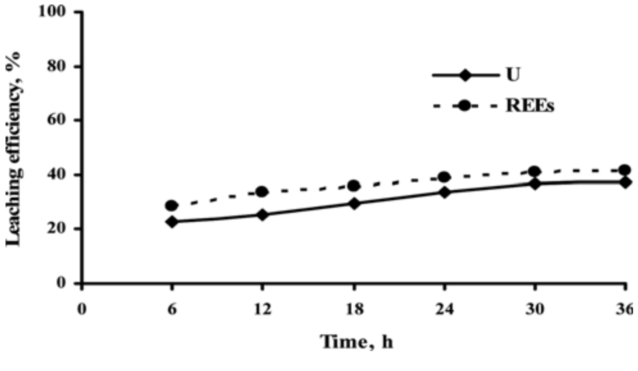


Fig. 2. Effect of agitation time upon the breakdown of the working xenotime concentrate.

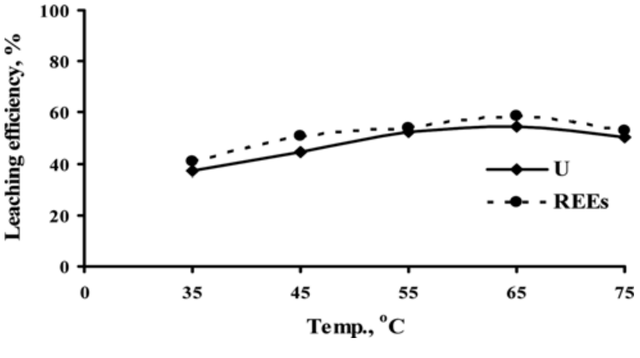


Fig. 3. Effect of leaching temperature upon the breakdown of the working xenotime concentrate.

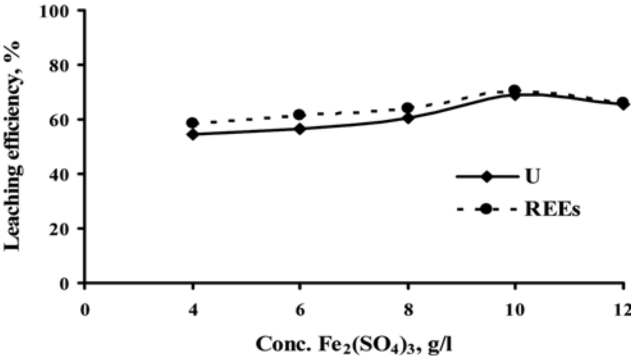


Fig. 4. Effect of ferric sulfate conc. upon the breakdown of the working xenotime concentrate.

leaching efficiencies of U and REEs are plotted in Fig. 4.

From the obtained data, under the applied leaching conditions, the leaching efficiency of U and REEs increased from 54.7 to 68.8% and 58.5 to 70.5%, respectively, by increasing the reagent concentration from 4 to 10 g/*l*. Further, increase in the leaching conc. at 12 g/*l* decreased the leaching efficiency of U and REEs. This decrease may be due to formation of jarosite, goethite minerals forms and their other assembly minerals. Therefore, it can be concluded that a reagent conc. of 10 g/*l* would be considered as an optimum concentration for leaching of uranium and the rare earths.

1-1-2. The Washing Step of the Slurry

After studying the optimum leaching conditions of the xenotime concentrate with ferric sulfate using distilled water in the washing of the obtained slurry, different concentrations of sulfuric acid were used and suggested an alternative washing type from the same acid formed through the leaching procedure to improve the leaching efficiencies of U and REEs. So, a series of washing experiments were performed using different concentrations of sulfuric acid in the range of 10 to 70 g/*l* with a fixed volume (25 ml). The other leaching conditions were fixed at temperature of 65 °C, ferric ion concentration of 10 g/*l*, S/L ratio of 1/20 and leaching time of 30 h. The obtained leaching efficiencies of U and REEs are tabulated in Table 2.

From the obtained data, under the applied washing conditions, the leaching efficiency of U and REEs increased from 72.3 to 96.5% and 77.5 to 97.1%, respectively, by increasing the sulfuric acid concentration from 10 to 70 g/*l*. So, the used acid is better than distilled water in washing of the obtained slurry. This is due to remobilizing U and REEs and preventing their precipitation in the form of jarosite, goethite and their assembly compounds. Therefore, it can be concluded that the washing by 70 g/*l* H_2SO_4 would be considered as an optimum concentration for almost complete extraction of uranium and rare earths elements.

Table 2. Effect of dist. water and sulfuric acid conc. upon the washing of the obtained slurry

Washing type	Leaching efficiency, %	
	U	REEs
Dist. water	68.8	70.5
10 g/*l* H_2SO_4	72.3	77.5
30 g/*l* H_2SO_4	80.5	82.8
50 g/*l* H_2SO_4	88.8	90.5
70 g/*l* H_2SO_4	96.5	97.1

Table 3. Chemical composition of the prepared REEs and U solution

Metal ion	Conc., g/*l*
U	0.184
REEs	11.200
Fe	0.347
Al	0.078
pH	1.600

From the above studied leaching factors of the working xenotime concentrate, it can be concluded that the optimum conditions for its almost complete breakdown involve:

Ferric sulfate concentration: 10 g/*l*
Leaching time: 30 hrs
Leaching temperature: 65 °C
S/L: 1/20
Washing of the slurry: 70 g/*l* H_2SO_4

1-2. Recovery of REEs and U

A 25 g sample portion of the working xenotime concentrate was subjected to the above studied optimum conditions of its ferric ion leaching. After filtration and proper washing of the obtained slurry by 70 g/*l* H_2SO_4, both the REEs and U have been presented in the sulfate liquor to attain up to volume 1 L. The pH of the obtained sulfate leach liquor was found to be 1.6 and its U assay attained 0.184 g/*l* U, while the assay of Fe and the REEs attained 0.347 g/*l* and 11.20 g/*l*, respectively (Table 3). The dissolution reaction of the xenotime mineral with ferric sulfate can be represented by the following Eq. (7).

$$2REPO_4+3U(PO_4)_2+2Fe_2(SO_4)_3+6H_2O \\ \rightarrow RE_2(SO_4)_3+3UO_2SO_4+4Fe(OH)_3+4P_2O_5 \quad (7)$$

1-2-1. Precipitation of REEs

The prepared sulfate liquor was subjected to REEs precipitation as oxalates leaving uranium in solution using 10% oxalic acid at pH 0.7 followed by its filtration and ignition at 850 °C for 1 h and subjected to an SEM analysis as shown in Fig. 5. In the meantime, RE_2O_3 in the obtained product was spectrophotometrically analyzed against arsenazo III and was found to assay up to about 93.4%.

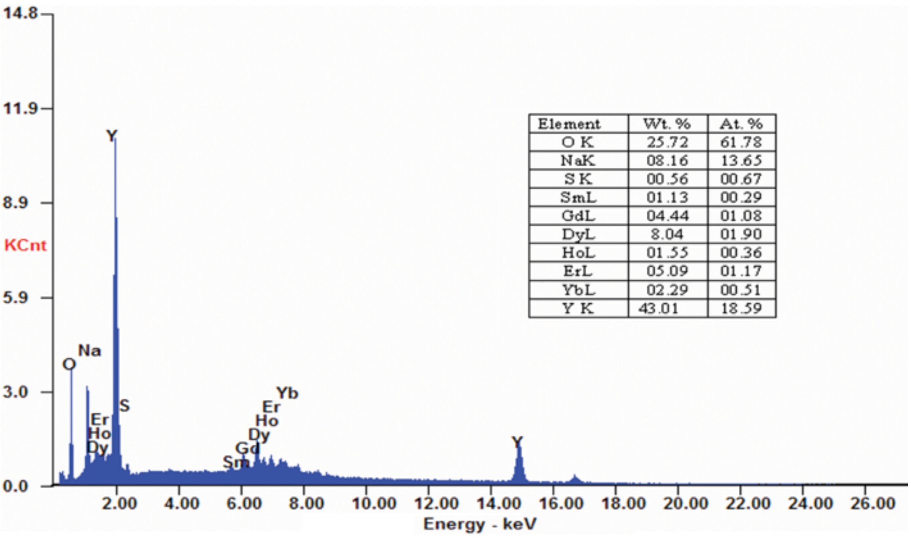
The expected chemical reaction of oxalic acid with REE sulfates can be represented by Eq. (8):

$$RE_2(SO_4)_3+3C_2O_4H_2 \rightarrow RE_2(C_2O_4)_3+3H_2SO_4 \quad (8)$$

2-2-2. Ion Exchange Recovery of Uranium

(i) Uranium Adsorption

After separation of the rare earths oxalates, the sulfate solution left behind was first evaporated till 1 L again and its pH was found to be 0.4. The latter was then subjected to uranium extraction via anion exchange resin technique using Amberlite IRA-400. After pH adjustment at 1.7, 1 liter of the working sulfate leach liquor was allowed to pass through a small glass column packed with Amber-



Element	Wt. %	At. %
O K	25.72	61.78
NaK	08.16	13.65
S K	00.56	00.67
SmL	01.13	00.29
GdL	04.44	01.08
DyL	8.04	01.90
HoL	01.55	00.36
ErL	05.09	01.17
YbL	02.29	00.51
Y K	43.01	18.59

Fig. 5. Analysis of the prepared RE_2O_3 product using SEM qualitative analysis.

lite IRA-400 at a contact time of 6 min. The obtained effluent was collected every 50 ml until saturation of the resin, and the results of uranium analysis in these samples are plotted as an adsorption or loading curve in Fig. 6. From the calculated adsorbed U values, it was revealed the resin capacity under the mentioned conditions attained 83.4 g U/L w.s.r. (wet settled resin).

(ii) Uranium Elution

After washing the working resin column by distilled water, the adsorbed U was eluted using 1 M sodium chloride solution acidi-

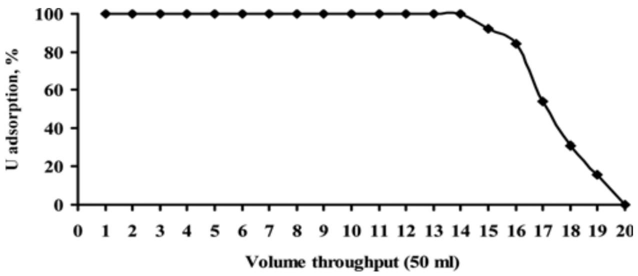


Fig. 6. Adsorption curve of uranium from the working sulfate leach liquor upon Amberlite IRA-400 resin.

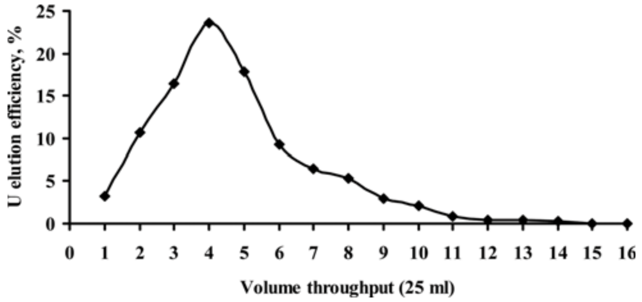
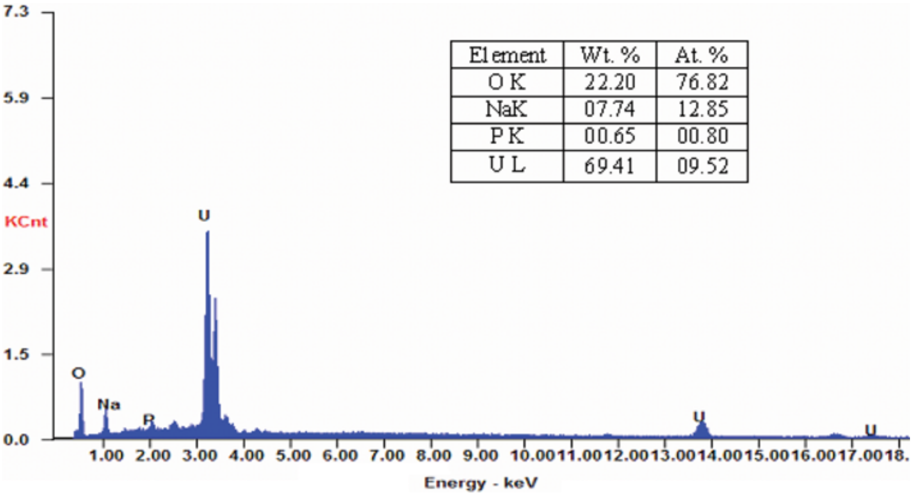


Fig. 7. Uranium elution curve from the working saturated Amberlite IRA-400 resin column using acidified 1 M NaCl solution.



Element	Wt. %	At. %
O K	22.20	76.82
NaK	07.74	12.85
P K	00.65	00.80
U L	69.41	09.52

Fig. 8. SEM qualitative analysis of the prepared $Na_2U_2O_7$ product.

fied with 0.15 N H_2SO_4 using a contact time of 9 min. The obtained eluate was collected every 25 ml and the results of their uranium analysis are plotted in Fig. 7 as the elution curve.

From the obtained results, it is clearly evident that the working resin was saturated from uranium content upon the used column attained 0.16 g. This corresponds at a capacity of 83.4 g U/*l* w.s.r.. The saturation capacity of the working resin was determined theoretically to be about 90 g U/*l* w.s.r.. In other words, the attained capacity in this work represents about 93% of the mentioned saturation capacity.

(iii) Uranium Precipitation

After collecting the obtained eluate samples, the pH of the solution was adjusted to about 7.5 using 10% NaOH solution where uranium was precipitated as sodium di-uranate. After filtration and calcination of the obtained product at 850 °C for 1 h, it was subjected to SEM analysis as shown in Fig. 8. In the meantime, uranium in the obtained product was titrimetrically analyzed against ammonium metavanadate and was found to assay about 70%. Finally, the obtained products of the REEs and U were properly formulated in a novel flowsheet as shown in Fig. 9.

CONCLUSION

The agitation leaching procedure using ferric sulfate salt was applied for treating the xenotime concentrate followed by washing of the obtained slurry in sulfuric acid for REEs and uranium recovery. The optimum conditions of the ferric ion leaching for its breakdown involved 1/20 concentrate/ferric ion ratio and a leaching temperature of 65 °C for 30 h. using 10 g/*l* $Fe_2(SO_4)_3$. After filtration and washing of the obtained slurry by 70 g/*l* H_2SO_4, both the U and REEs were dissolved in the solution with their corresponding leaching efficiencies of 96.5 and 97.1%, respectively. From the latter, the REEs were precipitated as their oxalates, while uranium was left behind. The latter was extracted using Amberlite IRA-400 resin and from the obtained elute, U was precipitated as sodium diuranate ($Na_2U_2O_7 \cdot 2H_2O$). The obtained RE oxalates and the sodium diuranate were properly calcined at 850 °C for 1 h to obtain on RE_2O_3 and $Na_2U_2O_7$. The two products were then analyzed by both SEM and titrimetrically.

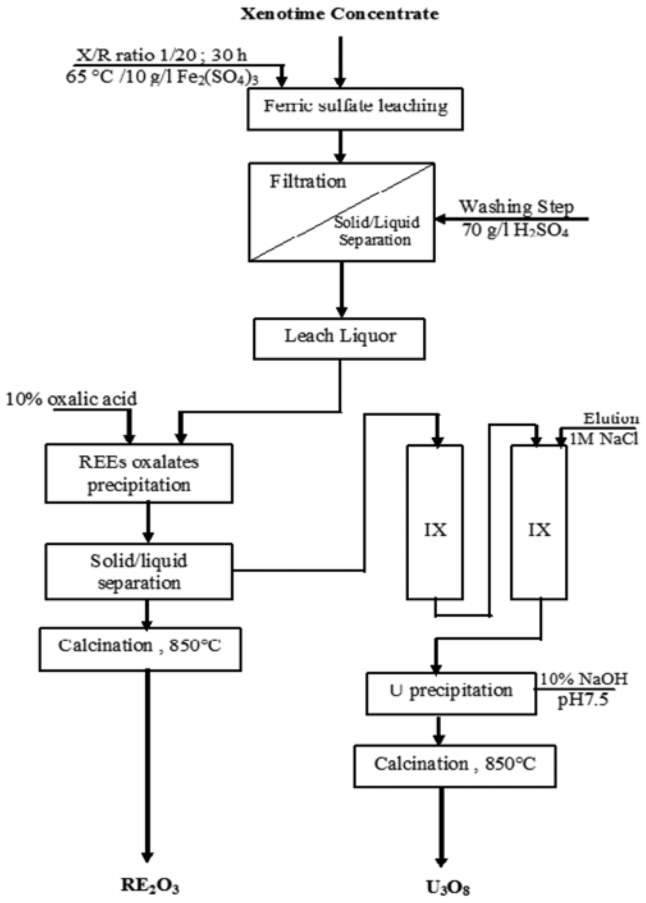


Fig. 9. A novel procedure for processing of the xenotime concentrate of SW Sinai.

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