Recovery of Uranium and Rare Earth Elements from Western Desert Phosphate Rocks with EDTA and Nitric Acid Solutions

4 authors:

Ahmed R. Bakry
Nuclear Materials Authority of Egypt
32 PUBLICATIONS **129** CITATIONS

SEE PROFILE

Sayed Hady
Nuclear Materials Authority of Egypt
16 PUBLICATIONS **118** CITATIONS

SEE PROFILE

Mohamed demerdash
Nuclear Materials Authority of Egypt
12 PUBLICATIONS **79** CITATIONS

SEE PROFILE

A. I. L. Abd El Fatah
Nuclear Materials Authority of Egypt
19 PUBLICATIONS **92** CITATIONS

SEE PROFILE

Recovery of Uranium and Rare Earth Elements from Western Desert Phosphate Rocks with EDTA and Nitric Acid Solutions

A. R. Bakry[a], M. Demerdash[a], E. A. Manaa[a], and A. I. L. Abd El Fatah[a,*]

[a] Nuclear Materials Authority, El Maadi, Cairo, PB530 Egypt
*e-mail: nuclic_science@yahoo.com

Abstract—Dissolution of uranium and rare earth elements from the Abu-Tartur phosphate rocks, Egypt using a mixed EDTA + nitric acid solution was studied. The effect of factors such as particle size, EDTA and nitric acid concentrations, agitation time, solid to liquid ratio, and dissolution temperature was studied, and the dissolution conditions were optimized. The optimized dissolution conditions were applied to a representative sample of the weathered rock assaying 1067 ppm rare earth elements. Before being processed, the representative sample was completely characterized using mineralogical and chemical analyses. From the obtained leach liquor, uranium was recovered by anion exchange with Duolite A 101D resin, and after uranium adsorption the rare elements were recovered by cation exchange with Dowex 50W-X8 resin.

Keywords: uranium, rare earth elements, phosphate rock, dissolution, ethylenediaminetetraacetic acid, nitric acid, ion exchange resins

INTRODUCTION

The Abu-Tartur phosphate deposit (Western desert, Egypt) is one of the largest phosphate deposits in the world. It is geomorphologically localized in the southeastern portion of the Abu-Tartur plateau (600 km from Cairo) and is the largest source of uranium and rare earth elements (REEs) in Egypt [1]. Abu-Tartur plateau has a huge reserve of phosphate ore reaching up to billion tons [2]. Most of the world's phosphate rocks are of sedimentary origin and are primarily composed of the apatite group in association with a wide assortment of some accessory minerals, fluorides, carbonates, clays, quartz, silicates, and metal oxides [3–5]. In general, the phosphate rock (PR) found in the earth's crust contains relatively high levels of environmentally toxic elements as well as precious elements having diverse applications in green and sustainable industrial energy production. Uranium is one of such elements found in PR, usually at levels between 50 and 200 ppm, but some deposits contain significantly higher levels of uranium (>800 ppm) [6]. Uranium contained in PR is dispersed during the use of fertilizers on agricultural soils; it contaminates soil and water and poses hazard to health via food chain [7]. Uranium is an essential element in the atomic energy program. In spite of its low concentration in phosphate ores, its recovery is significant from environmental and economical viewpoints [8]. Along with environmental impact minimization and human health protection, extraction of uranium from PR can reduce mine waste and extend resource use [9]. The previously used procedure for processing of phosphate ores involved hydrochloric acid leaching to obtain monocalcium phosphate (MCP) in the presence of a reductant to keep U(IV) in the insoluble form [10–14]. Recovery of U from Egyptian phosphorites is conducted only on an experimental scale by the Nuclear Materials Authority of the Egyptian government [15]. There are several techniques for recovering uranium from its leach liquors, including ion exchange [16, 17].

Phosphate rock associated with trace amount of REEs was considered as one of the potential new resources for REEs due to its huge reservation, about 300 billion tons in total [18, 19]. The recovery of REEs from phosphate rock attracted much attention recently [20].

Table 1. Chemical composition of the working Western Desert PR sample

Oxide	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	MnO	F	Cl	SO_3	L.O.I[a]	Total
Content, wt %	5.89	1.72	8.08	1.01	45.61	0.58	0.16	0.29	21.82	0.34	0.76	0.17	3.88	9.20	99.51

[a] L.O.I.: loss on ignition at 1000°C.

Table 2. Content of trace elements in Western Desert PR sample

Element	Sc	Cr	V	Co	Ni	Cu	Zn	As	Cd	Sr	U	Th	La	Ce	Nd	Sm	Yb	Y	Total REEs
Concentration, mg kg^{-1}	76	77	103	54	40	24	72	41	2	1495	30	18	201	337	176	42	19	283	1067

The diffusion-controlled process of REEs recovery is characterized by the activation energy of 11.76 kJ mol^{-1} [21]. The kinetics of REEs leaching from phosphate ore using HNO_3 were described [22].

On the other hand, polyamino polycarboxylates such as EDTA, TETA, or DOTA form strong complexes with alkaline earth elements, trivalent lanthanides, and actinides in aqueous solution [23, 24]. The dissolution of the phosphate rock is favored by the formation of stable $Ca(EDTA)^{2-}$ complexes in the solution [25, 26]. The ion exchange sorption of REE complexes with EDTA on an anion exchanger in the H_2EDTA^{2-} form can be performed.

This study is aimed at analyzing and optimizing the conditions for uranium and REEs dissolution from Abu-Tartur phosphate rocks using a binary EDTA + nitric acid solution.

EXPERIMENTAL

Materials and Chemicals

The phosphate samples were obtained from Abu-Tartur (Egypt); samples with different particle size were taken. The Duolite A 101D strongly basic anion-exchange resin (Auchtel Products, Mumbai, India) was used in the chloride form. The Dowex 50W-X8 cation-exchange resin (Merck) was used in the hydrogen form. Ethylenediaminetetraacetic acid disodium salt (Na_2EDTA), hydrochloric acid (37%), sulfuric acid (98%), and nitric acid (60%) were purchased from Merck. First, the calcareous Abu-Tartur phosphate sample was completely analyzed for determine major oxides and trace elements by X-ray fluorescence (XRF) technique with a PW 1510 Philips experimental

unit equipped with an automatic sample changer (JEOL-JSX-3222 Element Analyzer; Atomic Energy Authority, Cairo, Egypt). The device was interfaced with a computer system; X-40 program for spectroscopy was used. The detection limit was 5.0 ppm. Atomic spectrophotometry was used for identification and confirmation of trace elements including uranium and REEs [27]. The results obtained are given in Tables 1 and 2; as can be seen, P_2O_5 assays only 21.8%, the CaO content reaches 45.6%, and the loss in ignition amounts to 9.2%; U assays only 30 ppm, and the REEs content reaches 1067 ppm.

Experimental Procedures

The working sample was collected from an experimental mine of Abu Tartur plateau located in the Western Desert of Egypt. Five different size fractions (700, 500, 150, 74, and 63 μm) were obtained by sieving through ASTM standard sieves.

Dissolution of U and REEs. In each experiment, a weighed portion of the representative sample was agitated in a certain volume of the solution for a definite time period at definite S/L ratio. In these experiments, a 40-g portion of the fine Abu-Tartur representative sample was put in a 250-mL beaker. A number of experiments were carried out with different values of EDTA concentration, reaction time, solid/liquid ratio, and temperature. The reaction mixture was agitated using a mechanical stirrer. After that, the mixture was washed several times with distilled water to remove any soluble materials. The EDTA concentration ensuring the maximum dissolution of U and REEs was subsequently used for optimizing the other parameters.

Recovery of U and REEs. *Recovery of uranium.* Duolite A 101D anion exchange resin (2 mL) in the form of wet settled resin (w.s.r.) was packed in a small glass column 2 cm in diameter, the leach liquor of U/REEs was passed through it at a rate of 2 mL min^{-1}, and 100-mL effluent samples were collected for uranium analysis. In so doing, uranium was adsorbed on the resin and was absent in the effluent solution, whereas REEs remained in the effluent solution and were absent on the resin. After the adsorption, uranium was eluted with a 1 M NaCl solution acidified with 0.15 N H$_2$SO$_4$ at a flow rate of 3 mL min^{-1}, and 10-mL eluate samples were collected for uranium analysis.

Recovery of REEs. The effluent solution after the uranium adsorption, which contained the leached REEs, was passed through the Dowex 50W-X8 cation exchange resin (w.s.r.). 5 mL of the resin was fixed in a glass column 2 cm in diameter, and the REE solution was passed through it at a flow rate of 0.5 mL min^{-1}. 100-mL effluent samples were taken for REEs analysis. After complete adsorption, the resin was washed with 2 M HCl to remove all impurities except REEs; then, the REEs were eluted with 4 M HCl at a flow rate of 2 mL min^{-1}, and 10-mL eluate samples were collected for REEs analysis.

Analytical Control

The uranium content was determined by oxidimetric titration with ammonium metavanadate. For this purpose, uranium was preliminarily reduced with (NH$_4$)$_2$Fe(SO$_4$)$_2$ in the presence of diphenylamine sulfonate indicator until it changed color to slightly violet [28]. Also, the uranium content was determined spectrophotometrically with Arsenazo III (λ = 655 nm). The REEs content was determined spectrophotometrically with Arsenazo III [29].

Analysis of the Product

Finally, the precipitated sodium diuranate and rare earth oxide products were analyzed in the NMA Labs with an XL30 Philips type scanning electron microscope (SEM); the results were confirmed by ICP-OES [30].

RESULTS AND DISCUSSION

Optimization of Dissolution Conditions

Effect of grain size. The influence of phosphate rock fineness on the dissolution of U and REEs from phosphate rock was studied using fractions of 700,
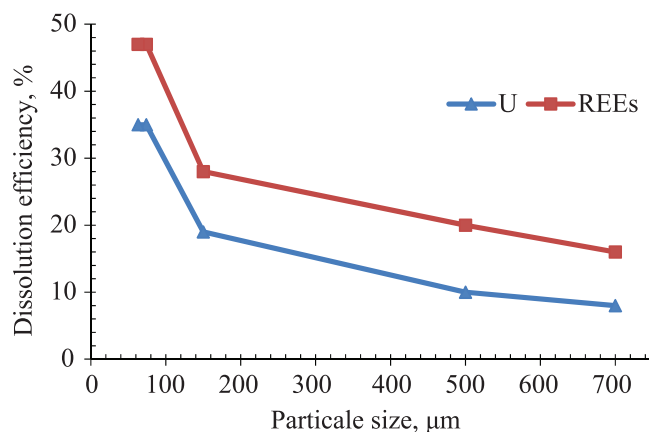

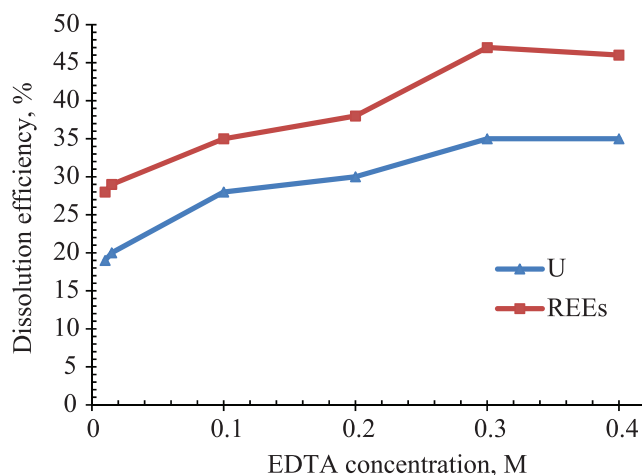**Fig. 1.** (Color online) Effect of particle size on the dissolution of U and REEs from Abu-Tartur PR.

Fig. 2. (Color online) Effect of EDTA concentration on the dissolution of U and REEs from Abu-Tartur PR (ore particle size 74 μm, S/L ratio 1/3, dissolution time 30 min, 25°C).

500, 150, 74, and 63 μm size. The other parameters were fixed: 0.3 M EDTA, S/L = 1/3, room temperature. The experimental results are shown in Fig. 1 as the dependence of the U and REEs dissolution efficiency on the particle size. As can be seen, the dissolution efficiency increases with a decrease in the particle size from 700 to 74 μm, which may be due to an increase in the active surface area, and does not further change in going to 63 μm; thus, 74 μm is the optimum phosphate particle size, and it was chosen for the other experiments on the U and REEs dissolution.

Effect of EDTA concentration. The effect of EDTA concentration on the U and REE dissolution efficiency was studied at the EDTA concentration in the range from 0.01 to 0.4 M. The other dissolution conditions were kept constant: sample weight 40 g, ore particle size
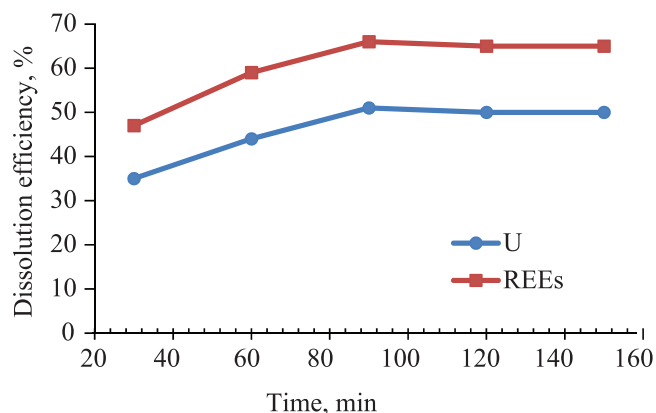
Fig. 3. (Color online) Effect of agitation time on the dissolution of U and REEs from Abu-Tartur PR (ore particle size 74 μm, 0.3 M EDTA, S/L ratio 1/3, 25°C).
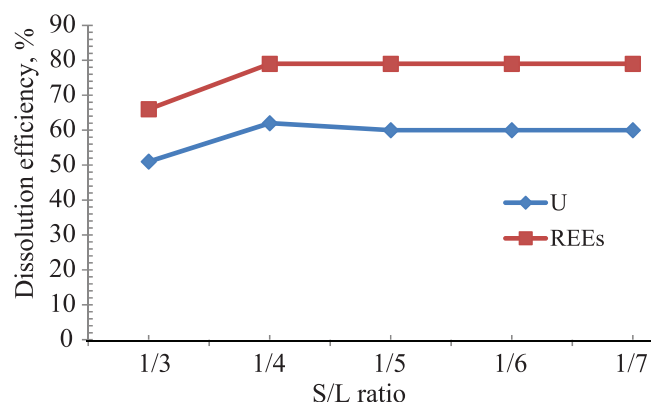

Fig. 4. (Color online) Effect of S/L ratio on the dissolution of U and REEs from Abu-Tartur PR (ore particle size 74 μm, 0.3 M EDTA, dissolution time 90 min, 25°C).
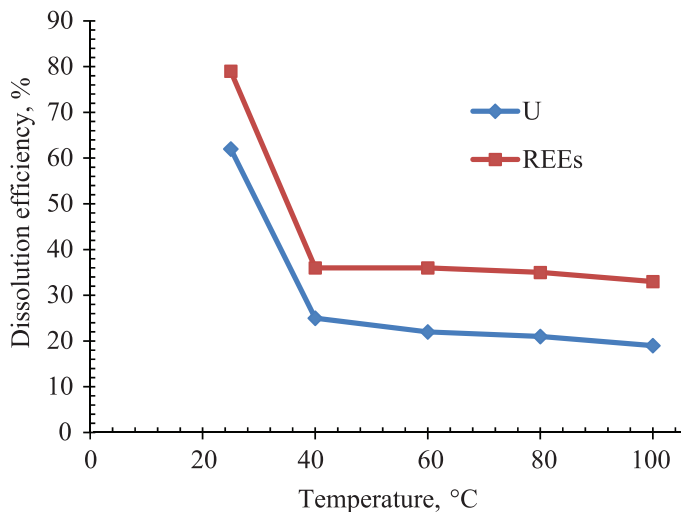

Fig. 5. (Color online) Effect of temperature on the dissolution of U and REEs from Abu-Tartur PR (ore particle size 74 μm, 0.3 M EDTA, S/L ratio 1/4, dissolution time 90 min).

74 μm, solid/liquid ratio 1/3, leaching time 30 min, and temperature 25°C. The results are shown in Fig. 2. As can be seen, with an increase in the EDTA concentration from 0.01 to 0.4 M the U dissolution efficiency increased from 19 to 35% and the REE dissolution efficiency increased from 28 to 47 %. Similar result was obtained already with 0.3 M EDTA, and this concentration was chosen as optimum. The residue mainly consists of SiO_2, CaO, Fe_2O_3.

Effect of dissolution time. To evaluate the influence of time on the dissolution of U and REEs from Abu-Tartur phosphate rock, the time was varied from 30 to 150 min. The other conditions were kept constant: sample weight 40 g, EDTA concentration 0.3 M, S/L ratio 1/3, and room temperature. Figure 3 shows that the U and REEs dissolution efficiency increases to 51 and 66%, respectively, at 90 min and does not appreciably increase at longer contact times; therefore, we used the agitation time of 90 min in all the experiments.

Effect of solid/liquid ratio. To evaluate the effect of the ore weight to solution volume ratio on the U and REE dissolution efficiency, we kept the ore sample weight constant (40 g) and varied the solution volume so that the S/L ratio ranged from 1/3 to 1/6. The other variables were kept constant: EDTA concentration 0.3 M, dissolution time 90 min, and room temperature. Figure 4 shows that the best S/L ratio is 1/4; at this ratio, the U and REE dissolution efficiency reaches 62 and 79%, respectively.

Effect of temperature. To determine the optimum temperature, we performed experiments at 25, 40, 60, 80, and 100°C, keeping the other factors on the optimum level (EDTA concentration 0.3 M, dissolution time 90 min, S/L ratio 1/4). Figure 5 shows that the best results are reached at room temperature. Probably, at elevated temperatures the EDTA complexes of U and REEs are unstable. Therefore, all the experiments were made at room temperature.

Effect of Mineral Acids Added to EDTA on the Dissolution of U and REEs

Different mineral acids (HCl, H_2SO_4, and HNO_3) were added to 0.3 M EDTA. The other variables were fixed: sample weight 40 g, ore particle size 74 μm, solid to liquid ratio 1/4, dissolution time 90 min, and 25°C. The best results were reached with HNO_3 (Figs. 6 and 7). With an increase in the nitric acid concentration from
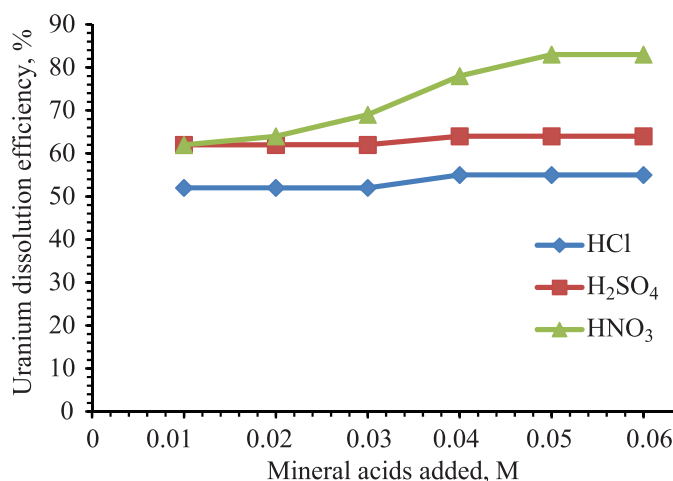
Fig. 6. (Color online) Effect of mineral acids added to EDTA on the dissolution of U from Abu-Tartur PR (ore particle size 74 μm, 0.3 M EDTA, S/L ratio 1/4, dissolution time 90 min, room temperature).
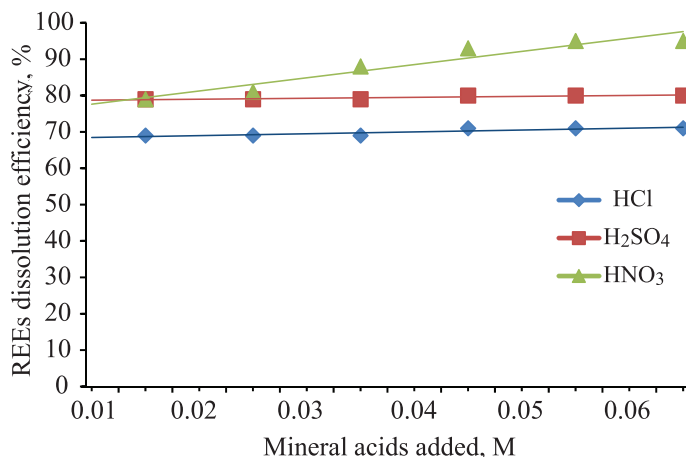

Fig. 7. (Color online) Effect of mineral acids added to EDTA on the dissolution of REEs from Abu-Tartur PR (ore particle size 74 μm, 0.3 M EDTA, S/L ratio 1/4, dissolution time 90 min, room temperature).

0.01 to 0.05 M, the U and REEs dissolution efficiency increases to 83 and 95%, respectively; it does not change with a further increase in the HNO_3 concentration to 0.06 M. The U and REEs dissolution efficiency is considerably higher than when using EDTA. The 0.05 M nitric acid concentration in combination with 0.3 M EDTA was chosen as the optimum.

Ion-Exchange Recovery of Uranium and REEs

The leach liquor was obtained under optimum conditions: PR particle size 74 μm, (0.3 M EDTA/0.05 M HNO_3), agitation time 90 min, S/L ratio 1/4, and room temperature. The leach liquor was analyzed colorimetrically to determine major oxides and by ICP-OES to determine trace elements. The results are given in Table 3. The leach liquor was passed through the anion-exchange resin to recover uranium and then through the cation-exchange resin to recover REEs.

Ion-exchange recovery of uranium. *Uranium adsorption.* After the dissolution of uranyl and REEs with EDTA + nitric acid, uranium was then adsorbed onto Duolite A 101D anion-exchange resin. The first 100-mL portions of the effluent contained virtually no
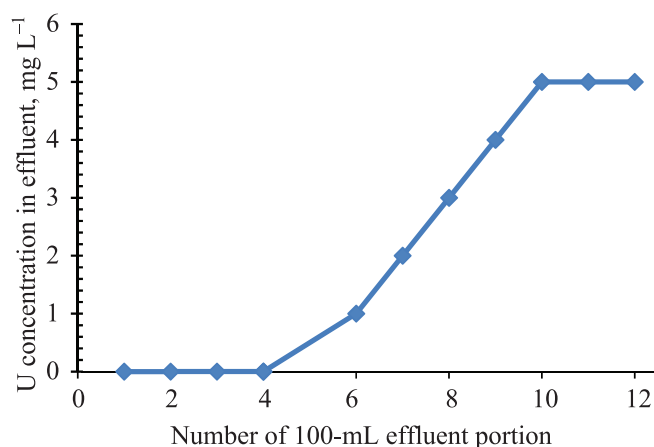

Fig. 8. Uranium adsorption onto Duolite A 101D anion-exchange resin.

uranium; i.e., uranium was completely adsorbed on Duolite A 101D (Fig. 8). The uranium breakthrough became appreciable in the sixth portion of the effluent sample and complete in the tenth portion.

Uranium elution. The uranium adsorption was followed by elution with a 1 M NaCl solution acidified with 0.15 N H_2SO_4. The elution curve is shown in Fig. 9;

Table 3. Chemical composition of Abu-Tartur PR leach liquor

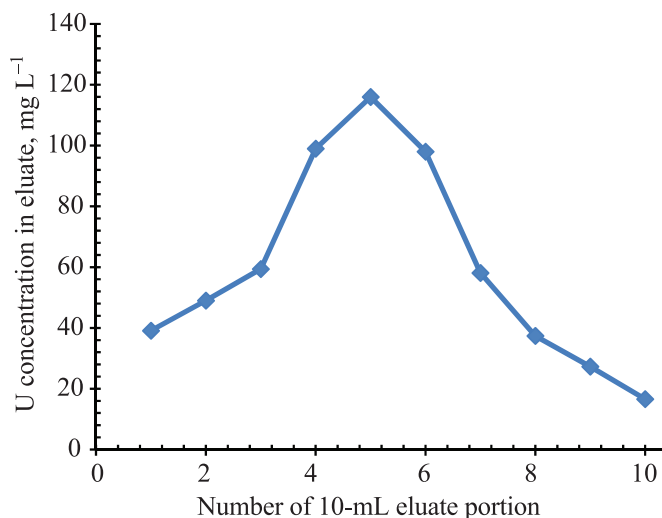| Ion	Si^{4+}	Al^{3+}	Fe^{3+}	Ca^{2+}	Mg^{2+}	NO_3^-	P^{5+}	UO_2^{2+}	REE^{3+}
Concentration, g L^{-1}	0.21	0.13	0.25	4.1	0.35	3.05	2.62	0.029	1.02

Fig. 9. (Color online) Uranium elution from the loaded Duolite A 101D resin.

it is bell-shaped with the maximum in the fifth eluate portion. The uranium elution was almost complete.

Ion-exchange recovery of REEs. *REE adsorption.* The effluent after Duolite A 101D anion-exchange resin was passed through the Dowex 50W-X8 cation-exchange resin for the REE adsorption. The effluent was collected in 100-mL portions and analyzed for the REE content; the results are shown in Fig. 10. The REE breakthrough was observed starting from the fifth portion, and starting from the tenth portion it became complete.

REE elution. First, the resin was washed with 2 M HCl to remove possible impurities adsorbed jointly with REEs, and then it was eluted with
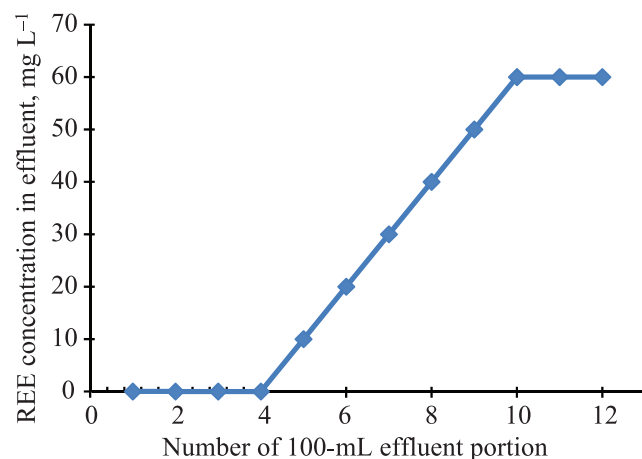
4 M HCl to desorb REEs. The REE elution curve is shown in Fig. 11; it is bell-shaped with the maximum in the fifth eluate portion. The REE elution was virtually complete.

Uranium and REE Precipitation

Uranium concentrate product. The eluate from Duolite A 101D was alkalized to pH 7 with sodium hydroxide to precipitate sodium diuranate ($Na_2U_2O_7$). The product was calcined at 750°C for 1 h, and the uranium content was determined chemically to be 69.0%. The product purity was 91.90%. The final uranium product was identified by scanning electron microscopy (SEM), Nuclear Material Authority, Cairo, Egypt (Fig. 12). The chemical composition of the uranium precipitate given by EDX analysis was follows: O 22.20%, Na 7.74%, P 0.65%, and U 69.41%. The results of ICP-OES analysis are given in Table 4.

Total rare earth oxides product (ΣREO). The total rare earth oxides (ΣREO) were prepared in the form of a light brownish powder from the uranium-free effluent. The eluate from Dowex 50W-X8 was partially neutralized with ammonia to pH 1.5, and 30% oxalic acid was added to pH 0.8. In so doing, REE oxalates completely precipitated. The precipitation efficiency reached 97%. The oxalate precipitate was filtered off, properly washed with distilled water, and calcined at 850°C for 1 h to obtain total rare earth
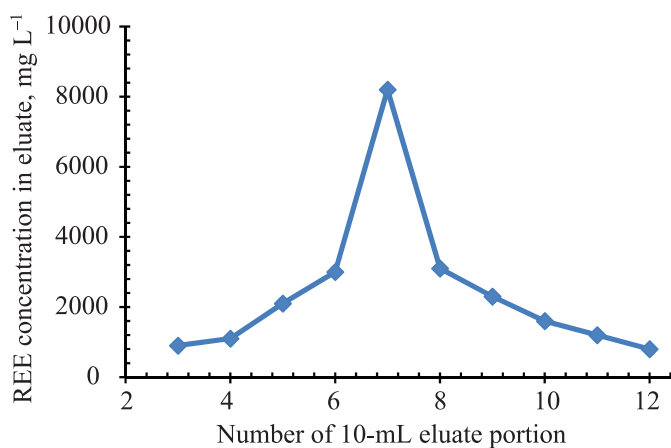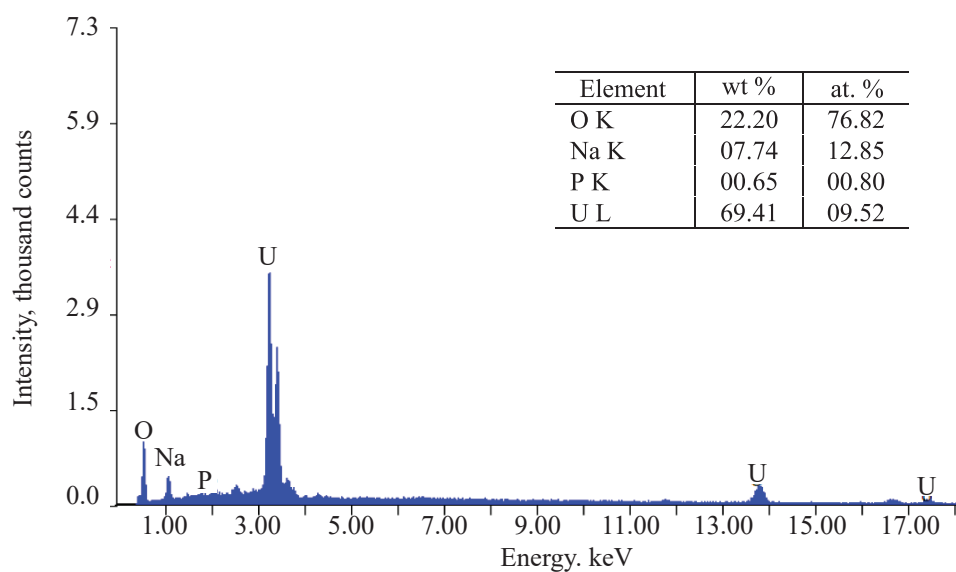

Fig. 10. REE adsorption onto Dowex 50W-X8 cation-exchange resin.

Fig. 11. REE elution from the loaded Dowex 50W-X8 resin column.

Fig. 12. SEM analysis of the prepared U product.

Element	wt %	at. %
O K	22.20	76.82
Na K	07.74	12.85
P K	00.65	00.80
U L	69.41	09.52
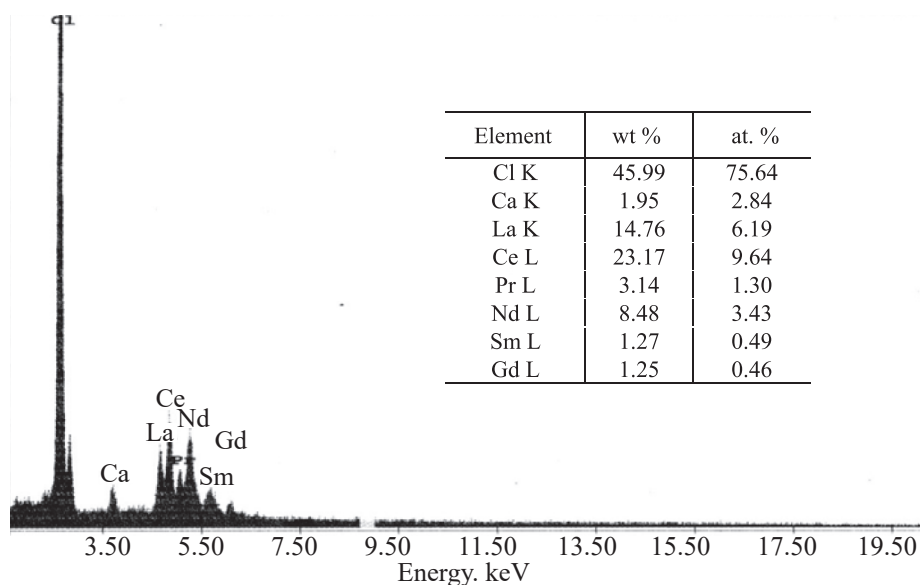
Fig. 13. SEM analysis of the prepared RE$_2$O$_3$ product.

Element	wt %	at. %
Cl K	45.99	75.64
Ca K	1.95	2.84
La K	14.76	6.19
Ce L	23.17	9.64
Pr L	3.14	1.30
Nd L	8.48	3.43
Sm L	1.27	0.49
Gd L	1.25	0.46

Table 4. ICP-OES specification of U concentrate produced after elution from Abu-Tartur PR

Element	U	Si	Al	Fe	Ca	Mg	Co	Ni	Cu
Content, %	69.25	0.004	0.0131	0.009	0.0431	0.0223	0.0015	0.0016	0.0041
Element	Cd	Cr	Na	K	Mn	V	Zn	Pb	Sr
Content, %	0.0007	0.0037	5.019	0.0150	0.0114	0.0053	0.0041	0.0045	0.0048

Table 5. ICP-OES specification of the total rare earth oxide (ΣREO) after elution from Abu-Tartur PR[a]

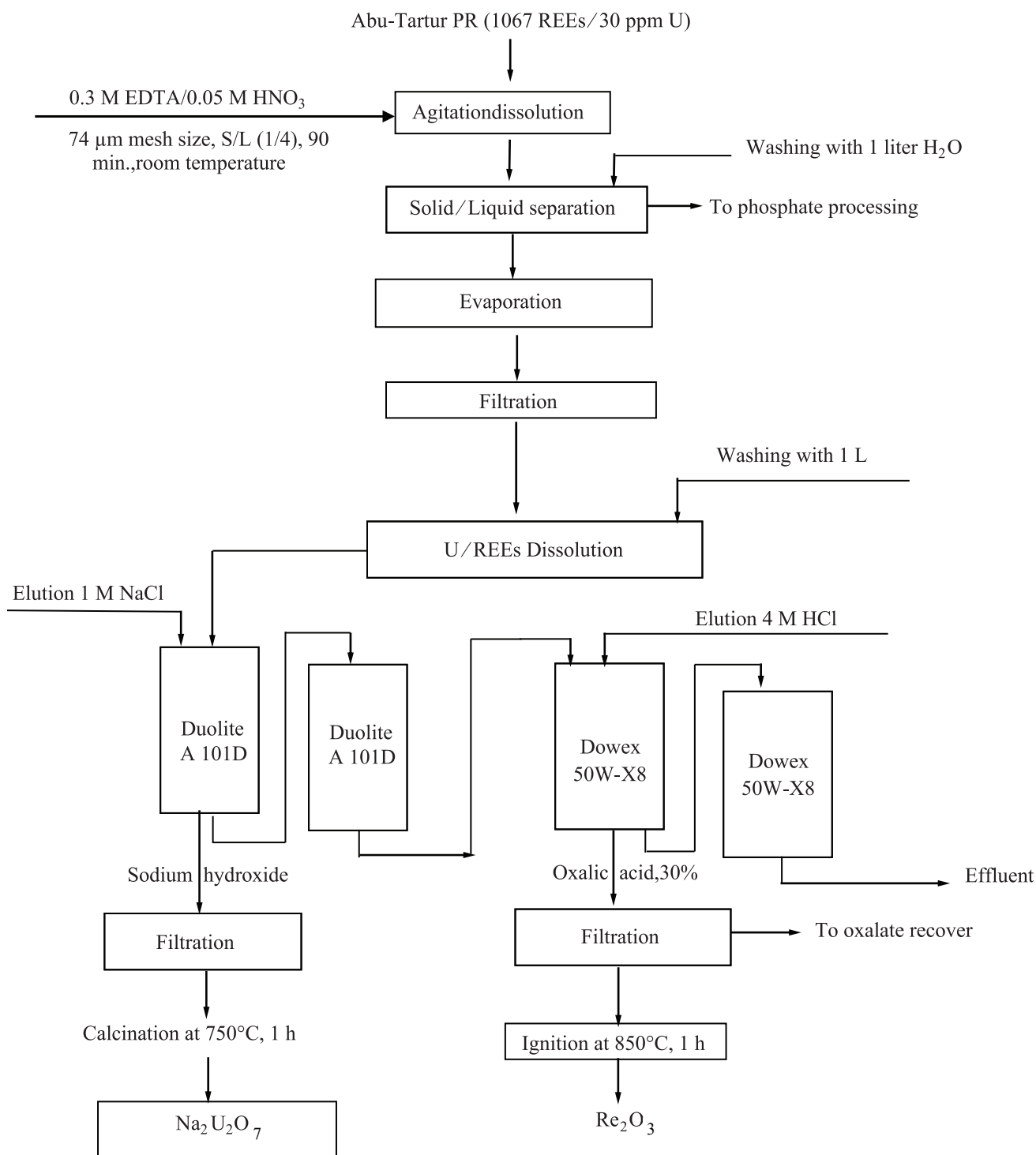| REO	La$_2$O$_3$	Ce$_2$O$_3$	Pr$_2$O$_3$	Nd$_2$O$_3$	Sm$_2$O$_3$	Tb$_2$O$_3$	Gd$_2$O$_3$	Y$_2$O$_3$
Content, wt %	21.76	39.90	2.45	9.85	1.66	0.10	0.82	N.D.
REO	Eu$_2$O$_3$	Dy$_2$O$_3$	Ho$_2$O$_3$	Er$_2$O$_3$	Tm$_2$O$_3$	Yb$_2$O$_3$	Lu$_2$O$_3$	ΣREO
Content, wt %	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	76.54

[a] N.D.: not detected.

Fig. 14. Flow diagram for the recovery of uranium concentrate + ΣREO from Abu-Tartur PR.

oxides (ΣREO). Analysis of the total rare earth oxides in the product by ICP-OES and EDX gave the ΣREO content of 76.54% (Table 5, Fig. 13). The major components of the product were valuable light rare earths (lanthanum, cerium, praseodymium, neodymium, and samarium). The content of Ce_2O_3, La_2O_3, and Nd_2O_3 was 39.9, 21.76, and 9.85%, respectively. Impurities of P_2O_5, CaO, MgO, and Fe_2O_3 were also detected.

CONCLUSION

Our study of the dissolution behavior of uranium and REEs from the Abu-Tartur PR shows that the optimum conditions ensuring almost complete dissolution of both these two metal values are as follows: parical size 74 µm, 0.3 M EDTA + 0.05 M HNO_3, agitation time 90 min, S/L ratio 1/4, and room temperature. From the leach liquor, uranium and REEs were successfully recovered by ion exchange. Duolite A 101D anion-exchange resin was used for uranium recovery, and Dowex 50W-X8 cation-exchange resin, for the REEs recovery from the uranium-free effluent. The flow diagram of the process is shown in Fig. 14.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

FUNDING

REFERENCES

1. Eid, M.A., Fakhry, A.A., Mahdy, A.A., Eid, K.A., and Broekaert, J.A., *Fresenius' J. Anal. Chem.,* 1995, vol. 351, pp. 190–196.

2. Al-Wakeel, M.I., *Int. J. Miner. Process.,* 2005, vol. 75, pp. 101–112.
https://doi.org/10.1016/j.minpro.2004.05.004

3. Gupta, C. and Krishnamurthy, N., *Extractive Metallurgy of Rare Earth,* CRC, 2005.

4. Al-Shammari, M.S., Ahmed, I.M., Nayla, A.A., Aly, H.F., Mohamed, G.G., and Abdel Rahman, M.S., *Adv. Environ. Biol.,* 2016, vol. 10(9), pp. 49–53.
http://creativecommons.org/licenses/by/4.0/.

5. Al-Emrajabi, M., Rasmuson, Å.C., Korkmaz, K., and Forsberg, K., *Hydrometallurgy,* 2017, vol. 169, pp. 253–262.
https://doi.org/10.1016/j.hydromet.2017.01.008

6. Paschalidou, P. and Pashalidis, I., *Hydrometallurgy,* 2019, vol. 189, p. 105118.
https://doi.org/10.1016/j.hydromet.2019.105118

7. Habashi, F., Awadalla, F.T., and Zailaf, M., *Chem. Technol. Biotechnol.,* 1986, vol. 36, pp. 259–267.

8. Nriagu, J.O. and Pacyna, J.M., *Nature,* 1988, vol. 333, pp. 134–139.

9. Haneklaus, N., Sun, Y., Bol, R., Lottermoser, B., and Schnug, E., *Environ. Sci. Technol.,* 2017, vol. 51, pp. 753–754.
https://doi.org/10.1021/acs.est.6b05506

10. Alter, I., *Int. Conf. on Peaceful Uses of Atomic Energy (Geneva, 1958),* New York: UN, 1958, vol. 3.

11. Ketzinel, Z., Yakir, D., Rosenberg, J., Shashua, J., Hasid, M., and Volkman, Y., Abstracts of Papers, *Recovery of Uranium: San Paulo Symp., IAEA-SM-135/12,* Vienna: IAEA, 1971.

12. El-Shazly, E.M., El Hazek, N.J., and El Mahrouky, F.A., *Congr. of Phosphate in ARE, Part II, Arabs Min. Petrol. Assoc. Trans.,* 1972, vol. 27, no. 2.

13. Hassan, F. and El-Kammar, A., *Egypt. J. Petrol.,* 1975, vol. 19, pp. 169–178.

14. Kim, J.A., Dodbiba, G., Tanimura, Y., Mitsuhashi, K., Fukuda, N., Okaya, K., Matsuo, S., and Fujita, T., *Mater. Trans.,* 2011, vol. 52, no. 9, pp. 1799–1806.
https://doi.org/10.2320/matertrans.M2011111

15. Ali, M.M., *The Impact of the Mineralogical and Geochemical Characteristics of West Sibaiya Nile Valley Phosphorites on the Extraction of Uranium: PhD Thesis,* Cairo (Egypt): Cairo Univ., 1990.

16. Muller, L.D. and Burton, C.J., *Metall. Congr. Aust.,* 1965, vol. 6, pp. 1151–1163.

17. Ortiz, I., Alonso, A., and Urtiaga, A., *Ind. Eng. Chem. Res.,* 1999, vol. 38, pp. 2450.
https://doi.org/10.1021/ie980686s

18. Zhang, P., *Procedia Eng.,* 2014, vol. 83, pp. 37–51.
https://doi.org/10.1016/j.proeng.2014.09.010

19. Wu, S.X., Wang, L.S., Zhang, P., El-Shall, H., Moudgil, B., and Huang, X.W., *Hydrometallurgy,* 2018, vol. 175, pp. 109–116.
https://doi.org/10.1016/j.hydromet.2017.10.025

20. Wu, S., Zhao, L., Wang, L., Huang, X., Zhang, Y., Feng, Z., and Cui, D., *J. Rare Earths,* 2018.
https://doi.org/10.1016/j.jre.2018.09.012

21. Abd El-Mottaleb, M., Cheira, M.F., Gouda, A.H., and Saiyd, A.A., *Chem. Adv. Mater.,* 2016, vol. 1, no. 1, pp. 33–40.

22. Shengdong, W., Kaixi, J., Xunxiong, J., Lingyong, F., Yanqing, F., Wei, J., and Denggao, Z., *Nonferrous Met. (Extr. Metall.),* 2011, vol. 10, no. 9, pp. 28−31.

23. Anderegg, G., *Critical Survey of Stability Constants of EDTA Complexes,* IUPAC Chemical Data Series, 1976, no. 14.

24. Martell, A.E. and Smith, R.M., *Critical Stability Constants. Amino Acids,* New York: Plenum, 1982.

Bookmark: https://trove.nla.gov.au/version/45272155.

25. Arena, G., Musumeci, S., Purrello, R., and Sammartano, S., *Thermochim. Acta,* 1983, vol. 61, pp. 129–138.
https://doi.org/10.1016/0040-6031(83)80309-8

26. Paschalidou, P. and Pashalidis, I., *J. Radioanal. Nucl. Chem.,* 2019, vol. 320, pp. 807–813.
https://doi.org/10.1007/s10967-019-06539-0

27. Farag, A.B., Bakry, A.R., Abdelfattah, N.A., and Elwy, A.M., *Int. J. Adv. Res.,* 2015, vol. 3, no. 5, pp. 32–41.

28. Mathew, K.J., Mason, B., Morales, M.E., and Narayann, U.I., *J. Radioanal. Nucl. Chem.,* 2009, vol. 282, pp. 939-944.

https://doi.org/10.1007/s10967-009-0186-4

29. Marczenko, Z. and Balcerzak, M., *Separation, Preconcentration, and Spectrophotometry in Inorganic Analysis,* Amsterdam (Netherlands): Elsevier, 2000.

30. Govindaraju, K., Mevelle, C., and Chouard, C., *Anal. Chem.,* 1976, vol. 48, pp. 1325–1331.