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A STUDY OF URANIUM AND P_2O_5 TRANSFER IN SYRIAN PHOSPHATE LEACHING BY COMMERCIAL NITRIC AND HYDROCHLORIC ACIDS

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ABSTRACT

This paper reports a study of leaching of uranium and P_2O_5 from Syrian phosphate using commercial nitric and hydrochloric acids. The effect of different parameters such as the liquid:solid ratio, the acid concentration, the leaching time and the reaction temperature is studied aiming optimization of various process parameters and hence maximization of uranium and P_2O_5 recovery. The optimal dissolution efficiency of uranium and P_2O_5 in hydrochloric acid referring to 88.46 % and 91.37 %, respectively, is obtained under the following conditions: a liquid:solid ratio of 5:1 mL:g, an acid concentration of 29.6 %, contact time of 25 min and a reaction temperature of 40°C. The optimal dissolution efficiency of uranium and P_2O_5 in nitric acid referring to 94.16 % and 93.07 %, respectively, is obtained under the following conditions: a liquid:solid ratio of 6:1 mL:g, an acid concentration of 40.9 %, contact time of 20 min and a reaction temperature of 50°C.

***Keywords:** Syrian phosphate dissolutions, phosphate acidulation, uranium recovery, phosphoric acid production.*

INTRODUCTION

The phosphate deposits, that are the prime supply of phosphate employed in the manufacture of phosphate fertilizers and orthophosphoric acid, contain low concentrations of uranium in the fine-grained mineral. They can be treated as an unconventional supply of uranium. This offers the phosphate trade a gorgeous chance to recover uranium as a fertilizers by-product and therefore has greatly revived the interest towards the wet-process of orthophosphoric acid production as a major supply of uranium [1].

Many researchers have attacked the phosphate rock by many materials. S. Matta uses a recycled hexafluosilicic acid to decrease the sulfuric acid quantities required for the phosphoric acid production in accord with the environmental advantages referring to the harmful fluorine gas recycling [2]. Yuksel Abali studies the dissolution kinetics of phosphate rocks with Cl_2

gas in aqueous media [3]. R. Marcato has designed a new process to obtain dicalcium phosphate. It is based on nitric acidulation of a phosphate rock concentrate, followed by neutralization of the extract liquor with calcium carbonate [4]. Other researchers use different acids such as hydrochloric, acetic, lactic, succinic and citric one [5 - 7].

Since 1950-es, a lot of research targets the recovery of uranium from a wet phosphoric acid (WPA) due to the potential of this resource to provide a major part of the world uranium production [8,9]. For instance, 11 000 tons of uranium have been produced on the ground of WPA in respect to the total 57 000 tons produced in the world in 2011 [10].

Most factories around the world use sulfuric acid for the treatment considered. Although the method is economical, it has many environmental hazards, especially in terms of the quantities of phosphogypsum produced

and its radioactive elements content.

In Syria, phosphoric acid has been produced since the early 1980s by the wet process using sulfuric acid. This has led to the production of tones of phosphogypsum. Its disposition outside the pit is currently prohibited aiming to avoid environmental pollution caused by the radionuclides remained [11].

The present study is designed to determine the effect of different parameters on uranium and P_2O_5 recovery during the dissolution of Syrian phosphate rock by commercial nitric and hydrochloric acids.

EXPERIMENTAL

Apparatus and materials

Uranium and P_2O_5 contents were determined in this research using UV-Vis device, model 402, manufactured by Optizen Pop Company. A shaker, model LSB-030S, manufactured by LabTech Company, was used with a maximum shaking speed of 190 rpm. All chemical materials, products of BDH Company, were of an analytical grade. The solutions used were prepared with distilled water.

Experimental methods

The phosphorus obtained by an acid extraction from the phosphate rocks used was determined by the ammonium molybdenum method. The corresponding complex obtained was measured at 420 nm [12]. The uranium obtained was determined on the ground of its reaction with theocyanate, while the complex obtained was measured at 360 nm [13].

Experimental procedure

Samples of a phosphate rock were taken from Khniefes mine in October 2017. The samples were dried to a constant weight in an electric furnace at 110°C for at least 4 h. Then they were pulverized and homogenized through sieving with an automatic vibratory screen to get particles of a size ranging between 0.25 mm - 0.01mm.

The uranium and P_2O_5 analysis showed that the phosphate rock samples contained 104 ppm of uranium and 30.15 % of P_2O_5 .

RESULTS AND DISCUSSION

Leaching Studies

The phosphate rock treatment process is based on leaching dry samples of 15 g using hydrochloric and

nitric acids. The effect of the liquid-solid ratio, the acid concentration, the contact time and the temperature on uranium and P_2O_5 leachability is followed.

A liquid-solid ratio effect

4 samples of a liquid-solid ratio ranging between 3:1 and 6:1 L:S mL/g are prepared to study the effect of the liquid-solid ratio on U and P_2O_5 leachability. Commercial nitric acid (49.09 %) and hydrochloric acid (29.6 %) are used. All samples are treated under identical conditions (contact time of 15 min, a reaction temperature of 40°C).

Fig. 1 exhibits the effect of the liquid-solid ratio (mL/g) on leachability (%) of uranium and P_2O_5 . The best results in case of a liquid-solid ratio of 6:1 mL:gr for nitric acid and 5:1 mL:g for hydrochloric acid refer to 92.3 % and 90.9 % for U and P_2O_5 , respectively, in nitric acid and to 84.6 % and 89.6 % for U and P_2O_5 , respectively, in hydrochloric acid.

As seen from the figure, the increase of the liquid:solid ratio leads to an increase of the leachability because of the increased contact surface and acid amount.

An acid concentration effect

Several samples of different concentrations of nitric and hydrochloric acid are prepared based on the previous results.

Figs. 2(a) and 2(b) illustrate the effect of the acid concentration on the leachability (%) of uranium and P_2O_5 . The best results refer to 92.3 % and 90.95 % for U and P_2O_5 , correspondingly, in case of nitric acid (49.09 %) and to 84.6 % and 89.82 % for U and P_2O_5 , correspondingly, in case of hydrochloric acid (29.6 %).

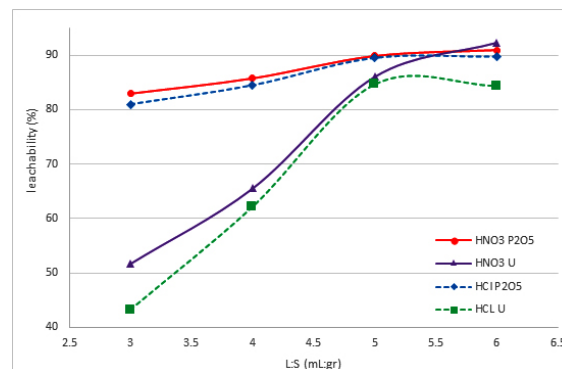


Fig. 1. A liquid-solid ratio effect on the leachability of U and P_2O_5 under the following conditions: 49.09 % HNO_3 , 29.6 % HCl , contact time of 15 min, a reaction temperature of 40°C.

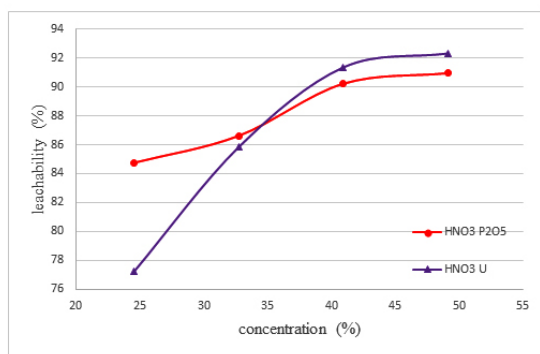


Fig. 2(a). A nitric acid concentration effect on the leachability of U and P_2O_5 under the following conditions: a liquid:solid ratio of 6:1, contact time of 15 min, a reaction temperature of 40°C.

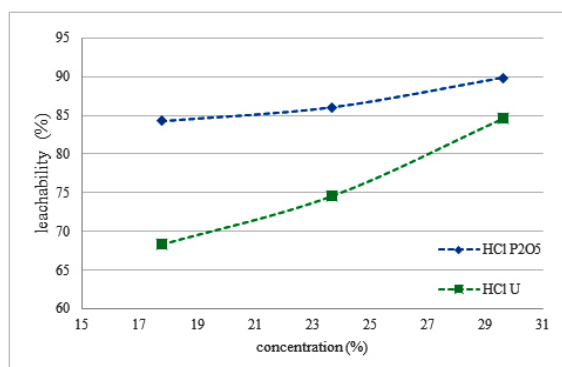


Fig. 2(b). A hydrochloric acid concentration effect on the leachability of U and P_2O_5 under the following conditions: a liquid:solid ratio of 5:1, contact time of 15 min, a reaction temperature of 40°C.

The graphs presented in Fig. 2 show that the leachability increases with the increase of the acid concentration.

A contact time effect

The effect of the contact time is studied in the range between 10 and 30 min. The phosphate rock samples are treated with HNO_3 under the following conditions: an acid concentration of 40.9 %, a liquid-solid ratio of 6:1 mL:g and a reaction temperature of 40°C. In case of hydrochloric acid the acid concentration is 29.6 %, the liquid-solid ratio is 5:1 mL:g, while the reaction temperature is 40°C.

The contact time effect on the leachability (%) of P_2O_5 and U is illustrated in Fig. 3. The best results refer to 93.3 % and 92.08 % for U and P_2O_5 , respectively, at contact time of 20 min in case of nitric acid and to 88.46 % and 91.37 % for U and P_2O_5 , respectively, at contact time of 25 min in case of hydrochloric acid.

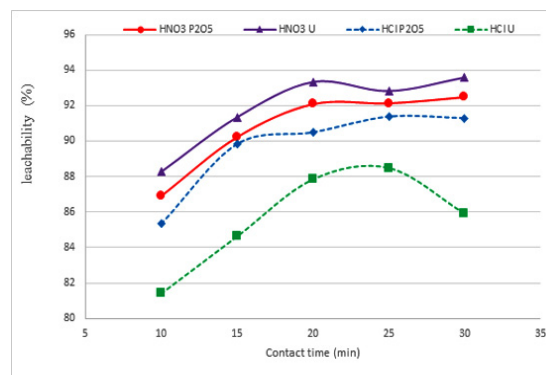


Fig. 3. A contact time effect on the leachability of U and P_2O_5 under the following conditions: a liquid:solid ratio of 6:1, an acid concentration of 40.9 %, a reaction temperature of 40°C in case of HNO_3 ; a liquid:solid ratio of 5:1, an acid concentration of 29.6 %, a reaction temperature of 40°C in case of HCl.

The graphical data shows the contact time effect on the leachability of U and P_2O_5 . As evident the increase of the contact time leads to an increase of the leachability within the initial 20 min. Then a balance is reached.

A reaction temperature effect

The temperature effect is studied in the range of 25°C - 60°C for phosphate rocks samples treated with (i) HNO_3 under the following conditions: an acid concentration of 40.9 %, a liquid-solid ratio of 6:1 mL:g and contact time of 20 min, and (ii) hydrochloric acid under the following conditions: an acid concentration of 29.6 %, a liquid-solid ratio of 5:1 mL:g and contact time of 5 min.

Fig. (4) demonstrates the temperature effect on the leachability (%) of P_2O_5 and U. The best results refer to 94.1 % and 93.07 % for U and P_2O_5 , correspondingly, at 50 °C in case of nitric acid, and to 88.46 % and 91.37 % for U and P_2O_5 , correspondingly, at 40°C in case of hydrochloric acid.

It is evident from the figure that a significant decrease of P_2O_5 leachability is brought about by temperature increase in HCl. This behavior is attributed to hydrochloric acid evaporation which is observed at higher temperature values [14].

CONCLUSIONS

A new process for Syrian phosphate treatment is developed using commercial nitric and hydrochloric acids. Factors affecting U and P_2O_5 transformation are studied aiming the process optimization. They refer to the liquid:solid ratio, the acid concentration, the leaching

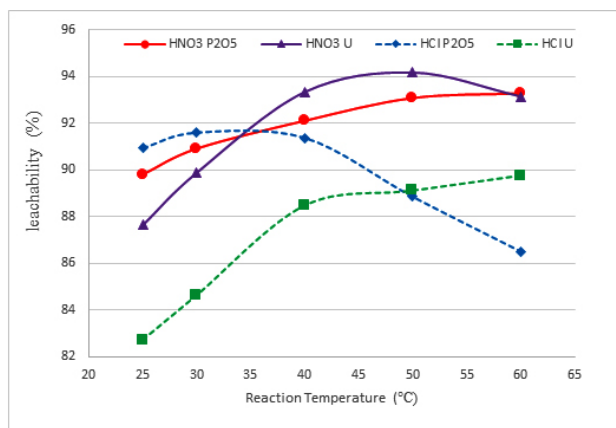


Fig. 4. A reaction temperature effect on the leachability of U and P_2O_5 under the following conditions: (i) HNO_3 : a liquid:solid ratio of 6:1, an acid concentration of 40.9 %, contact time of 20 min; (ii) HCl: a liquid:solid ratio of 5:1, an acid concentration of 29.6 %, contact time of 25 min.

time and the reaction temperature. The best results are attributed to 94.16 % and 93.07 % of U and P_2O_5 , respectively, in case of using nitric acid with a liquid:solid ratio of 6:1 mL:g, an acid concentration of 40.9 %, contact time of 20 min and a reaction temperature of 50°C. The corresponding results obtained in HCl refer to 88.4 % and 91.3 % for U and P_2O_5 , respectively, in case of a liquid:solid ratio of 5:1 mL:g, an acid concentration of 29.6 %, contact time of 25 min and a reaction temperature of 40°C.

The obtained optimal conditions of U and P_2O_5 maximal leaching are related only to the range of the parameters studied.

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