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Potentiality of uranium adsorption from wet phosphoric acid using amine-impregnated cellulose

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ABSTRACT

The prepared amine-impregnated cellulose (AIC) was found to be an efficient adsorbent for uranium originated from El-Sebaiya phosphate ore. The impregenation process was carried out by copolymerization between pretreated cellulose and a mixture of (tri-ethyl amine and epi-chorohydrine).

In the present work, two working solutions were used. The first solution was a synthesized phosphoric acid (35% P_2O_5 and 100 ppm U) used for optimizing the loading process of uranium. The second one was used as a case study for adsorption of uranium from a real phosphoric acid (WPPA) prepared from El-Sebaiya phosphate ore (P_2O_5 35%, 100 ppm U). The obtained equilibrium data were found to be satisfactory fitted with Langmuir isotherm. A maximum-metal uptake of 56.5 mgU/g AIC was observed at the obtained optimum conditions. Also, elution process of uranium has been achieved at 1 M of 30 ml Na_2CO_3 solution/g AIC after 15 min contact time. From the latter, a marketable product of sodium di-uranate was prepared.

1. Introduction

The energy crisis is actually considered as a worldwide problem due to increasing consumption of the fossil fuel resources. In the meantime, increasing uranium exhaustion of the classical uranium ores in the last century would recently re-orient research attention towards the non-conventional uranium resources. The latter include mainly the phosphate ores, shales and lignite in which the uranium content is usually quite low. Phosphorites actually considered as an important alternate or secondary source for uranium with an average uranium content of 50–200 ppm. According to the International Atomic Energy Agency (IAEA, 2012), uranium recovery from phosphoric acid could easily reach 10% of the world uranium production whereby this percentage is believed to increase in the future.

Egyptian production of phosphate rocks is about 6.0 million tons represents about 3.0% of the world total phosphate rock production (Jasinski, 2012). Phosphoric acid can be

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manufactured using two principal methods, the thermal process and the wet process (WPPA). The acid produced by thermal method is extremely pure; however, it is also expensive. The wet process which is based on the acidulation of phosphate ores using any mineral acid is the most popular. Phosphoric acid manufactured using the wet process has many environmental calamities, among these are the presence of heavy trace elements (Becker, 1989).

With respect to uranium recovery from phosphoric acid, the low selectivity and capacity of available resins for the extraction of uranium from P_2O_5 solutions and their low density compared with that of the acid have limited their use until now. The density and viscosity of phosphoric acid was such that they make resin beads float.

On the other hand, many efforts has been done for recovering uranium from phosphoric acid by using different organic solvents e.g. D_2EHPA with TOPO studied by Cordero, Jodra, Otero, and Josa (1977) and Hurst, Arnold, and Ryon (1977), the OPPA [octyl pyrophosphoric acid] by Ghosh, Bellary, and Keni (1994). Also, using a synergistic mixture; namely DOPPA – TOPO (Krea & Khalaf, 2000), di-nonyl phenyl phosphoric acid (DNPPA) and tri-n-butyl phosphate (TBP) (Singh, Mishra, & Vijayalakshmi, 2003), D_2EHPA-TOPO [di-(2-ethyl hexyl) phosphoric acid/trioctyl phosphine oxide] (Ali, Ali, Taha, & Ahmed, 2012), synergistic reagent in mixture with D_2EHPA (Beltrami, Chagnes, Haddad, et al., 2014; Beltrami, Chagnes, Mokhtari, et al., 2014), bis(1,3-dialkyloxypropan-2-yl) phosphoric acids and tri-n-octylphosphine oxide (Beltrami et al., 2013), bis(1,3-dibutoxypropan-2-yl) phosphoric acid (BiDiBOPP)/di-n-hexyl octylmetoxy phosphine oxide (di-n-HMOPO) (Beltrami, Chagnes, Haddad, et al., 2014; Beltrami, Chagnes, Mokhtari, et al., 2014), di-nonyl phenyl phosphoric acid (DNPPA) and bidentate *n*-octyl (phenyl)-*N*,*N*-di-isobutyl carbamoyl methyl phosphine oxide (CMPO) in *n*-dodecane (Mondal et al., 2014) and cetrimide (Orabi, El-Sheikh, Mowafy, Abdel-Khalekb, & El Kady, 2015).

Beltz et al. (1983) reported that both primary and secondary as well as tertiary amines can be used as alkylamines for recovering uranium from phosphoric acid. Some amines, however, may form a third phase which can be prevented if other organic substances with solubilizing properties such a n-octanol, iso-decanol, reaction mixtures from the oxysynthesis in the range from C3 to C12, cyclohexanone or tridecanol are added to the organic phase of amine polyphosphate and/or amine metaphosphate and organic solvent. However, the addition of a solubilizer represents a considerable complication as a result of the change of the distribution equilibriums between the phases.

The great advance in recent years drives towards bridging the gaps between ion exchange resin and solvent extraction systems through development of new effective media called solvent impregnated resin (SIR). The latter requires the impregnation of the required solvent upon a synthetic polymeric resin (e.g. Amberlite-XAD, silica gel, activated carbons and cellulose, … etc) devoid of any functional groups. The extraction behavior involves the counter diffusion of metal ions from the aqueous solution and the ions from the resin phase through various different resistances. The species in solution must diffuse across the liquid film surrounding the SIR particle, transfer across the particle interface, diffuse into the bulk of the impregnated resin and then interact with an impregnated extracting. The species initially within the impregnated particle experience the same resistances but in the opposite order during the elution step (Amir, 2014).

In this work, amine-impregnated cellulose (AIC) has been prepared as solvent impregnated resin to avoid many problems when using only amine as uranium extractant. The present work was first directed towards optimization of the relevant factors affecting both adsorption and elution of U on amine -impregnated cellulose (AIC) from a synthetic phosphoric acid solution WPPA (35% P_2O_5). The work has then been shifted to investigate the potentially of the prepared (AIC) for the recovery of U from a real wet phosphoric acid prepared from El-Sebaya phosphate ore.

2. Materials and methods

2.1. Materials

2.1.1. Preparation of synthetic phosphoric acid

The relevant factors affecting uranium adsorption and elution processes with the study (AIC) have first been optimized using stimulated synthetic uranium-bearing phosphoric acid solution. The latter (assays 35% P_2O_5) has thus been prepared from Adwic 85% P_2O_5 phosphoric acid by proper dilution. Also, a properly weighed sample of uranyl acetate was dissolved in the prepared synthetic phosphoric acid solution to attain an assay of 100 ppm uranium to be comparable to the wet process phosphoric acid (WPPA) produced from El-Sebaiya phosphate ore sample.

2.1.2. Preparation of El-Sebaiya WPPA (case study)

El-Sebaiya WPPA was used in the present work as a case study where the optimum factors obtained for uranium extraction and elution from the synthetic acid were applied.

According to El-Sayed & Abdel-Aal (2000), El-Sebaiya WPPA was prepared using HCl (300 g/L) with particle size of −0.25 mm at room temperature (25 °C) within S/L ratio of 2:1.

2.2. Preparation of synthesized amine-impregnated cellulose (AIC)

2.2.1. Pretreatment of cellulose

Prior to impregnation process, 20 g of whatman powder cellulose was firstly, refluxed with 0.5% sulfuric acid using solid liquid ratio (S/L) of 1/6 at 55 °C for 4 h. The produced cellulose air-dried for two days. The purpose was to remove the amorphous part of cellulose to resist the high acidity when using phosphoric acid.

2.2.2. Impregnation process

The synthetic amine-impregnated cellulose (AIC) was prepared by co-polymerization between one mole of pretreated cellulose and 5 mol of tri-ethyl amine and epichorohydrine (1:1.2 mol ratio) refluxed for 4 h at 55 °C until granulation. The latter was used as solvent impregnated resin (AIC).

2.3. Uranium recovery procedures

In order to study the relevant factors affecting the recovery process of uranium from phosphoric acid, many series of