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Modeling Uranium Adsorption Using Hydroxyapatite Derived From Natural Phosphate Via Wet Precipitation Method

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ABSTRACT

The main objective of this study is the recovery of uranium VI present in radioactive effluents through adsorption, using synthesis phosphate (SP) as an adsorbent. In the first phase, we developed (SP) through wet precipitation method. The (SP) was characterized using X-ray diffraction, scanning electron microscopy, infrared spectroscopy. The second part of this work focused on the batch adsorption study of uranium VI by SP. We employed a numerical approach to analyze and optimize the parameters of the uranium VI adsorption process by SP, such as pH, adsorbent dose, and initial uranium VI concentration. The conclusions of this study indicated that the optimization was efficient to predict the uranium recovery by (SP) and the exploitation of abundant natural resources as sources of calcium and phosphorus has yielded significant results.

Keywords: U (VI), Adsorption, Natural Phosphate, Hydroxyapatite.

1. Introduction

Environmental contamination by radionuclides, such as uranium, is a pressing issue resulting from industrial and nuclear activities. Unlike other contaminants, radionuclides are non-biodegradable and pose serious risks to human, animal, and plant health. The nuclear industry's growth has led to increased discharge of radioactive wastewater, threatening water resources and public health. Traditional methods for removing radionuclides, like chemical precipitation and ion exchange, have significant drawbacks, including high costs, toxic waste production, and energy consumption [1]. Adsorption offers a promising solution due to its high capacity, ease of regeneration, and rapid kinetics. Calcium phosphates, especially apatites, are effective adsorbents for removing heavy metals and radionuclides from wastewater [2]. This study investigates the adsorption of U(VI) ions by apatites synthesized via a wet method from natural phosphate resources as precursor of calcium and phosphorus.

2. Experimental

2.1. Synthesis of Hydroxyapatite

Hydroxyapatite was synthesized by dissolving 30 g of natural phosphate, extracted from Kefsnoun ore, in 2 L reactor containing 500 ml of distilled water. This dissolution process was initiated by adding 20 mL of 65% nitric acid solution, with continuous agitation maintained using a magnetic stirrer for 3 hours at 25°C at pH 2. The resultant mixture underwent vacuum filtration. Subsequently, the precipitation was done by adding 200 ml of 25% NH₄OH in continuous magnetic stirrer, and the pH was then adjusted to 10. The resulting precipitate underwent maturation under magnetic agitation for 24 hours at room temperature. After maturation, the precipitate was filtered, washed with distilled water, and dried at 100°C for 24 hours in an oven.

2.2. Uranium adsorption procedure

Adsorption experiments were conducted in batch under mechanical agitation using 50mL beakers/Erlenmeyer flasks containing U(VI) solution with concentrations ranging from 100 to 310 mg/L,



masses of SP (0.01 to 0.05 g), and initial pH solution ranging from 1 to 5. The mixing speed was set at 200 rpm at 25° C for all experiments. After agitating for fixed time 200 min, the mixture (adsorbate and adsorbent) was filtered using Whatman No. 04 filter paper.

2.3. Design of Experiments for the U(VI) Adsorption by SP

The Box-Behnken matrix design was employed for the three variables (initial concentration of U(VI), initial pH of the solution, and adsorbent dosage) with the aim of optimizing the selected parameters in this study in a total of seventeen sequential experiments. The second-order polynomial equation was used to correlate the adsorption of U(VI) and the independent variables.

2.4. Characterization of SP

Characterizations by X-ray diffraction were performed on powder samples at room temperature using a Philips X'Pert MPD system diffractometer. The diffractograms were obtained within the 2θ region (20°–60°) with a scanning step of 1.2. The **Scanning Electron Microscopy of the SP** was conducted using (Philips XL30; Japan) at an alternating voltage ranging from 20 to 30 kV resolution, coupled with an Energy Dispersive Spectrometer (EDS). For the detection of chemical bonds present in the SP, the Fourier Transform Infrared Spectroscopy was carried out using a JASCO Spectrophotometer (USA) in the region of 400–4000 cm⁻¹.

2.5. Experimental method for U(VI) Analysis

For U(VI) quantification, UV-Visible spectrophotometry was employed. The concentration of U(VI) in all adsorption experiments was determined using the arsenazo III complexation method, which forms a pink-colored complex with U(VI) at 650 nm.

3. Results and Discussion

3.1. Phase identification of (SP) by X-ray Diffraction

X-ray diffraction patterns of the prepared product are presented in (Fig. 1). A multiple diffraction peaks corresponding to the hkl planes: (200), (111), (002), (102), (210), (211), (300), (202), (301), (212), (310), and (213) of the hydroxyapatite structure, as identified according to the JCPDS 09-432 PDF file. Other minor phases were also detected, such as Ca₂FH₃O₅Si at 2θ (26.69° and 44.83°), calcium oxide CaO at 2θ (37.5° and 54°), and calcium carbonates Ca₆C₆O₁₈ at 2θ (29.26° and 47.5°).

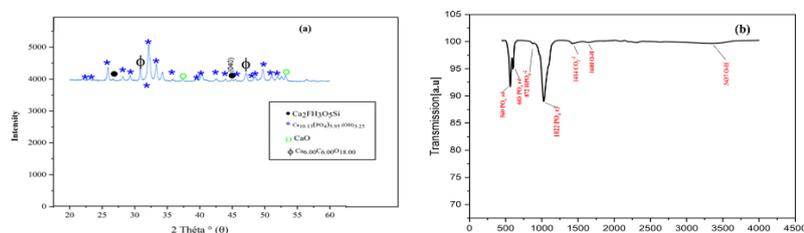


Figure 1: Characterization of SP sample: (a) DRX. (b)FTIR

3.2. Characterization of SP by SEM

The results from (Fig. 2) of the SEM characterization of synthesized phosphate reveal fine grains with irregular shapes ranging in size from 10 to 100 μm. Semi-qualitative analyses obtained by EDS reveal the major elements present in these structures, namely, Ca (13.28% by mass), and P (9.76% by mass), F (7.61% by mass) and Si. These results are in good agreement with those from X-ray Diffraction.

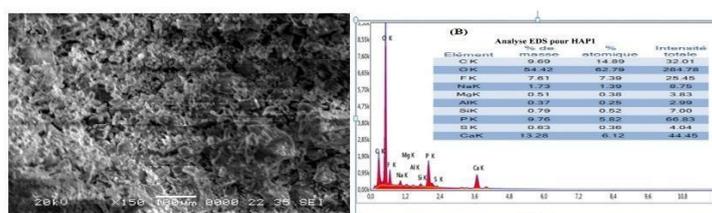


Figure 2: Characterization by SEM: (a) morphology analysis of SP, (b) :EDS analysis.

3.3. Modeling uranium adsorption by RSM

The results of the modeling of uranium adsorption by are summarized in fig. XX:

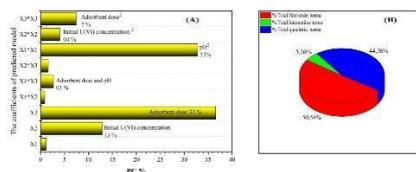


Figure 3: percentage contribution terms of predicted model: (a) contribution terms of the model, (b): different way contribution on uranium adsorption.

The interaction of pH and adsorbent dosage are the two dominant parameters contributing to the adsorption of U(VI) with the highest PC values in a quadratic effects which is the most dominant term on uranium adsorption.

4. Conclusions

(I) Characterization by (XRD), (FTIR), and (SEM) revealed the presence of apatite phases characteristic of hydroxyapatite. (II) Design of Experiments (DOE) planning demonstrated that pH and adsorbent dosage are the two most influential parameters on the adsorption of U(VI) by SP. (III) Utilizing of natural sources as calcium and phosphate precursors for apatite synthesis resulted in low-cost materials with appreciable U(VI) adsorption capacity.

References

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