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Adsorptive removal of uranium ions from liquid waste solutions by phosphorylated chitosan

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HIGHLIGHTS

- In this study, Phosphorylated chitosan was synthesized.
- Phosphorylated chitosan used to investigate the adsorption of U(VI) ions.
- The P-chitosan exhibits good adsorption characteristics for uranium (VI).
- The adsorption of uranium on P-chitosan was not a pH dependent.
- Langmuir model describe the adsorption, adsorption capacity was 54.95 mg/g.

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ABSTRACT

The purpose of this work is to take a closer look for the phosphorylation of chitosan and its potential applications for uranium removal from waste effluents. The synthesized P-chitosan has first been characterized using FTIR spectroscopy and scanning electron microscope before and after adsorption of uranium. From the former it is showed that the phosphate groups may be more responsible for the adsorption of uranium. The relevant factors that influence the uranium adsorption onto the phosphorylated chitosan namely solution pH, contact time, temperature, Co-ions, solid–liquid ratio and initial uranium concentration were studied in detail. The obtained results are fitted into the different adsorption isotherms and according to Langmuir adsorption model the uranium adsorption capacity of the synthesized P-chitosan is estimated as 54.6 mg/g. It was also observed from the obtained kinetic data that the pseudo-second order model is more suitable to explain the adsorption kinetics of uranium up on the P-chitosan. Finally the thermodynamic parameters show the adsorption reaction to be a spontaneous process and more favorable at room temperature.

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1. Introduction

Chitosan is a natural-based non-toxic, biodegradable and biocompatible polymer and has been used in bio-medical areas as well as in various industrial applications. However, practical use of this polymer has been mainly confined to the unmodified forms. But recently, there has been a growing interest in chemical modification of chitosan to improve and widen its application. In addition, phosphorylated chitosan has attracted considerable interest because of its various advantages: anti-inflammatory property and the ability to form metal complexes.

The high sorption capacities of modified chitosan for metal ions can be of great use for the recovery of valuable metals or the treatment of contaminated effluents. A great number of chitosan derivatives have been obtained with the aim of

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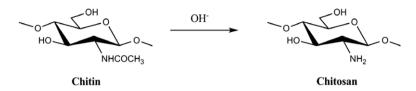


Fig. 1. Chemical structure of chitosan.

adsorbing metal ions (Park et al., 2006; Shi et al., 2006), treating water (Prabaharan and Mano, 2006; Crini, 2006), biomedical (Ng and Swami, 2005; Crompton et al., 2005), cosmetic (Crompton et al., 2005; Tsigos et al., 2000), agricultural (Baños et al., 2006) or food industrial (Muzzarelli, 1996; Ravi Kumar, 2000), by including new functional groups onto the chitosan back-bone. In other words the chemical modification of chitosan to generate new bio-functional materials is of primary interest because such procedure would not change the fundamental skeleton of chitosan and would keep the original physio-chemical and biochemical properties depending on the nature of the group introduced (Prabaharan et al., 2006; Tsigos et al., 2000; Kweon et al., 2003).

The new functional groups are incorporated into chitosan to increase the density of sorption sites, to change the pH range for metal sorption and to change the sorption sites in order to increase sorption selectivity for the target metal. Introduction of groups such as phosphonic acid or phosphonate onto chitosan by reaction of a phosphorylating agent onto the amino groups are known to increase the chelating properties of chitosan (Hendrickson, 1967; Schwarzenbach et al., 1949a). Several techniques to obtain phosphate derivatives of chitosan have been proposed due to the interesting chemical properties of such compounds (Sakaguchi et al., 1981; Ramos et al., 2002).

In fact, Schwarzenbach et al. (1949b) considered that the phosphonic complexing agents are as effective or even more than those containing carboxylic groups. The amino alkyl phosphonic ligands $(NH_2-CH_2-PO_3^{2-})$ have well known strong chelating properties owing to the donor effect of the amine group $(-NH_2)$ and the monodentate ligand as $-PO_3^{2-}$ (Hendrickson, 1967). They have tendency to form chelates in ring structure with possibilities of different conformations owing to the metal ion nature. In the case of NMPC $(NH_2-CH_2-PO_3^{2-})$ the presence of an amine group from chitosan combines these two effects, increasing the metal binding abilities and its ability to chelate not only transition-metal ions, but also calcium ions (Westerback et al., 1965). From the latter the NMPC has a powerful chelating agent of transition metal and calcium ions. It is reported that P-chitosan adsorb uranium much more than the other heavy metal ions (Ramos et al., 2003).

The present investigation was undertaken to obtain fundamental information on the removal of uranium from waste effluents using Phosphorylated chitosan. Factors that influence the uranium adsorption onto phosphorylated chitosan such as solution pH, contact time, temperature, Co-ions, solid–liquid ratio and initial uranium concentration as well as a case study have been studied in detail.

2. Experimental

2.1. Materials

All the chemicals used (di-methylformamide, urea, ortho-phosphoric acid, HCl, NaOH) are analytical grade, purchased from Merck and/or Sigma-Aldrich, while commercial chitin (Otto Kemi, C1904) was obtained in the form of flakes from M/s BR Corporation, Mumbai. The raw sample was crushed and sieved into less than 0.2 mm using scientific sieves.

2.2. Chitosan preparation

According to the method of Muzzarelli and Rochetti (1985), chitosan was prepared by chitin deacetylation, where a known weight from chitin was stirred in 50% NaOH solution for 2 h at 100 °C. The mixture was then placed under the hood and cooled for 30 min at room temperature. Afterwards, the product was continuously washed by 50% NaOH and filtered in order to retain the solid matter, which is the chitosan (Fig. 1). The final product was oven-dried at 110 °C for 6 h.

2.3. Phosphorylated chitosan synthesis

P-chitosan was prepared by the method described by Sakairi et al. (1998). In brief, 10 g of chitosan powder was mixed with 50 g urea and 100 ml of N,N-dimethylformamide followed by the addition of 11.8 g orthophosphoric acid(85%). The mixture was then allowed to react at 150 °C for 1 h. After cooling, the P-chitosan product was filtered, washed with distilled water until nearly neutral pH, dried at room temperature.

2.4. Preparation of stock solutions

A uranium stock standard solution assaying 1000 ppm (mg/L) was prepared by dissolving 1.782 g of uranyl acetate $[UO_2(CH_3COO)_2-2H_2O]$ of BDH Chemicals Ltd. Poole, England. The standard solutions of Cu, Ni and Fe were prepared from (Merk) stock solutions containing 1000 mg/L, of each studied element. The working standard solutions were then prepared from their stock solutions by dilution with double distilled water to give the desired concentration.

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