

Removal of Uranium Ions in Lagoon Waste by Electrosorption

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Abstract

A study on the electrosorption of U(VI) onto porous activated carbon fibers (ACFs) was performed to treat uranium-containing lagoon sludge. Effective U(VI) removal is accomplished when a negative potential is applied to the activated carbon fiber(ACF) electrode. For a feed concentration of 100mg/L, the concentration of U(VI) in the cell effluent is reduced to less than 1mg/L. The adsorbed uranium could be desorbed from the ACF by passing a 1M NaCl solution through the cell and applying a positive potential onto the electrode. The regeneration of ACF from the cycling experiments was confirmed.

Introduction

For the removal and recovery of uranium(VI) from contaminated water and waste streams, a variety of physical and chemical methods such as precipitation, coagulation, ion exchange and adsorption have been used. But, these techniques have been restricted in application due to their limited capacity when the concentration of U(VI) in the waste water is relatively high [1-3]. As an efficient electrochemical method for the removal of a high concentration of uranium, electro-deposition on carbon materials has been extensively investigated and used very effectively. For uranium having a high reduction potential, electrodeposition is not a practical method [4]. An alternative to electrodeposition is electrosorption that is defined as the reversible adsorption or the reversible retention of ions, molecules, or particles from a liquid phase on or near an electronic conducting surface as a function of the electric potential difference between the surface and the liquid [5]. The amount of material adsorbed in electrosorption depends on the electrochemical potential applied to the adsorbent. This technique has reversible characteristics for purifying a waste solution by adsorption and concentrating contaminants by desorption.

In this study, we conducted experiments on the selective adsorption of uranium(VI) from a high concentration of chemical salt to find the optimal conditions of the electrosorption process using ACF as an electrosorption adsorbent. The adsorption behavior with an applied potential was investigated and compared with that in the potential free conditions. Desorption was examined by first adsorbing at a negative potential, then reversing the electric potential back

to positive. In order to confirm the regeneration of ACF, cycling experiments consisting of the adsorption and desorption steps were carried out.

Experimental

Flow-through adsorption experiments were carried out using a three-electrode electrochemical cell where the electric current flows parallel to the solution flow, as shown in Fig. 1. Pitch-based ACF was used as the working electrode and placed on a platinum mesh (current collector). The counter electrode was platinum wire and the Ag/AgCl electrode was used as the reference electrode. All the potentials reported in this paper are relative to this reference electrode. The electro-chemical cell was connected with a potentiostat (EG&G Model 273). The fixed flow rates through the cell were controlled by a peristaltic pump. Various types of salts such as NH_4NO_3 , NaNO_3 and $\text{Ca}(\text{NO}_3)_2$ were tested as supporting electrolytes. The ACF felt electrode was kept under various potentials ranging from 0.3 to 0.9V in order to remove the uranium ions in a high concentration of electrolyte salt by electrosorption. A desorption test was performed at potentials ranging from +0.5 to +1.2V and an influent pH in the range of 3 to 5.

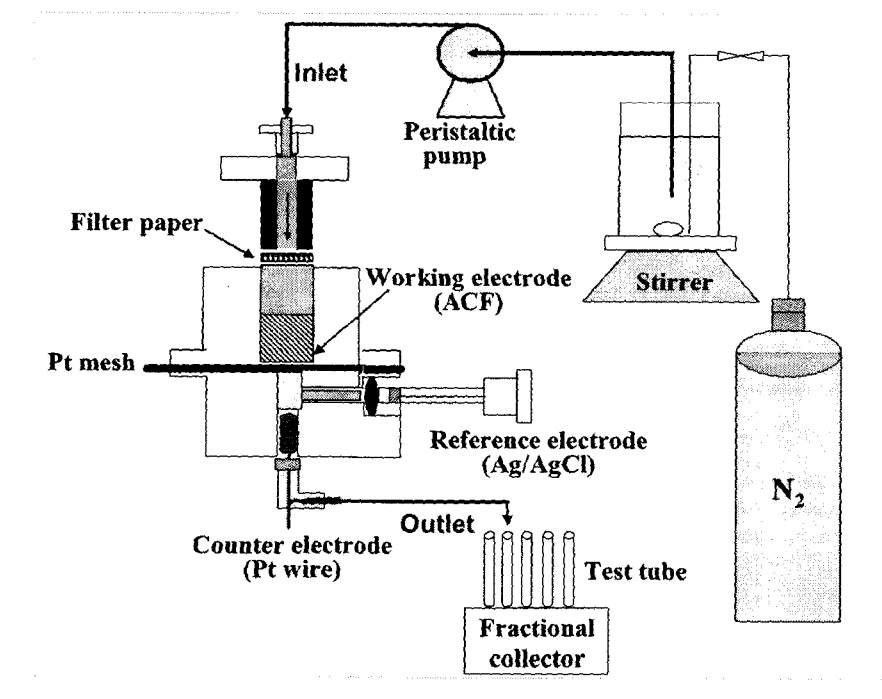


Fig. 1. Schematic diagram of the electrosorption cell.

In the cycling experiments to test the regeneration of ACF, desorption was examined by first adsorbing at -0.9V for 30h, then reversing the electric potential back to $+1.2\text{V}$ for 30h.

Results and Discussion

ACF felt used as an electrode has a large BET specific surface area(1536 m²/g), and is microporous adsorbent with an average pore diameter of about 15 Å. It has a surface acidity of 2.45 with a high proton liability of the surface OH and COOH groups. The electrosorption tests on the U ion was performed in the electric potential ranges from -0.3 to -0.9 V(vs Ag/AgCl). Fig. 2 shows the selective removal of the U ions in the multi-component system composed of the Na⁺, NH₄⁺ cations. Adsorption of the cations is related to the charge density or ionic potential of the cations. The higher the charge density(i.e. z/r, where z is the valency and r is the radius of the adsorbed ion), the stronger the cation-carbon affinity, with a variation of the applied potentials. The selective adsorption of the U ions existing as UO₂²⁺ is due to their higher charge density than that of the Na⁺, NH₄⁺ cations. Experimental test on the electrosorption with a 100 mg/L U(VI) feed onto the ACF felt at various negative potentials in the range of 0.3 to 0.9 V (vs. Ag/AgCl) is shown in Fig. 3.

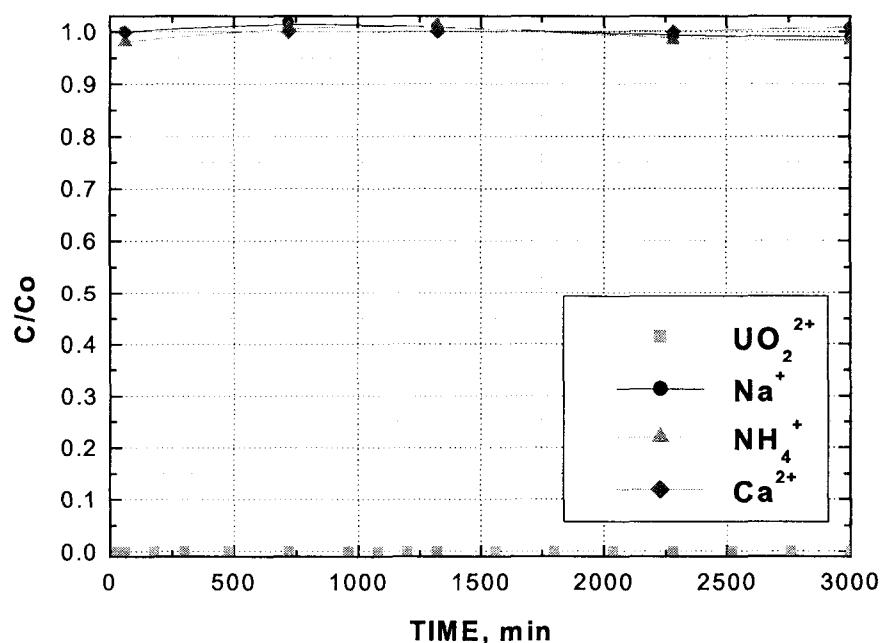


Fig. 2. Electrosorption of U(VI) in multi-component salt medium at -0.9V.

Effective U(VI) removal is accomplished at a potential of -0.9V. At a potential of 0.3V, the U(VI) concentration in the effluent is reduced to 1mg/L within 4h, then breakthrough started to appear. At 0.5V, the U(VI) ion in the effluent was detected after the test run of 10h. At a potential of 0.9V, a complete removal of U(VI) is maintained throughout the test. However, in the case of OCP(open-circuit potential) the effluent concentration of U(VI) increased within 1h and finally reached the level of the feed, indicating the saturation of the

sorption capacity of the ACF. From these results, it can be confirmed that the external negative potential exerted on the ACF electrode has a great impact on the adsorption capacity of the ACF.

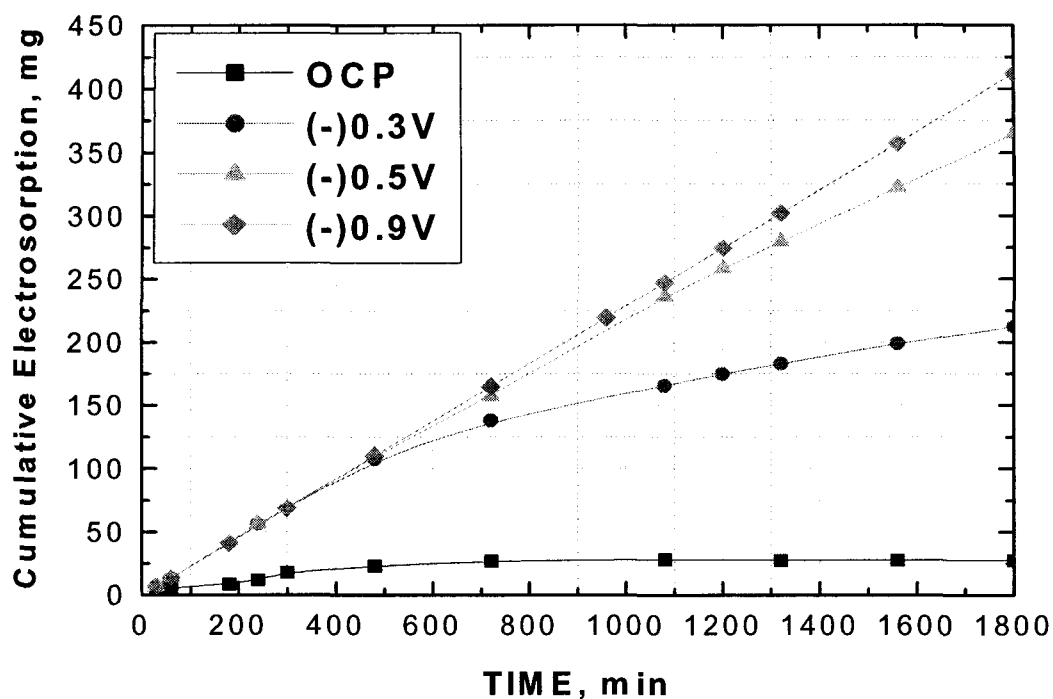


Fig. 3. Electro sorption of U(VI) with variation of potential in multi-component salt medium .

To confirm the continuous use of an electro-adsorbent, cycling experiments consisting of an adsorption step at -0.9V and desorption step at $+1.2\text{V}$, were performed. An adsorption/desorption cycle is shown in Fig. 4. The adsorption capacity of the ACF electrode measured for a multiple recycling up to 5cycles does not significantly change. These results indicate that reversing the potential of the ACF electrode results in the regeneration of the ACF carbon while electro-desorbing the adsorbate ions.

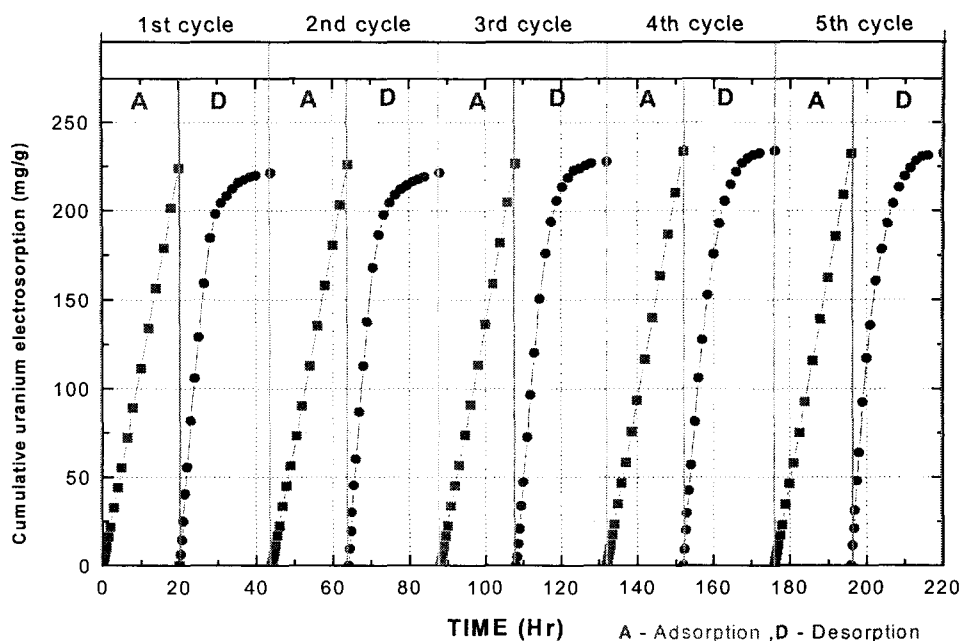


Fig. 4. Electroadsorption of U(VI) and regeneration of ACF electrode.

Conclusions

The electroadsorption process using an ACF felt electrode was confirmed to be effective in the removal of uranium ions, and an ACF felt electrode could be continuously used in the purification of a liquid waste. Also, the selective adsorption of U ions in Na^+ and NH_4^+ cations could be obtained. This electroadsorption process as a new technology without producing secondary wastes, appears to be a promising technique in liquid waste treatment and the separation of radioactive ions.

Acknowledgements

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