

THE SELECTIVE REMOVAL OF URANIUM IONS FROM SLUDGE WASTE BY ELECTROSORPTION

ChongHun Jung¹, WonZin Oh¹, YuRi Lee², SooJin Park³, SeungKonRyu²

¹*Decontamination & Decommissioning Technology R&D Division,
Korea Atomic Energy Research Institute*

²*Dept. of Chemical. Eng., Chungnam Univ.*

³*Korea Research Institute of Chemical Technology*

Corresponding author e-mail address: nchjung@kaeri.re.kr

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, electrodeposition is not a practical method [4] for the uranium having a high reduction potential.

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 the contaminants by desorption.

In this study, we conducted the experiments on a selective adsorption of uranium(VI) from a high concentration of chemical salts, to investigate the uranium removal mechanisms and the application conditions of the electrosorption technique using ACF as a good conductive electrosorption adsorbent.

Experimental

Flow-through adsorption experiments were carried out using a three-electrode electrochemical cell where the electric current flows parallel to the solution flow. 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.

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 were performed in the electric potential ranges from -0.3 to -0.9 V(vs Ag/AgCl).

Fig. 1 shows the selective removal of the U ions in the multi-component system composed of the Na⁺, Ca²⁺, NH₄⁺ cations at a potential of -0.9V. No break-through of uranium is observed for 50h, and it corresponds to an electrosorption capacity of 690mg U/g ACF. 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⁺, Ca²⁺ and NH₄⁺ cations.

Experimental tests 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 0.9V and a conventional adsorption(with open-circuit potential) is shown in Fig. 2. 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 starts 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. When the potential is -0.5V or more negative, the cumulative sorption plots are linear, indicating a constant sorption rate. From these results, it is clear that the external negative potential exerted on the ACF electrode has a great impact on the adsorption capacity of the ACF and controls the uranium sorption rate.

The adsorption capacity of the carbon depends on the double layer capacitance, specific surface area, and the applied potential. One gram of ACF used for this study (specific surface area of 1536 m²/g, double layer capacitance of 30F/g) may adsorb 74mg U/g ACF-V. This value is only 1/10 compared to the observed capacity. Therefore it is expected that other reactions may occur during electrosorption. A sample of uranium adsorbed was examined by X-ray diffraction and SEM, and they are shown in Fig. 3. From this test, uranium precipitation on the ACF has been observed, and it is identified as metaschoepite in the form of UO₃·2H₂O.

Conclusions

The electrosorption process using an ACF felt electrode was confirmed to be effective in the removal of uranium ions to treat uranium-containing sludge. The selective adsorption of U ions in the Na⁺, Ca²⁺, and NH₄⁺ cations could be obtained when a potential of -0.9V is applied to the activated carbon fiber(ACF) electrode. Electrosorption behaviors for U(VI) is considered to be the result of a double layer charging, and an electrochemical precipitation of the uranyl ions. Adsorbed uranium on the ACF is confirmed to be the precipitate of UO₃·2H₂O by XRD analysis.

References

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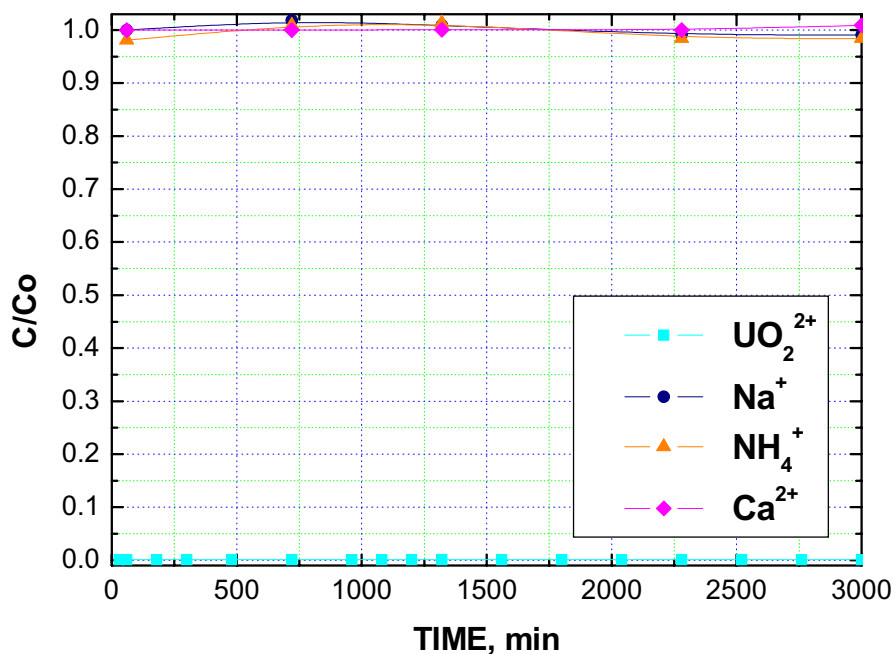


Figure 1. Electro-sorption of U(IV) in multi-component salts medium at -0.9V.

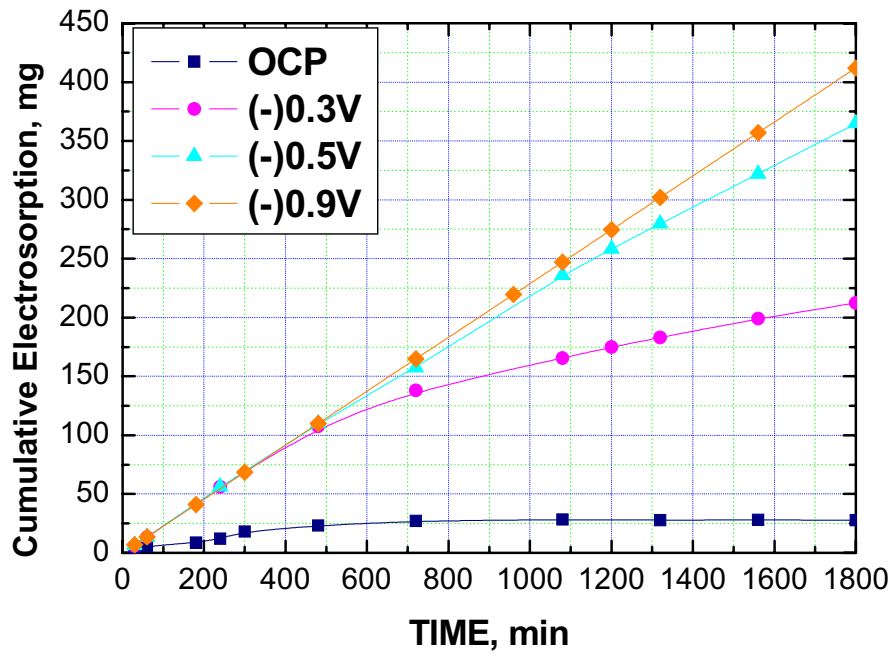


Figure 2. Electrosorption of U(IV) with variation of potential in multi-component salts.

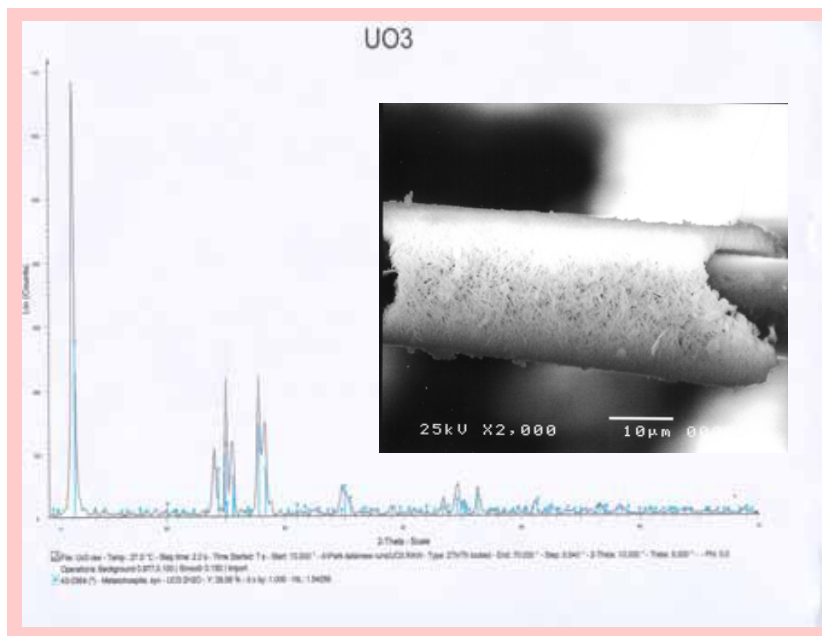


Figure 3. XRD and SEM analysis for adsorbed uranium on the ACFs.