The treatment of U-containing wastewater by electro-deposition

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Abstract. A new method for treating U-containing wastewater and recovering of uranium simultaneously by electro-deposition was proposed. The complexity of adsorbent preparation and difficulty in uranium recovery that existed in traditional adsorption method are solved by electro-deposition approach. The iron and graphite are used as anode electrode and cathode electrode in this experiment, respectively. The innovative view in this study is to promote the rapid generation of magnetite stemmed from iron dissolution under direct current. The uranium is incorporated or adsorbed to the magnetite through accurate controlling experimental conditions. The effects of initial U-concentration, voltage, electrode spacing, ion concentration, pH value on uranium removal efficiencies were investigated. Uranium removal efficiencies reached 83.5% under laboratory conditions that uranium concentration at 10mg/L, voltage at 30V, electrode spacing at 5 cm, ion concentration at 3 g/L, at pH 3.45. It is the advantages of U-containing magnetic is easily separated from aqueous solution and can be utilized as secondary low grade uranium deposit after treatment that making this method has a bright prospect.

Keywords: Uranium, Electro-deposition, Magnetite, U-containing wastewater.

1. Introduction

With the development of nuclear energy power, uranium is used extensively in all fields of the nuclear industry. Uranium containing wastewater is produced in the process of mining, purification, enrichment of mining and smelting [1-2]. As a result of long radioactive half-life, uranium containing wastewater can pollute the water source and cause radiation damage to the human body. Therefore, treating uranium-containing wastewater not only reduces pollution of water resources but also prevents people from being exposed to uranium radiation [3-4].

Recently, various methods have been applied for removal of uranium from U-containing wastewater, such as adsorption, chemical precipitation, ion-exchange, membrane separation and bioremediation [5]. Among these techniques, adsorption is considered an effective method with the advantages of high adsorption efficiency and adsorbent diversification. Commonly used adsorbent material such as activated carbon, chitosan, bentonite and zeolite were extensively used to prepare adsorbents for treatment of uranium containing wastewater[6-8].Wang uses synthetic mesoporous silica to purify U(VI) solution with initial concentration of 100 mg/L, and the saturated adsorption capacity can reach 203 mg/g, the efficiency was 95.25%[9]. Akhtarused calcium alginate immobilized trichoderma harzianumas adsorbent in wastewater purification and recycling of uranium, the results showed that calcium alginate immobilized trichoderma harzianumcould enhance adsorption capacity of uranium [10]. However the separation of adsorbent from aqueous solution is the difficulty of wastewater purification.

Therefore magnetic is utilized to purify U-containing wastewater because of being separated from aqueous solution easily. Furthermore magnetic is generated by electro-deposition approach that avoiding the preparation of absorbent. Electro-deposition used iron as anode electrode and graphite as cathode electrode, in which iron ions are generated by anodic iron and Fe^{2+} and Fe^{3+} converted to $Fe(OH)_2$ and $Fe(OH)_3$ colloids immediately[11]. Under the influence of electric field, Fe $(OH)_2$ and $Fe(OH)_3$ will generate magnetite further to immobilize uranium into its structure cell. The magnetic properties of can be easily separated from the uranium containing wastewater.

2. Materials and Methods

2.1 Materials

Stock solution of U (VI) (500 mg/L) were prepared by dissolving U5O8 (purity 99.9%) and then dilute to the desired concentration. Iron and graphite sheet were used as electrode materials.

2.2 Electro-Deposition Method

The main factors affecting of this experiment are the initial concentration of uranium solution, voltage, electrode spacing, ion concentration, pH value. The specific experimental process is as follows: 500ml of predetermined uranium solution was poured into the electrolytic cell. This experiment adopts the DC power supply, and recorded the pH value before energizing. After the power is turned off, the solution was filtered, the filter residue was dried and weighed, and the filtrate was analyzed and the removal rate of uranium was calculated.

2.3 Uranium Content Analysis Method

The filtrate of uranium content was analysis by ammonium vanadate titration method, the amount of ammonium vanadate was recorded, and the removal rate of uranium was calculated by formula (1).

$$W = \frac{C_0 - C_i}{C_0} \times 100\% \tag{1}$$

3. Results and Discussion

3.1 Effect of Initial Uranium Concentration

The initial uranium concentration range from 1 to 10 mg/L on U removal efficiency was investigated at the 32V voltage, 10 cm electrode spacing, 1.5 g NaCl dosage and electrolysis time is 2 h. The relationship between the initial uranium concentration and the removal rate of uranium is shown in theFig.1a.

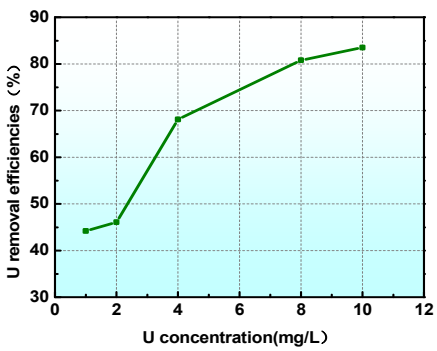

Figure.1a.Effect of Uranium Concentration on U Removal Efficiencies

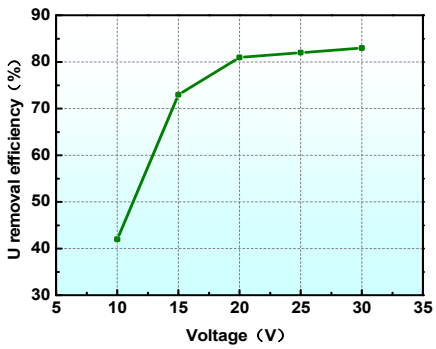

Figure.2.Effect of Voltage on U Removal Efficiencies

The results in the Fig.1.indicated that U removal efficiency raised with the increasing of U concentration and reach a highest value of 83.5% at 10 mg/L U. An increase of U concentration is beneficial of U removal. U removal efficiency in creased slowly during the range from 1 to 2 mg .Uranium recovery increased significantly when the initial uranium concentration was more than 4 mg /L.

3.2 Effect of Voltage

The voltage was varied from 10 to 30 V at the uranium solution is 10 mg/L, electrode spacing is 10 cm, NaCl dosage is 1.5 g and electrolysis time is 2 h. The result is shown in Fig.2.

This is a clear demonstration of Fig.2. that the rising of voltage increased the U removal efficiency. U remoa1val efficiencies increasing rapidly at voltage varied from 10 to 20 V. The U removal efficiencies tend to be gentle when the voltage is from 20 to 30V. U removal efficiencies reach the highest value 83% in 30V voltage.

3.3 Effect of Electrode Gap

The electrode spacing was controlled in the rang of 2.5-10 cm in the condition of 10 mg/L initial uranium solution, 30V, 10 cm electrode spacing and 2 h reaction time. The result is shown in Fig.3.

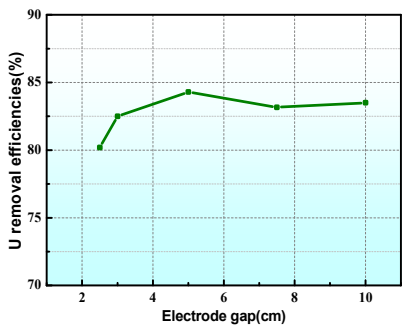

Figure.3.Effect of Electrode Gap on U Removal Efficiencies

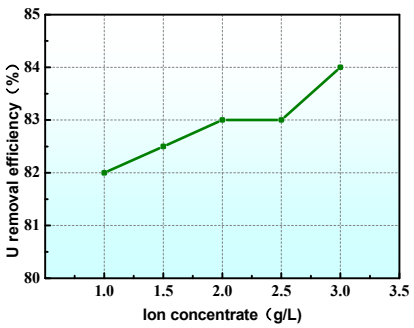

Figure 4.Effect of Ion Concentrate on U Removal Efficiencies

When the electrode spacing is from 2.5 cm to 5 cm, U removal efficiency increases with the raise of electrode spacing. When the electrode spacing is 5 cm, the removal rate of uranium is 84.3%. Increasing the distance between the electrode and the graphite can reduce the possibility of the Fe_2O_3 flocculation plugging between the two poles, helping the free ions to diffuse in the solution and increasing the uranium removal efficiency. However, when the distance between the electrodes is too large, the anode is easy to passivate, so the best electrode plate spacing is 5 cm.

3.4 Effect of Concentration

This experiment changes the ion concentration by adding NaCl. The effect of varying NaCl dosage from 1 to 3 g/L at uranium solution of 10 g/L, 30 V voltage, and 10 cm electrode gap. The result is given in Fig.4.

It can be seen from the graph that the dosage of NaCl has slightly affected. When the solid concentration of NaCl is between 1.0 and 2.0 g/L, the removal rate of uranium varies greatly. When the concentration of NaCl is between 2.0 to 3.0 g/L, the change of uranium removal rate is gentle, and the final uranium removal rate can reach 84%. The results show that the conductivity of the solution is better with more free ions in the solution than less free ions.

3.5 Effect of pH

This experiment changes the pH value by adding NH3·H2O and HCl. The effect of varying initial pH from 3.5 to 7.7 at uranium solution of 10 g/L, 30 V voltage, and 10 cm electrode gap. The result is given in Fig.5.

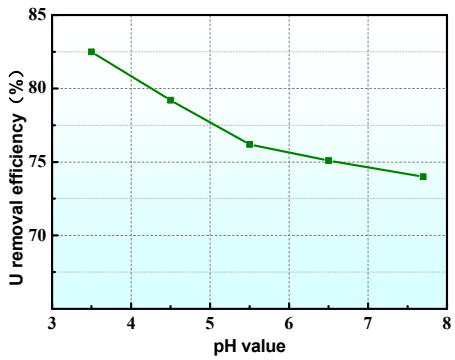

Figure 5. Effect of pH on U Removal Efficiencies

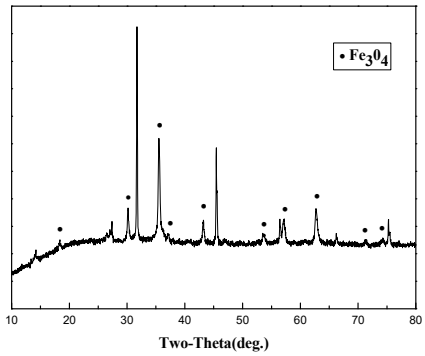

Figure 6. XRD Pattern of Precipitate

The results in the Fig.5.presented that U removal efficiencies is getting lower as the initial pH increased. U removal efficiencies was 82.5% at an initial pH of 3.5, while U removal efficiencies was reduced to 72.8% at an initial pH of 7.7.The experimental results show that U removal efficiencies has a relationship with the initial pH . Under acidic conditions, U removal efficiency is higher than alkaline solution.

3.6 Characteristics of Precipitate

Analysis of the precipitates by XRD gave results consistent with the reaction stages previously determined from analysis of experimental phenomenon. The precipitate obtained in electro-deposition was identified basically as magnetite by XRD. The diffraction pattern for sample is shown in Fig.6. All peaks have been labeled as the corresponding ones to Fe3O4.

4. Conclusion

In summary, the initial concentration of uranium has a great effect on the U removal efficiency and increasing with uranium initial concentration raised. The electrode spacing has influence on the U removal efficiencies. U removal efficiency reach the highest at the electrode spacing is 5 cm. The results indicated that the maximum removal efficiency was obtain at voltage 30 V, initial pH 2.6, dosage of NaCl is 3 g/L and electrode gap of 5 cm.

Acknowledgments

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