



Immobilization of uranium into magnetite from aqueous solution by electrodepositing approach



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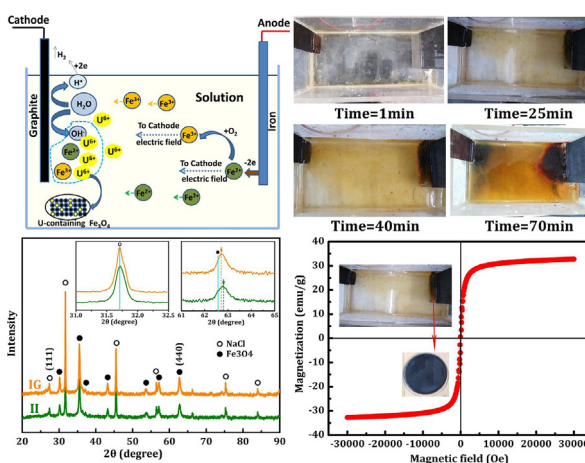
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HIGHLIGHTS

- Incorporating uranium into magnetite by electrodepositing method was proposed.
- Process parameters were investigated.
- The mechanism of incorporating uranium was discussed.
- Uranium-containing precipitate has environmental stability and can be used as resource.

GRAPHICAL ABSTRACT



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ABSTRACT

Immobilization of uranium into magnetite (Fe_3O_4), which was generated from metallic iron by electrochemical method, was proposed to rapidly remove uranium from aqueous solution. The effects of electrochemical parameters such as electrode materials, voltage, electrode gap, reaction time and pH value on the crystallization of Fe_3O_4 and uranium removal efficiencies were investigated. More than 90% uranium in the solution was precipitated with Fe_3O_4 under laboratory conditions when uranium concentration range from 0.5 mg/L to 10 mg/L. The Fe_3O_4 crystallization mechanism and immobilization of uranium was proved by XPS, XRD, TEM, FTIR and VSM methods. The results indicated that the cationic (including Fe^{2+} , Fe^{3+} and U(VI)) migrate to cathode side under the electric field and the uranium was incorporated or adsorbed by Fe_3O_4 which was generated at cathode while the pH ranges between 2–7. The uranium-containing precipitate of Fe_3O_4 can exist stably at the acid concentration below 60 g/L. Furthermore, the precipitate may be used as valuable resources for uranium or iron recycling, which resulted in no secondary pollution in the removal of uranium from aqueous solution.

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1. Introduction

A large amount of uranium-containing wastewater were produced by activities associated with using of radioisotopes in nuclear power plants, nuclear weapon testing, mining and medical research. Uranium is a serious threat to the environment due to its chemical toxicity and radioactivity. Uranium usually exist in several oxidation states such as U(III), U(IV), U(V) and U(VI), among which U(VI) and U(IV) are the dominant states in the environment [1]. It is well known that U(VI) of high mobility and migration exists in aqueous solutions causes serious pollution and harms to human health unless it is absorbed or changed into immobile U(IV) by reduction or precipitation methods. Therefore, it is imperative to find an environment friendly and cost-effective method to treat U-contaminated wastewater.

To date, several methods, such as ion-exchange [2], chemical precipitation [3,4], reduction [5,6], membrane separation [7], extraction [8], bioremediation [9] and adsorption have been extensively applied for the removal of U(VI) from aqueous solutions. Especially, adsorption is considered as an attractive method because of its high adsorption efficiencies and available raw materials. Various materials such as organic materials [10–15], inorganic materials [16–21] and biological materials [22–26] were widely used to prepare adsorbents for the removal of uranium. However, the separation of adsorbents from aqueous solution, the extraction of uranium from adsorbents, and disposal of used adsorbents are difficult problems worth considering. Therefore, magnetic materials are exploited to treat U-containing wastewater by the mechanism of incorporation or adsorption. Incorporation of uranium into iron oxide minerals gained more attention of investigators to immobilize U(VI) in the environment due to its simple treatment process. Derrell Hood [27] successfully incorporated U into hematite by injecting nitrogen into ferric chloride solution under alkaline conditions. Timothy [28] have also incorporated U into hematite by using ferrihydrite crystallizes to hematite under near-neutral conditions, but this method has limited efficiency of uranium removal. For the adsorption methods, it can obtain a high uranium removal ratio, but the preparation technology of magnetic materials, such as magnetic Fe₃O₄ nanoparticles [29], Core-Shell Magnetic Fe₃O₄@poly(*m*-Phenylenediamine) Particles [30], La-EDTA coated Fe₃O₄ nanomaterial [31], magnetic Fe₃O₄/CNT nanoparticles [32], magnetic Fe₃O₄@SiO₂ composite particles [33], and novel floating macroporous alginate-agarose-magnetite cryobeads [34], is complex. The combine of advantages from incorporation and adsorption mechanisms may be a promising approach to extract uranium from waste water by magnetite materials.

Currently, electrochemical treatment is a notable clean technology, versatile and environmentally compatible for the removal and recovery of inorganic pollutants from wastewater [35,36]. The Main electrochemical methods including electrocoagulation [37,38], electroflotation [39], electrosorption [40,41] and electrodeposition. The traditional electrodeposition mechanism mainly refers to oxidation and reduction at the anode and cathode respectively, when direct current is introduced to the electrodes. Heavy metal ions in the electrolyte were reduced to their elemental forms and deposited on the surface of the cathode due to the chemical reaction. The researchers have successfully recovered nickel and copper from the wastewater by electrochemical methods [42,43], and K. Yuan [44] investigated the uranium reduction on magnetite by electrochemical method, and U(VI) were reduced to U(V) on the surface of magnetite electrode. However, it is difficult to change uranium ions into metal uranium state from solution because of the high oxidation properties of uranium. Thus, the best way is to convert uranium into a precipitate and then separated from the solution.

Based on this, an innovative U-containing wastewater treatment method, incorporating and adsorbing U simultaneously to magnetite generated in a simple electrodeposition process (EPP) by using specific electrode, was proposed in this study. Compared with the traditional methods for U removal, the formation of magnetite and capture of uranium by using electrodeposition method don't need complex process for the preparation of adsorbent, and the only raw material is iron. The uranium can be incorporated by Fe₃O₄ and the fixed uranium cannot be released again unless at a high acidity conditions (above 60 g/L H₂SO₄), which reduced the risk of secondary environmental pollution. Besides, the uranium-containing magnetite can be simply separated from aqueous solution by magnetic separation and maybe used as resource for uranium and iron recovery after treatment.

The primary objective of this study was to investigate the effects of electrode materials on uranium removal efficiency by controlling main electrochemical parameters such as initial uranium concentration, voltage, electrode gap and reaction time during electrochemical process and the mechanism for uranium removal was also discussed.

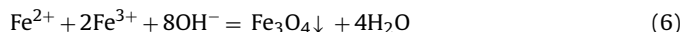
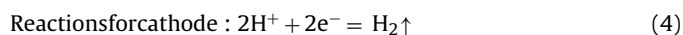
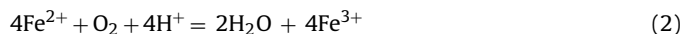
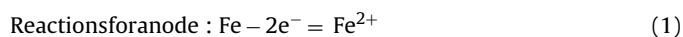
2. Material and methods

2.1. Materials

Stock solution of U(VI) (500 mg/L) was prepared by dissolving U₃O₈ (purity 99.99%) in concentrated nitric acid at 150 °C and then diluted to the desired concentration. Iron (purity 99.999%) and graphite sheet (purity 99.999%) were used as electrode materials.

2.2. Principle

The EPP was carried out by using iron as anode (AI), graphite (IG) or iron (II) as cathode to remove uranium from acidic solution. The AI was dissolved in acidic solution at the beginning of EPP with the generation of Fe²⁺ or Fe³⁺ ions. These ions were then transformed to Fe₃O₄ by adjusting the significant electrodeposition parameters including voltage, electrode gap, reaction time and pH value. The uranium was synchronously incorporated into the crystal lattice or adsorbed to the surface of Fe₃O₄ during its crystallization. The main reactions occurred in the EPP may be expressed by the following equations:



2.3. Batch experiment for U removal

500 mL of predetermined uranium solution was decanted into a self-made electrolytic cell with dimensions are 25 cm (length) × 10 cm (width) × 8 cm (height). The initial pH, current and weight of the AI were recorded before the experiment started. The EPP were subsequently initiated by controlling these main parameters including initial uranium concentration, voltage, electrode gap and reaction time. A certain amount of liquid from the electrolytic cell was taken out to measure the uranium concentration and pH value at specific intervals, and the current was recorded simultaneously. The residual liquid was filtered and the precipitate was dried under vacuum conditions at 80 °C for other analyses at the end of the EPP.

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