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Removal of uranium from aqueous solution by two-dimensional electrosorption reactor

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ABSTRACT

The radioactive wastewater discharged into the environment resulted in hazardous to public health. Electrosorption could be regarded as one of the efficient methods for removal of the uranium U(VI) containing radioactive wastewater. Herein, graphite and ACF were evolved as cathodes to design two-dimensional electrosorption reactor for elimination of U(VI) radioactive contamination. Effect of electrolyte concentration, electrode distance and voltage on the uranium removal efficiencies were investigated for optimization of sorption behavior. The removal efficiencies of U(VI) increased remarkably in the presence of voltage for both ACF and graphite, comparing to that in the absence of voltage. The ACF showed a greater removal efficiency of U(VI) (i.e., 76%) rather than that derived from graphite system (i.e., 61%). Under the conditions of 0.05 mol L^{-1} $NaNO_3$ electrolyte, 3 cm electrode distance and 5 V applied voltage, the electrosorption exhibited the highest removal efficiency of U(VI). The ACF could be reused as the absorbed U(VI) was able to be effectively desorbed by switching the electrodes in the two-dimensional reactor. The results of desorption and SEM–EDX analysis of ACF electrode suggested that the enhanced U(VI) removal efficiency was ascribed to electric induced polarization on the surface cathodes.

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1. Introduction

The nuclear industry activities, such as mining (Morrison and Cahn, 1991) and nuclear power generation (Antonijević et al., 2017), inevitably generate a considerable amount of highly toxic radioactive wastewater. The release of radioactive wastewater into the natural environment poses a severe threat to public health. The radioactive pollutants, particularly for those containing uranium (U(VI)), would cause a great number of fatal diseases, such as kidney damage, lung insufficiency, muscle cramp, liver damage and even death (Faghihian et al., 2012; Manigandan and Shekar, 2014). Prior to discharging U(VI) into the environment, the segregation of U(VI) from the waste stream is of great paramount to ensure the environmental safety. A number of techniques, for instance, co-precipitation (Carli et al., 2015), membrane filtration (Wannachod et al.,

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2016), nano-zero valent iron reduction (Klimkova et al., 2011; Ding et al., 2015; Kong et al., 2016), ion exchange (Tang and Reeder, 2009; Liu et al., 2016), adsorption onto activated carbon (Kutahyali and Eral, 2004), carbonaceous nanofibers (Sun et al., 2016), graphene oxide (Li et al., 2012; Wang et al., 2016) and functionalized graphene oxide (Sun et al., 2015, 2017) and electrosorption (Wang et al., 2012), have been developed in removing U(VI) from aqueous solutions. Of all the methods mentioned above, electrosorption technique possesses unique features of low cost, high antipollution ability, environment friendly and simple operation (Wang et al., 2012) and thus is more prevalent in treating radiative wastewater. Electrosorption applies the electrical potential as the driving force and combines with the outstanding properties among traditional adsorption and ion exchange mechanisms. The metal ions in wastewater were effectively trapped by an opposite charge on the electrode surface, adsorbed and fixed by the electrical double layer. Consequently, these metal ions were able to be successfully removed from the aqueous phase (Choi and Choi, 2010; Nadakatti et al., 2011).

To design a highly efficient electrosorption system, electrode plays a critical role in the removal of various contaminants (Shmidt et al., 1997). An electrode with great chemical and electrochemical inertness, high electronic conductivity and large surface area (Zhou et al., 2015; Sharma et al., 2015; Dong et al., 2015) is often attracting a huge attention because it could significantly enhance the removal efficiency. Carbon-based materials, including activated carbon fiber and graphite, are deemed to match with the aforementioned, being considered to be the most prevalent electrode materials in electrosorption process (Xu et al., 2015a,b; Zhu et al., 2016). Notably, activated carbon fiber (ACF), which can be easily attainable, has high specific surface area and good chemical and electrical inertness (Hatfield et al., 1996), but it is limited in adsorption for uranium due to its limit ability. The ACF has the potential of being superior electrode for enhancing sorption pollutants. The ACF adsorption ability to aniline from aqueous solutions was highly enhanced after the application of voltage (Han et al., 2006). Desorption ability in the presence of electric was not discussed. Therefore, electrosorption equipped with ACF electrode should be paid more attention.

In this work, electrosorption reactors equipped with ACF and graphite cathodes were designed to adsorb and remove uranium from aqueous solution. The aim of this paper is to comparatively explore the electrosorption behavior of uranium on the ACF and graphite electrodes for further guiding the design of the reactor. The influences of processing parameters, such as the distance between cathode and anode, applied voltage and electrolyte concentration in the electric induced sorption process were investigated in detail. The electro-desorption performance was investigated by switching the electrodes.

2. Materials and method

2.1. Materials

Uranyl nitrate hexahydrate $UO_2(NO_3)_2 \cdot 6H_2O$ was purchased from Chushengwei Chemical Co. in Hubei province. Uranium stock solution of 1000 mg L^{-1} was prepared by dissolving $UO_2(NO_3)_2 \cdot 6H_2O$ in deionized water. Other reagents were analytical grade purchased from Sinopharm Chemical Reagent Co. Ltd. and used as received without further purification. The activated carbon fiber (ACF), as an electrode, was purchased from Zichuan Carbon Fibers Co., Qinghuangdao, China. ACF, with a weight of 0.2000 g, was fixed at plastic net (as supporter) to fabricate an ACF electrode. The graphite electrode sheet with 1 mm thickness was purchase from Baofeng Jieshi Carbon Co. in Henan, China. The graphite electrode was cut into rectangular sheet with an identical sectional area of ACF electrode.

2.2. Design of the electrosorption reactor

The electrosorption reactor used in this work was designed and processed with Lucite plate with a volume of 200 ml. The as-prepared ACF and graphite electrode was placed in the reactor, respectively. The distance between cathode and anode was adjusted according to the experiment condition. The voltage was supplied from a DC power supply (1051A, Yihua Co., China).

2.3. Electrosorption test

The electrosorption experiment was conducted in a batch experiment. For each test, 100 ml aqueous solution containing 10 mg L^{-1} uranium was used. The voltage was provided by a DC power supply. The net adsorption by the electrode was carried out without any applied voltage. During the sorption process, 5 ml reaction solution was withdrawn at given time intervals. The concentration of uranium in the aqueous solution was determined by a reported spectrophotometric method using a Shimadzu UV-3150 spectrophotometer (Nie et al., 2013). The removal efficiency of uranium was calculated by $(C_0\text{-}C_t)/C_0 \times 100\%$. The C_0 and C_t (mg L^{-1}) were the initial concentration and residual concentration at t time (min), respectively. The electro-desorption was conducted by exchanging the electrode of the DC powder. Scanning electron microscopy (SEM) images were recorded using a JEOL JSM-6330F-mode Field Emission Scanning Electron Microscope (JED-2300).

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