Removal of Uranium from Radioactive Liquid Waste by Surface-modified Biosorbents

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1. Introduction

The radioactive liquid waste containing uranium mainly generated during the operation of nuclear power plants or related facilities such as nuclear research laboratories and nuclear fuel manufacturing plants. Due to its radioactivity and high toxicity, uranium is one of the most seriously hazardous materials for the environment and finally human body [1]. Therefore the development of treatment technology for the uranium-containing liquid waste has been required. Recently, biosorption technology using microorganisms, plants, and other various biomasses has been considered as an alternative to the metal adsorption process by chemical adsorbents [2]. In this study, the feasibility test of biosorption for uranium removal from liquid waste was performed, and the removal properties of uranium by pretreated brown algae were evaluated in various experimental conditions.

2. Experimental section

In this section, the details on the materials and methods to evaluate the removal efficiency of uranium by surface-modified brown algae biomass are described.

2.1 Uranium Liquid Waste

The simulated radioactive liquid waste was prepared by dissolving uranyl nitrate ($UO_2(NO_3)_2 \cdot 6H_2O$) into deionized water as 10 mg/l and 100 mg/l of initial uranium concentrations. In order to evaluate the removal efficiency of uranium at different conditions, the ionic strength and pH of the simulated liquid waste were changed by sodium nitrate ($NaNO_3$) and nitric acid (HNO_3), respectively.

2.2 Adsorbents

Several chemical adsorbents were used for the comparison of the uranium adsorption efficiency. The selected adsorbents such as activated carbon (AC), acidified-activated carbon (AAC), zeolite 4A, X, Y, and mordenite (MOR) were tested at the same condition where the prepared biosorbents were tested.

The non-living biomass of brown algae was obtained from Korean southern coast. Three types of brown algae such as *Sargassum fluvellum*, *Undaria pinnatifida*, and *Laminaria japonica* were selected for the preparation of the surface-modified biosorbents. The brown algae were dried in an oven at 60 ^oC overnight, and ground in a mortar. They were sorted by sieving as four different sizes (>2mm, 2mm ~ 0.2mm, 0.2mm ~ 0.05mm, and <0.05mm). Surface-modification of biomass was carried out by mixing 0.1 M hydrochloric acid (HCl) as 10g biomass per liter and stirring for 12 hrs in room temperature. After the acidification, the biomass rinsed several times with deionized water. Then the surface-modified biomass was dried in an oven at 60 ^oC overnight.

Fig. 1. Picture of *L. japonica* biosorbent and its raw material.

2.3 Adsorption experiments

The uranium adsorption efficiency of the prepared biosorbents was analyzed in various experimental conditions by several types of biosorbents prepared with different particle sizes. All the batch experiments were conducted by using a rotating shaker in room temperature. The uranium concentrations of the liquid samples obtained from the adsorption experiments were analyzed by a colorimetric method using Arsenazo III to calculate the removal efficiency. The finally obtained solid samples loading uranium were analyzed by scanning electron microscopy (SEM, SNE 4500M, SEC, Korea), energy dispersive spectroscopy (EDS, QUANTAX, Bruker, Germany), and X-ray diffractometer (XRD, D2 Phaser, Bruker, Germany).

3. Results and Discussion

3.1 Comparison of adsorbents

Uranium removal efficiencies of several chemical adsorbents and prepared biosorbents were evaluated at 100 mg/l initial uranium concentration and pH 4 (Fig.

2.). One gram per liter of each adsorbent was used in the batch experiment for 6hrs.

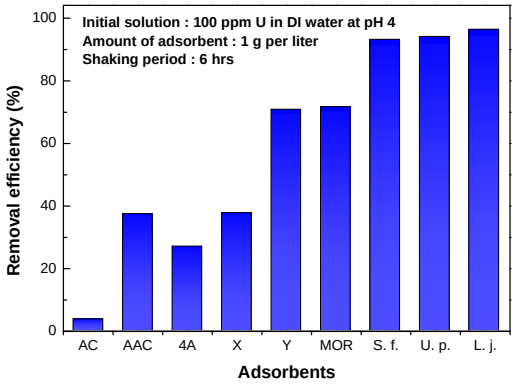

Fig. 2. Removal efficiencies (%) of uranium by several adsorbents at 100 mg/l initial uranium concentration and pH 4.

Activated carbon is a common adsorbent for the removal of various toxic materials, and the acidified-activated carbon is known to be more efficient for uranium adsorption because of the increase of surface functional groups [3]. Also zeolite is one of very useful materials for cation exchange in both liquid and atmospheric environments. In the case of zeolites, the sorption of uranium ions from liquid phase has been studied by using zeolite X, Y, mordenite, clinoptilolite etc [4]. It is known that uranyl ions (UO_2^{2+}) are mainly exchanged with the cations from the large cavities of those zeolites. Although these chemical adsorbents are commonly considered materials for uranium removal processes, the surface-modified biosorbents showed higher uranium removal efficiencies (more than 90%) at the same experimental condition than chemical adsorbents compared (less than 70%). This result demonstrates that the application of brown algae biosorbent is feasible for the uranium removal from liquid waste.

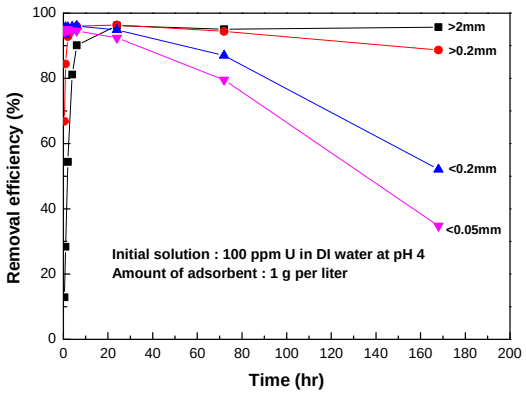

Fig. 3. Changes of uranium removal efficiencies (%) by *L. japonica* biosorbents with contact time.

3.2 Adsorption properties of biosorbents

Fig. 3 showed that the kinetics of uranium uptake was mainly dependent on the particle size of the prepared *L. japonica* biosorbent, which gave us the optimal particle size for the stable operations of liquid waste treatment process. The adsorption rate was significantly increased with decrease of particle size, moreover the desorption rate after long contact time was increased at the small particles. The adsorption capacity decreased with increase of the ionic strength of the solution. Although the biosorbent have a strong affinity for uranium ion, it does not have selectivity for a specific single ion. Therefore it should be considered that to treat the real liquid waste containing complex and various ions might have more limitations than the simulated liquid waste used. The effect of pH on the uranium adsorption efficiency of *L. japonica* biosorbent showed that highest uptake occurred at pH 4 in a solution with 1 M sodium nitrate.

The fact that the biosorption process could alternate with the former uranium removal processes using chemical materials was demonstrated from this study. In conclusion, the surface-modified biosorbent from brown algae is shown to be a favorable biosorbent for uranium removal from radioactive liquid waste.

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