

Dicarboxylic acid로 그래프트 공중합한 chitosan에 의한 우라늄 제거

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Removal of uranium in water system by grafted chitosan with dicarboxylic acid

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Introduction

WHO (1998) has recommended 20g uranium per liter of water as a safe limit for drinking purposes for residents in USA. These levels are set to represent a concentration that does not result in any significant risk to health over the lifetime drinking of water. The health and environmental protection agencies have recommended safe limit of uranium in drinking water for human beings. The World Health Organization(WHO, 1998) and the United States Environment Protection Agency (USEPA, 1992) in general have recommended $2.0\mu\text{g}/\ell$ of uranium concentration in drinking water as the safe limit. ICRP-30 (1979) has suggested this limit as $1.9\mu\text{g}/\ell$.

Recently a country presented research report(The Institute for Environmental Research, 1999~2002) showed the actual condition of radioactive pollution on groundwater. USEPA presented removal method of uranium in groundwater. And USEPA presented following treatment methods : coagulation-sedimentation, soft water, adsorption of activated carbon, an anion exchange, NF(Nanofiltration) and RO(Reverse osmosis).

Chitosan, a maximally deacetylated chitin, is soluble in dilute acidic solutions depending on the degree of acetylation of chitin. Chitin and chitosan had been left unexploited, even though they are widely distributed in natural environment. Their usefulness has recently been noticed and they have been used as important materials in medicines, soil conditioners, and a hypocholesterolemic agent.

The authors developed more effective flocculants or metal uptaking agents through graft-copolymerization of deliberately selected alkenoic acids onto chitosan.

Graft-copolymers are of importance in view of new industrial applications such as water absorbents, ion exchangers, flocculants, membranes, modified electrodes, and principally chelating agents.

The amide ligand has played a very important role in the development of actinide chemistry and carboxylic groups are often considered the only functional groups of substances responsible for the complexation of metal ions. However, graft-copolymerization on chitin and chitosan have been scarcely reported in the literature,

while on cellulose, starch, wool, and cotton, they have been studied to a greater extent.

The nitrogen in the amino group of the chitosan molecule acts as an electron donor and is presumably responsible for selective chelating with metal ions.

The chitosan grafted with itaconic acid(CsIa) was found more effective. The removal rate of uranium by CsIa was compared with pure chitosan(Cs) and activated carbon(AC) in this experiment.

The removal experiment was carried out through two systems(static system and flow system).

Materials and methods

Cs and CsIa were casted into beads(2mm in wet form) and treated with hexamethylene diisocyanate for stability in acidic aqueous solution through cross-linking of the beads surfaces. The removal rate of by chitosan grafted with Itaconic acid is superior to compared with removal rate by pure chitosan in uranium removal both in the static and the flow systems for standard solution(analysis degree uranium solution solubled in 5% HNO₃) and groundwater.

In the static system, a certain amount of testing water is confined in a vessel and some amount of beads are casted into in order to adsorb uranium(U) for a certain time. And then the remained uranium in water vessel is determined by ICP.

In the flow system, a certain amount of beads are packed in a column(Φ40×H300) and the test water is forced to pass through so that uranium is adsorbed on the surface of beads.

The author tried to develop a more effective and economical use of chitinous demineralization systems by changing sizes of beads packed in(dry wet. 8g) filtering columns and controlling flow rate of test water on down flow(uranium standard solution 1ppm, pH 6.0) passing through the columns.

Beads with different sizes(2mm, 3mm) were packed in columns separately. And then the primarily treated test water was introduced into columns with change of flow rate.

The removal rate is appreciated by determining the uranium amount contained in the effluent.

Fig.1 is flow sheet for graft-copolymerization reaction and Fig.2 is preparation method of chitosan beads and Itaconic acid grafted chitosan beads.

Results and discussion

Fig.3 and Fig.4 showed the FT-IR spectra of Cs, CsIa. Characteristic absorption on the IR spectra, wave number 1650cm⁻¹ were assigned to the carboxyl group. The IR spectra showed absorption bands at 1589 and 1650~1740cm⁻¹, attributed to the -NH₂ and C=O stretching. Therefore monomers were graft-copolymerized on chitosan.

The experimental results are as below :

Fig.5 showed uranium uptake at CsIa bead sizes. The experimental results explain that the efficiency of metal uptake increased with decrease of bead size.

Fig.6 showed the uranium removal ability of Cs beads, CsIa beads, and the same amount of activated carbon(dry wet 0.5g) in the static system which 1ppm of uranium contained 100ml(pH 6) with adsorption time(96hr). The experimental results showed that the grafted chitosan CsIa was greatly superior to Cs or AC compared with removal rate of uranium.

Fig.7 showed uranium removal rates by Cs beads, CsIa beads, and the same amount of activated carbon in the flow system with the effluent amount(ml). Flow rate was 5

ml/min. Fraction amount was 10ml.

The experimental results showed that the grafted chitosan CsIa was superior to Cs or AC in uranium uptaking.

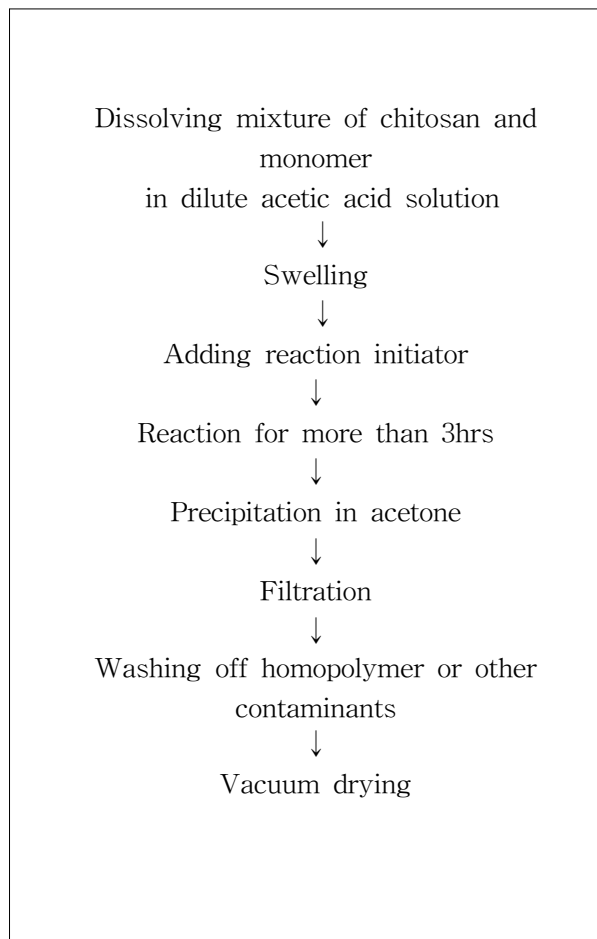


Fig.1. Flow sheet for graft-copolymerization reaction

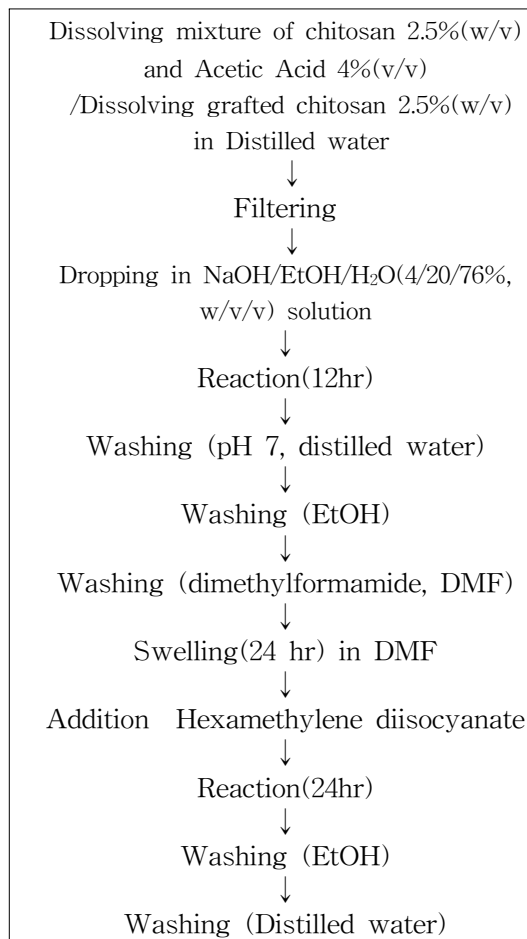


Fig.2. Preparation method of chitosan beads and Itaconic acid grafted chitosan beads

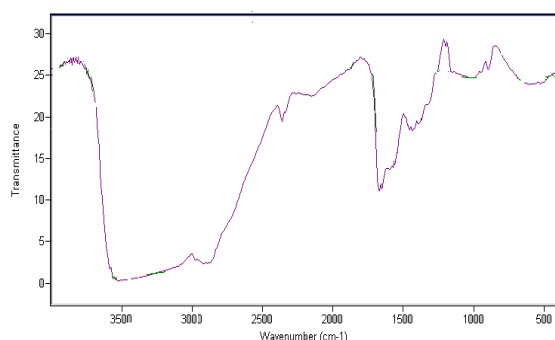


Fig.3. FT-IR spectra of Chitosan

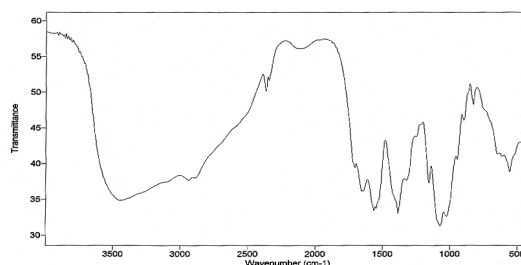


Fig.4. FT-IR spectra of Itaconic acid grafted chitosan

The experimental results explained that the efficiency of uranium uptake increased with decrease of beads size. Uranium ion uptaking efficiencies were examined with variations of sizes of beads in column.

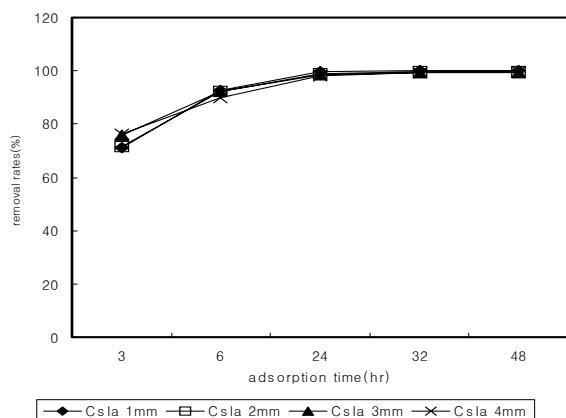


Fig.5. Uranium uptake with CsIa bead sizes

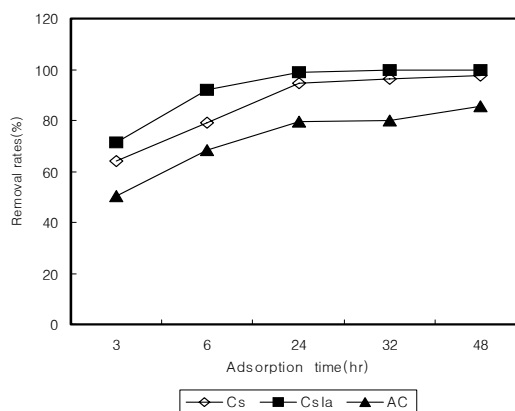


Fig.6. Comparison of uranium removal rates with Cs and CsIa, and activated carbon in a static system with 1ppm U

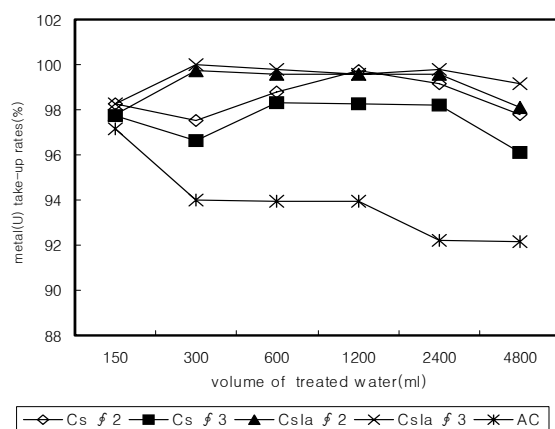
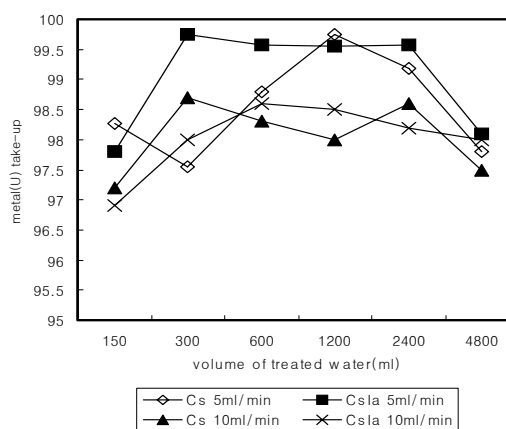
Fig.7. Comparison of uranium removal rates with Cs and CsIa, and activated carbon packed in $\varnothing 4\text{cm} \times L 30\text{cm}$ columns with function of effluent volume in the flow system with 1ppm UFig.8. Effect of flow rates on U removal rates by the beads of Cs and CsIa packed in $\varnothing 4\text{cm} \times L 30\text{cm}$ columns with 1ppm U (beads size : 2mm)

Fig.8 showed uranium removal rates by Cs beads, CsIa beads, and the same amount of activated carbon in the flow system. Flow rate was 5ml/min. Fraction amount was 10ml. The experimental results showed that the grafted chitosan CsIa was superior to Cs or AC in uranium uptaking. The experimental results explained that the efficiency of uranium uptake increased with decrease of bead size.

The chitosan grafted with itaconic acid was superior to pure chitosan and activated carbon in compared of uranium removal both in the static and the flow systems.

Reference

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