

REMOVAL OF URANIUM BY SODIUM ALGINATE-BASED DIPHOSIL BEAD

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ABSTRACT

Chemical wastes containing small amounts of uranium pose an environmental problem, when we finally dispose of them as industrial wastes. Especially for the removal of actinides and multivalent ions, a new organic-inorganic resin has been developed by ANL, which is called as diphosil resin, having chelating diphosphonic acid groups in the silica support. diphosil has been applied to treat uranium-contaminated wastewaters due to its high affinity for uranium ions, but these applications are limited for a continuous process, because the resin is a powder type. The conventional process for removing heavy metals from aqueous solutions has used immobilized biosorbents in the form of alginate gel beads.

In this study, a method of immobilizing diphosil powder within alginate beads is adopted to make a bead type from the powdered resin. Diphosil powder was mixed with a 4% sodium alginate solution and the mixed solution was dropped into a 0.1M CaCl₂ solution by an injector to make the beads.

The experiments were performed to determine the physical properties such as the water content in the bead, adsorption rate with various compositions of the diphosil, and the effect of the uranium concentration on adsorption rate. The adsorption efficiency between the dried beads and wet beads were compared for a further application of continuous process.

The diphosil content is well distributed in each bead when viewed from the surface of the dried bead. The sodium alginate bead itself showed the capability for uranium uptake to above 60%, but the value was decreased to below 30% after equilibrium.

The rate of uranium adsorption increased with an increasing content of the diphosil in the sodium alginate bead. The diphosil resin itself showed a very fast uptake of uranium from the early stages and then the rates leveled off. The adsorption rate from 0.5g of the bead with 4% sodium alginate and 4% diphosil showed a similar level of adsorption with the one from 0.25g of diphosil. As a result, the diphosil bead showed a remarkable capability for the uptake of uranium with a factor of 4, when considering the diphosil content in the bead.

In conclusion, a considerable potential exists for further applications of a continuous adsorption process by using a bead form of diphosil within sodium alginate.

INTRODUCTION

Chemical wastes containing small amounts of uranium cause environmental problems, if those wastes exceed the concentration of the EPA standard, 20 $\mu\text{g/L}$, and cannot be disposed of as an industrial waste. The concentrated sludge generated from the treatment of chemical wastes should be additionally dried and packaged into a drum, and categorized as an radioactive waste.

Recently, the Argonne National Laboratory(ANL) has developed diphosil resin to specifically remove actinides or multivalent metals, which have sulfonic and diphosphonic acid groups in their silica base. The resin is applied for the removal of uranium due to its characteristics of a high affinity with uranium[1]. Diphosil has an excellent selectivity for uranium, but the type of the resin is a power form, and its application for a continuous process is limited.

On the other side, the immobilization technology for biosorbents or biotissues for the uptake of heavy metals is being developed, and sodium alginate is used to make a bead form of the biosebents from bacteria, yeast[2], or biotissues from plant roots[3].

In this study, the immobilization technology is applied to make a bead form from the powdered diphosil by embedding diphosil powder into sodium alginate.

Physical properties, adsorption characteristics for uranium, effect of the uranium concentration, and the stability of the bead generated are studied in this paper.

EXPERIMENTAL METHOD

Materials

Chemicals and resin used in this study are as follows:

- Diphosil: Silica diphonix resin, 60-100mesh, Eichrom Technologies Inc., U.S.A.
- Alginic acid, sodium salt: Aldrich Chemical Co. Inc., Germany
- Uranium solution: 957 $\mu\text{g/ml}$ of U, Atomic adsorption standard solution, Aldrich Chemical Co. Inc., Germany

Immobilization of the Diphosil Powder into Sodium Alginate

Sodium alginate is solved into the deionized water little by little with a constant stirring, and after mixing for several minutes, the diphosil powder is introduced into the mixing solution very slowly because of the insolubility in water, and the mixing is maintained for several minutes. Then, the stirring is slowed down to remove the air bubbles within the mixed solution.

Preparation of the Bead

The mixture of sodium alginate and diphosil powder with water is transferred into the injector and it is dropped into 0.1M CaCl_2 solution. The bead produced is washed with the deionized water and stored at room temperature.

Measurement of the Uranium Adsorption

Various concentrations of the uranium solution were prepared by the amount of standard solution added. The adsorption characteristics of the bead were studied using 100 ml of the uranium solution with various concentrations. Uranium concentrations were measured by an ICP-AES (Inductively Coupled Plasma-Atomic Emission Spectrometry).

RESULTS AND DISCUSSION

Physical Characteristics of the Bead

Sodium alginate is added into water with an increasing content. The concentration of sodium alginate was fixed at 4%, because it was too viscous to mix in the case of a 5% addition of sodium alginate. The remove of the bubbles within the mixed solution should be checked in the final stage of mixing by lowering the stirring speed. The mixture needs to be injected into the CaCl_2 solution carefully drop by drop.

The bead produced is an ivory-colored granular form having a dimension of 3~4mm in diameter. Water and diphosil content is listed in Table 1. Water content of the naturally dried bead at room temperature is within a 2~3% difference of original content added, and the diphosil content in the dried bead is increased in accordance with the increasing rate of original addition.

Table I. Water and diphosil content in the sodium alginate-diphosil bead

Alginate (%)	Diphosil (%)	Weight(g)		Water Content (%)	Diphosil Content (g)
		Drying			
		Before	After		
4.0	-	0.501	0.028	94.4	-
	1.2	0.498	0.037	92.6	0.009
	2.0	0.497	0.044	91.2	0.015
	4.0	0.502	0.057	88.6	0.029

The sodium alginate bead itself showed a strong brown color for the appearance of the dried bead, but the bead with the diphosil became whiter with an increasing diphosil content (Fig.1). It is considered from these observations that the diphosil is well distributed in each bead without any loss of the amount originally added.

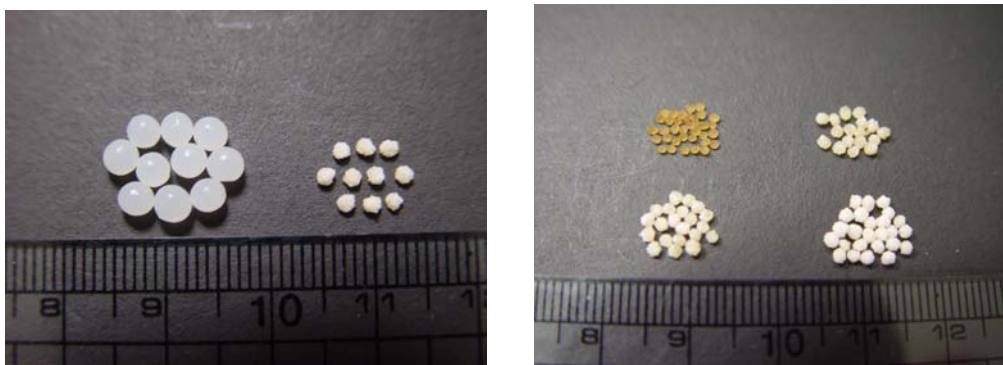


Fig.1 The wet and dried bead, and the distribution of the diphosil

Characteristics of the Uranium Adsorption

Characteristics of the uranium removal from the wet bead with various contents of the diphosil were studied using 100 ml of the uranium solution with a concentration of 11.3 mg/L at room temperature. Figure 2 shows that the sodium alginate itself showed a 68% uranium uptake, although the rate was decreased by 30% after equilibrium. It is considered to be the effects of the adsorption and desorption by the sodium alginate.

The adsorption rate of the uranium for the bead with diphosil increased with an increasing content of the diphosil. Diphosil itself shows a very fast uptake of uranium in the early stages and then it levels off. The amount of the bead with 4% diphosil, 0.5g showed a considerably high adsorption rate, 85%, when considering the same amount with diphosil. The escalating effect of the uranium adsorption of the bead is concluded due to the adsorption effect of the sodium alginate itself.

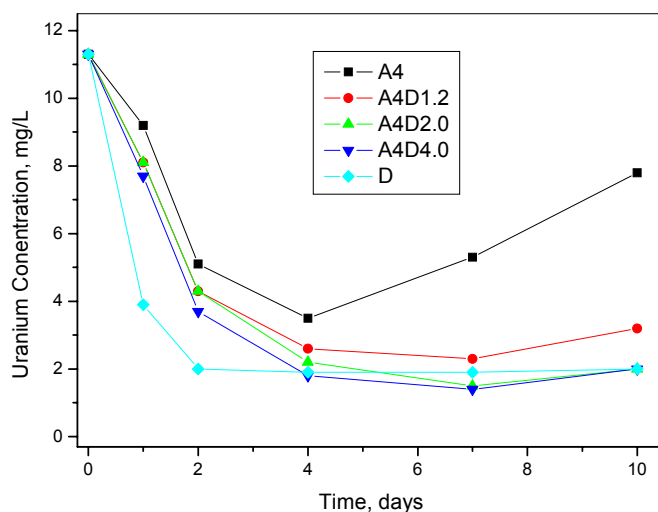


Fig. 2 Uranium removal at various concentrations of the diphosil

Fig. 3 shows the effect of the uranium concentration on the adsorption rate. Adsorption rate is decreased with an increasing concentration of the uranium. About 80% of the uranium from the 100ml solution of 95mg/L was removed by 2g of the bead.

In order to estimate the safe characteristics of the bead, the adsorption rates between the wet and dried beads were compared. A same amount of the wet bead is dried at room temperature and subjected to the uranium solution. As shown in Fig.4, there was little difference in the adsorption rate between the wet and dried beads. These results indicate no loss of the diphosil content through the expansion after drying in the solution. The 0.5g of the bead showed a similar adsorption rate to the 0.25g of the diphosil. As a result, the adsorption efficiency is improved by a factor of 4 considering the net amount of the diphosil itself except for the water content in the bead.

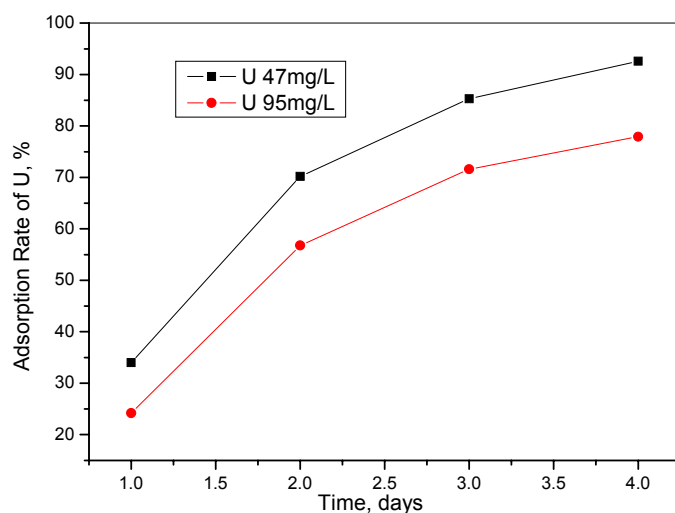


Fig. 3 Effect of the uranium concentration on the adsorption rate

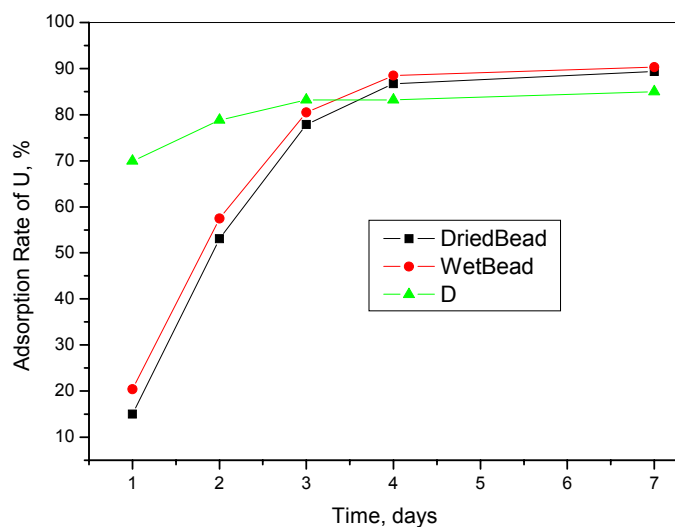


Fig. 4 Comparison of the stability and adsorption efficiency between the wet and dried beads

CONCLUSION

The bead form of diphosil has been prepared by the immobilization of the diphosil powder into sodium alginate. Characteristics of the bead produced are as follows:

- The bead produced showed a good physical stability. There was little difference in adsorption rate between the wet and dried beads.
- The bead with 4% diphosil, 0.5g showed about 90% adsorption rate for the 100ml solution of 11.3mg/L. Characteristics of the uranium removal are improved by a factor of 4.

The bead could be further utilized in a continuous process for the concentrated chemical wastes containing uranium.

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