

**Treatment of Liquid Radioactive Waste with High Salt Content by Colloidal Adsorbents –
13274**

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ABSTRACT

Treatment processes have been fully developed for most of the liquid radioactive wastes generated during the operation of nuclear power plants. However, a process for radioactive liquid waste with high salt content, such as waste seawater generated from the unexpected accident at nuclear power station, has not been studied extensively. In this study, the adsorption efficiencies of cesium (Cs) and strontium (Sr) in radioactive liquid waste with high salt content were investigated using several types of zeolite with different particle sizes. Synthesized and commercial zeolites were used for the treatment of simulated seawater containing Cs and Sr, and the reaction kinetics and adsorption capacities of colloidal zeolites were compared with those of bulk zeolites. The experimental results demonstrated that the colloidal adsorbents showed fast adsorption kinetic and high binding capacity for Cs and Sr. Also, the colloidal zeolites could be successfully applied to the static adsorption condition, therefore, an economical benefit might be expected in an actual processes where stirring is not achievable.

INTRODUCTION

Over the last decades, the processes associated with liquid radioactive waste treatment have been extensively investigated and applied into the field of industrial and environmental engineering. Liquid radioactive wastes containing short lived beta/gamma activity are simply kept in storage, but those containing long lived radionuclides and having high radioactivity have been treated by several conventional techniques, such as precipitation, adsorption, ion exchange, evaporation, micro/ultrafiltration, solvent extraction, and electrochemical processes.[1-2]

Among the techniques, adsorption processes have been considerably studied for the removal of radionuclides from various types of liquid wastes. In general, most adsorbents are obtained as fines, which might show excellent adsorption performance due to higher surface area and fast kinetics, but they also have a drawback in the difficulty of handling in the treatment process.

Therefore, many studies have been focused on the synthesis of physicochemically stable granular adsorbents to support column operations. However, forming granular adsorbents might also impact the active sites, and consequently decrease the overall adsorption efficiency and reaction kinetics.

Batch operations based on precipitation with coagulation/flocculation of fine particles is another treatment option that may better utilize the initial properties of fine adsorbents. Moreover, in general, scaling down adsorbents from bulky to colloidal size enhances the adsorption capacity

and catalytic activity, which has led to recent efforts to synthesize sub-micron colloidal adsorbents.[3]

Up to the present, the treatment of most radioactive liquid waste generated during the operation of nuclear power plants is well established and successfully commercialized. However, a process for radioactive liquid waste with high salt content, such as waste seawater generated from the unexpected accident at nuclear power station, has not been studied extensively.[4] In this study, the adsorption efficiencies of cesium (Cs) and strontium (Sr) in radioactive liquid waste with high salt content were investigated using several types of zeolite with different particle sizes.

MATERIALS AND METHODS

Synthesized and commercial zeolites were used for the treatment of the simulated liquid waste based on the seawater containing Cs and Sr, and the reaction kinetics and adsorption capacities were investigated depending on the various conditions. Table I shows the concentrations of major cations and anions in the simulated wastewater prepared by dissolving Cs and Sr into the seawater collected from Korean East Sea. The concentrations of both contaminants were 100 mg/l, and the contents of collected seawater were very similar with the average seawater contents from reference.

Table I. Concentrations (mg/l) of Major Ions in the Simulated Wastewater.

	Ca ²⁺	K ⁺	Mg ²⁺	Na ⁺	Br ⁻	Cl ⁻	SO ₄ ²⁻	Cs ⁺	Sr ²⁺
Seawater ^a	421	407	1309	10998	69	19743	2645		
Simulated wastewater	420	424	1200	10675	73	19700	2610	100	100

^a Average seawater contents of Korea

Two types of zeolites, Na-chabazite and 4A, were selected for the adsorption experiments. Each type was prepared as a colloid, full-sized particles, and ground particles.

Colloidal Na-chabazite was prepared by dispersing synthesized Na-chabazite fine powder (2.78 μm mean particle size) into water.[5-7] Pellet type AW500 (Aldrich) and its ground powder (less than 200 μm particle size) were also prepared for comparison with bulk zeolites.

Colloidal zeolite 4A was prepared by dispersing 4A fine powder (Aldrich, 2 μm mean particle size) into water. Pellet type 4A (Aldrich) and its ground powder (less than 200 μm particle size) were also prepared for comparison with bulk zeolites.

The concentrations of Cs and Sr before and after the adsorption experiments were analyzed by Atomic Absorption Spectroscopy (AAS). Also, the physicochemical and mineralogical properties of the zeolites before and after the adsorption reactions were observed by Particle Size Analysis (PSA), Scanning Electron Microscopy (SEM), Energy Dispersive Spectroscopy (EDS), and X-Ray Diffraction Analysis (XRD). The physicochemical and mineralogical data made it possible to explain the mechanisms of the different adsorption reactions. The analytical equipment used is specified in Table II.

Table II. Analytical Equipment.

Analysis	Equipment
AAS	AAAnalyst 900, PerkinElmer, USA
PSA	3500S, Microtrac, USA
SEM	SNE 4500M, SEC, Korea
EDS	QUANTAX, Bruker, Germany
XRD	D/MAX 2500G, Rigaku, Japan

RESULTS AND DISCUSSION

Colloidal Adsorbents

EDS results showed the molar ratios of Na, Al and Si of both synthesized Na-chabazite and commercial zeolite 4A was very close with those of theoretical values (Fig. 1). Through the PSA and SEM analysis, it was observed that the particle size of zeolite ranged from hundreds nm to several μm . These zeolite fine powders were dispersed in DI water with sonication and stirring pretreatments, and most of colloidal particles kept a dispersed condition for at least 2hrs.

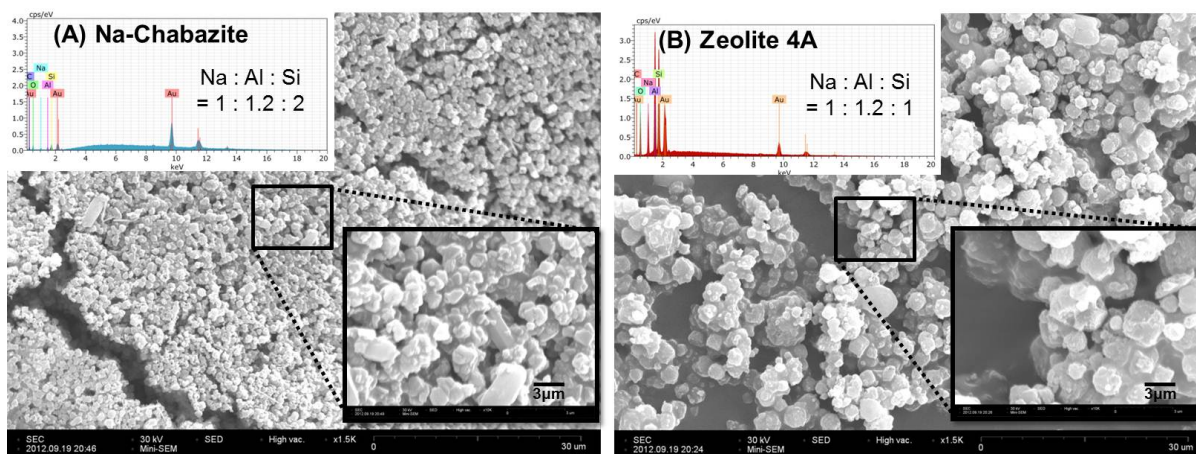


Fig. 1. SEM images and EDS results of (A) synthesized Na-chabazite and (B) commercial zeolite 4A fine powders

On the other hand, the colloidal zeolite injected into seawater was slowly precipitated with time (Fig. 2), which might be caused by the change of surface characteristics of colloidal zeolite and its self-coagulation in the condition of high salt content.



Fig. 2. Picture showing the different dispersion properties of colloidal zeolite in seawater and DI water.

Adsorption Properties

Fig. 3 represents the adsorption yield of Cs and Sr after shaking for 2hrs with different zeolite doses. In the case of Cs, Na-chabazite showed higher removal efficiency than zeolite 4A, and colloidal Na-chabazite showed the highest binding capacity for Cs for 2hrs. On the other hand, zeolite 4A showed higher removal efficiency for Sr than Na-chabazite, and colloidal 4A was the best adsorbent within 2hrs.

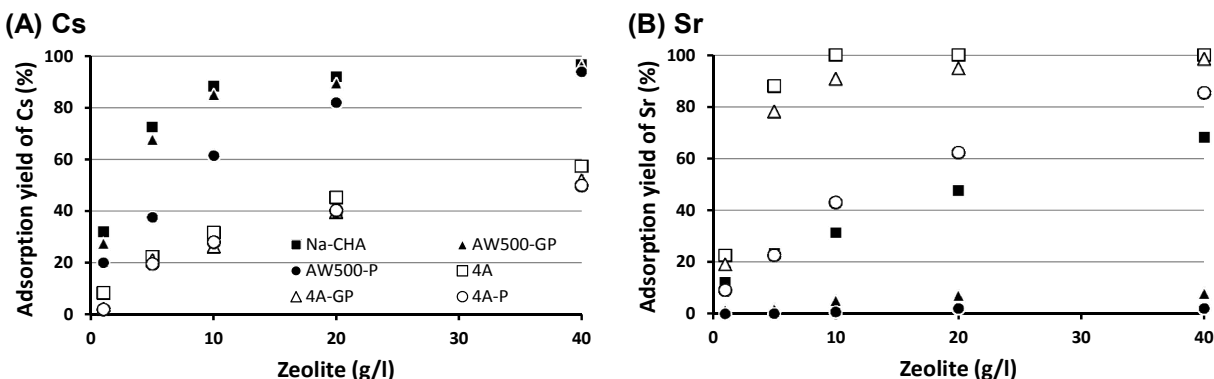


Fig. 3. Adsorption yield (%) of (A) Cs and (B) Sr after shaking for 2hrs with different zeolite doses. (GP: ground pellet; P: pellet)

The colloidal Na-chabazite also showed relatively good performance for the removal of Sr compared to the bulk Na-chabazite. Therefore, the colloidal Na-chabazite might be the optimum choice for the actual waste seawater generated from the nuclear power accident, which contains lower Sr content than Cs.

In order to confirm the reaction kinetics of Cs and Sr adsorption in static conditions, the adsorption yield in the constant condition (without shaking, 10g/l zeolite dose) was evaluated over time. In these kinetic experiments, both colloidal zeolites showed significant higher efficiencies compared to bulk zeolites. Therefore, colloidal zeolite should offer a significant advantage to wastewater treatment processes where stirring is not achievable.

The results also show that the adsorption rate increases with the decrease of particle size. The adsorption reaction of most zeolites reached to the steady state within around 5 min for Cs and 20 min for Sr in this experimental condition, which indicates that the reaction rate of zeolite is very fast.

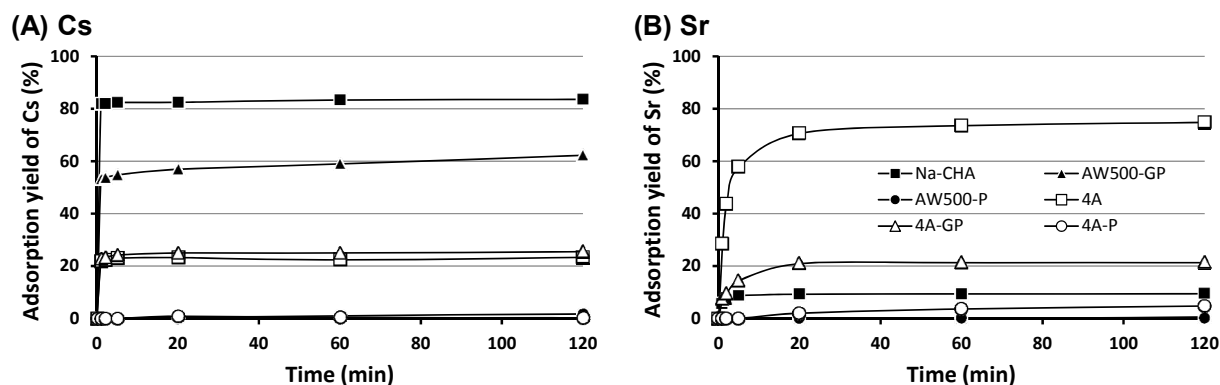


Fig. 4. Adsorption yield (%) of (A) Cs and (B) Sr in the static condition (without shaking, 10g/l zeolite dose) with time. (GP: ground pellet; P: pellet)

CONCLUSIONS

The experimental results emphatically demonstrated that colloidal zeolite adsorbents have fast adsorption kinetic and high binding capacity for the target contaminants. Because colloidal zeolites may be successfully applied to the static adsorption condition, an economical benefit might be expected where stirring is not achievable. In order to improve the performance of colloidal adsorbents for radioactive liquid waste treatment, common post- adsorption steps such as coagulation/flocculation for suspended particles and solid/liquid separation, should be employed.

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