

Surface Modification of Alginate Beads for Enhanced Affinity to Barium Ions in ^{131}Cs Separation Process

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1. Introduction

^{131}Cs with a short half-life time and high average radiation energy is attractive radioisotope for brachytherapy because they can treat a cancer effectively within a short time, with its shorter half-life and higher average radiation energy than commonly used radioisotopes: ^{125}I or ^{103}Pd .

It is necessary to separate the ^{131}Cs (i.e., the decay product) from the ^{131}Ba (i.e., the mother isotope) to obtain a carrier-free radioactive source with a high specific radioactivity. There are two typical separation processes for the ^{131}Cs production. The first process based on the precipitation of barium with a salt such as carbonate or nitrate [1]. However, this method requires long process time for the repeat of several precipitation steps to achieve high nuclidic purity of ^{131}Cs , resulting in increased decay loss. The other process is a separation using chelating agents with a high selectivity to cesium [2]. While this process has fewer separation or purification steps followed by the solvent extraction, its application to large-scale processes is limited.

Previous study [3] presented the alternative methodology of ^{131}Cs production based on the column separation with alginate beads. ^{131}Cs as a medical radioisotope requires a high radionuclidic purity in solution, it is important to keep the Ba adsorbed on the alginates does not desorb. Herein, we suggested a strategy for the surface modification of alginate beads to reduce its affinity of alginate beads to cesium ions while reinforcing the affinity to barium ions. Through the adsorption experiments of alginate beads, we could present the effectiveness of functional group modification by understanding its adsorption behavior such as the isotherm and kinetics.

2. Experiments

2.1. Functional group modification of alginate beads

The sulfonate group, which is the functional group used for strong acid cation exchange resin, has a higher selectivity for cations as the ion valency of the cation increases. Because the Ba^{2+} has a higher ion valency than the Cs^+ , substitution of the sulfonate group is

expected to reduce the cesium affinity of the alginate beads while reinforcing their affinity to barium ions. The carboxyl group in the alginate beads was modified with an amide coupling reaction using EDC ((3-Dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride) and sulfanilic acid. The prepared Sr-alginate beads [3] were immersed in a 100 mM MES solution (2-(N-Morpholino)ethanesulfonic acid hydrate), followed by injecting 20 mM EDC and 20 mM sulfanilic acid into the solution.

3. Results and Discussion

3.1. Adsorption kinetics of modified alginate beads

The adsorption kinetics of the modified alginate beads were investigated. 0.1 g of native or modified Sr-alginate beads were mixed with 10 mL of barium solution for different times (i.e., 10-1,440 min). The initial Ba^{2+} concentration of the solution was 250 ppm (with 25 ppb Cs). We analyzed the adsorption kinetics with the pseudo-second order kinetic model (PSOKM). Figure 1 and Table 1 show the results of kinetic analysis. Both adsorption behaviors were well fitted to the PSOKM. The modified alginate beads had a higher rate constant of adsorption and reached adsorption equilibrium faster than the native alginate beads. It is essential to reduce the process time in the radioisotopes production process because the radioisotopes continuously decay to their daughter isotope. Faster adsorption kinetics through the modification is expected to be useful for the ^{131}Cs production process.

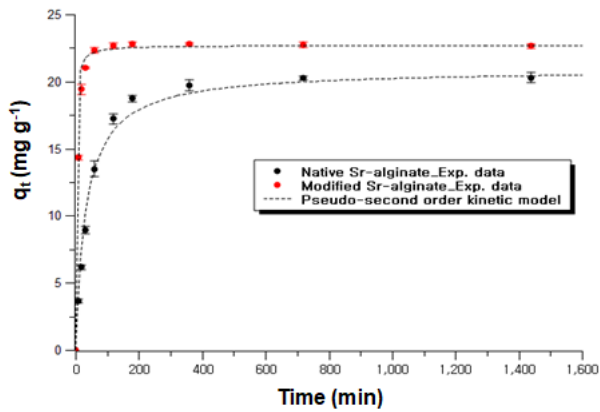


Figure 1 Kinetics of native and modified alginate beads

Table 1 PSOKM parameters of native and modified alginate beads

Sample	PSOKM		
	q_e (mg g^{-1})	K_2 ($\text{g mg}^{-1} \text{min}^{-1}$)	R^2
Native alginate beads	20.9	0.00143	0.99
Modified alginate beads	22.7	0.0314	0.99

3.2. Adsorption isotherm of modified alginate beads

The correlation between the adsorption equilibrium concentration and the Ba adsorption capacity was analyzed using the Langmuir isotherm model to estimate the maximum adsorption capacity. Both the native and modified alginate beads fitted well with the Langmuir isotherm with a high correlation coefficient, as shown in Figure 2 and Table 2. It was notable that the modified Sr-alginate had a higher maximum adsorption capacity (132.9 mg g^{-1}) and Langmuir constant ($1.163 \text{ L mmol}^{-1}$) than the native Sr-alginate (112.3 mg g^{-1} and $0.427 \text{ L mmol}^{-1}$), which represented that the modified alginate beads had the higher affinity to barium ions than the native alginate beads.

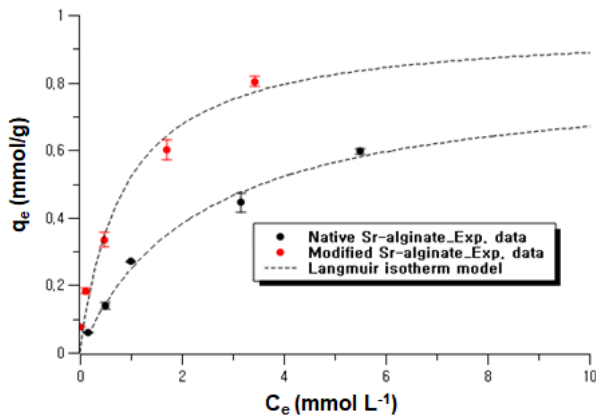


Figure 2 Isotherm of native and modified alginate beads

Table 2 Langmuir isotherm parameters of native and modified alginate beads

Sample	q_{max} (mg g^{-1})	K_L (L mmol^{-1})	R^2
Native alginate beads	112.3	0.427	0.99
Modified alginate beads	132.9	1.163	0.97

3.3. Comparison of cesium adsorption

The relatively high affinity of alginate beads to cesium ions makes it difficult to elute the low concentration of cesium ions from the beads, resulting in the decrease of the ^{131}Cs production yield. We evaluated the decrease of affinity to the cesium ions through the modification. 0.1 g of modified Sr-alginate beads were added into 10 mL of solutions with various concentrations of Cs^+ (10, 25, and 50 ppb) and mixed for 12 h. As shown in Figure 3, the modification could reduce the Cs adsorption to approximately one-third, indicating that the functional group modification of the alginate beads could reduce the affinity to the cesium ions, resulting in an increase of the ^{131}Cs production yield.

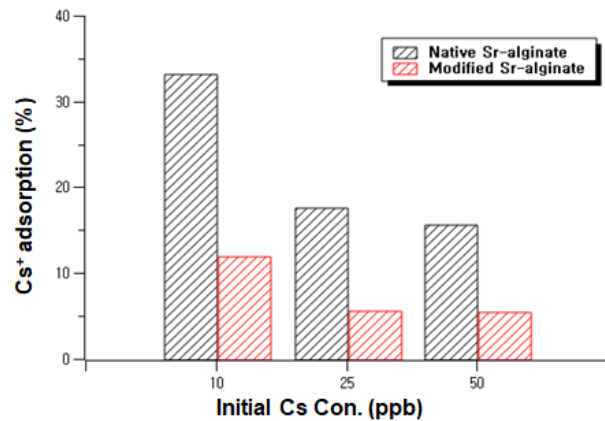


Figure 3 Comparison of Cs adsorption in native and modified alginate adsorption

3. Conclusions

To achieve higher radionuclidic purity in ^{131}Cs separation process, we presented a strategy to reduce the affinity of alginate beads to cesium ions through the functional group modification. The functional group modification of alginate beads reduced the Cs adsorption about 20% and increased the adsorption capacity of Ba from 112.3 mg g^{-1} to 132.9 mg g^{-1} . The modified Sr-alginate had a faster adsorption rate and higher affinity to barium as well.

In conclusion, surface modified alginate beads exhibit better performance in ^{131}Cs production with shorter

process time, higher radionuclidic purity as well as larger production yield.

Acknowledgements

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