

Adsorption and Separation Behavior of Sr(II) and Y(III) by Extraction Chromatography using a Silica-based DtBuCH18C6 Adsorbent and Its Medical Application

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To separate ⁹⁰Y from the ⁹⁰Sr-⁹⁰Y mixture, a silica-based (DtBuCH18C6+Dodec)/SiO₂-P adsorbent was prepared by impregnating 4',4'(5'')-di(tert-butylcyclohexano)-18-crown-6 (DtBuCH18C6) extractant and a molecule modifier 1-Dodecanol into SiO₂-P support with a mean diameter of 60 μm. Adsorption and separation behavior of Sr(II) and Y(III) in the concentration of NaNO₃ in the aqueous feed solution ([H⁺]: 0.1, 0.5 M) onto adsorbent was investigated by batch and extraction chromatography methods.

I. INTRODUCTION

The nuclide partitioning of high-level liquid waste (HLLW) generated from the spent nuclear fuel reprocessing is thought to be a favorable method from the viewpoints of minimizing the long-term radiological risk and the radioactive waste accumulation. In our previous research, Sr(II) which is one of the heat-generating elements in HLLW could be separated from simulated HLLW using distilled water as eluent by extraction chromatography [1-2]. For the utilization of large amounts of separated ⁹⁰Sr from HLLW, ⁹⁰Y, a daughter nuclide of ⁹⁰Sr, is expected to be isolated from ⁹⁰Sr-⁹⁰Y group and used as radioactive medicines.

For the purpose, we have attempted to load the DtBuCH18C6 and a molecule modifier, 1-Dodecanol, into SiO₂-P by using its high immobilizing ability for the separation of Y(III) from the Sr(II)-Y(III) mixture solution. The present article deals with the preparation of a DtBuCH18C6 loaded adsorbent, characterization, adsorption equilibrium and kinetic modeling, and breakthrough and elution properties of Sr(II) for the extraction chromatography column.

II. EXPERIMENTAL

All batch experiments were performed at a phase ratio of 20 cm³·g⁻¹ and the resultant mixtures were shaken at 160 rpm. After the adsorbent was separated from the aqueous solution by filtration, the concentration of the metal ions in the solution was analyzed via inductively coupled plasma-atomic emission spectrometry (ICP-AES, Shimadzu ICPE-9000) and atomic absorption spectrophotometry (AAS). The distribution coefficients (K_d , cm³·g⁻¹), uptake ratios (R , %), and adsorbed amounts (Q , mg·g⁻¹) of the metal ions were calculated as follows:

$$K_d = \frac{(C_0 - C_e)}{C_e} \times \frac{V}{m} \quad (1) \quad R = \frac{(C_0 - C_t)}{C_0} \times 100 \quad (2) \quad Q = \frac{(C_0 - C_t) \times V}{1000 \times m} \quad (3)$$

where C_0 , C_e , and C_t are the concentrations of metal ion before adsorption, after adsorption and after reaching equilibrium in the liquid phase, respectively, in mg/L. m , and V are the weight of dry adsorbent in g, volume of the liquid phase in cm³, respectively.

The separation behavior of metal ions with adsorbent was evaluated by column experiment for chromatographic separation. 4.0 g dry adsorbent was packed into a pyrex glass column and the experiment was performed at 298 K by circulating thermostatic water through the jacket of the column.

III. RESULTS AND DISCUSSION

Figure 1 shows effect of the concentration of NaNO₃ of Sr(II) and Y(III) onto (DtBuCH18C6+Dodec)/SiO₂-P adsorbent, the concentration of NaNO₃ in the aqueous feed solution ([H⁺]: 0.1, 0.5 M (M = mol dm⁻³)) was varied. As shown in figure, (DtBuCH18C6+Dodec)/SiO₂-P adsorbent showed strong adsorption affinity to Sr(II) and weak adsorption for Y(III) in the nitrate concentration in the aqueous solution. On the other hand, the K_d of Sr(II) rapidly increased with the increase of nitrate concentration in the initial stage to 2 M and then decreased slowly over 2 M. The extraction chromatographic separation of Y(III) from Sr(II)-Y(III) mixture can be achieved by controlling the concentration of nitric acid.

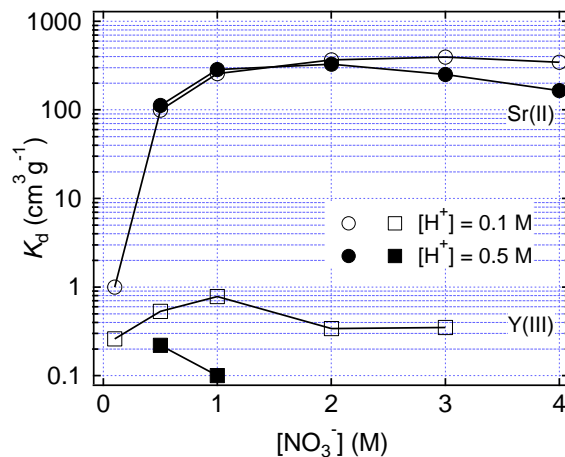


Fig. 1 Variation in the distribution coefficients (K_d) of Sr(II) and Y(III) as a function of the nitrate ion concentration in the aqueous solution. ([H⁺]: 0.1, 0.5 M; Time: 5 h; Temp.: 298 K)

Adsorption of Sr(II) reached equilibrium state within 1 h and this adsorption process could be expressed by Langmuir mode, and be governed by the chemisorption process. In extraction chromatography experiments, breakthrough curve of Sr(II) had S-shaped profile and more than 99.9% Sr(II) could be separated from Sr(II)-Y(III) mixture using distilled water as elution solution.

II. CONCLUSIONS

It was found that the adsorbent showed strong adsorption affinity to Sr(II) and weak adsorption ability to Y(III) under the experimental conditions. The K_d of Sr(II) rapidly increased with the increase of nitrate ion concentration in the initial stage to 2 M and then decreased slowly over 2 M. The adsorption equilibrium was attained within 3 h. Kinetic data were successfully modeled using pseudo-second-order kinetics. This adsorption process could be expressed by Langmuir mode, and be governed by the chemisorption process. Thermodynamic parameters were calculated using the Van't Hoff equation; these calculations indicated that the adsorption of Sr(II) was a spontaneous and exothermic process. (DtBuCH18C6+Dodec)/SiO₂-P adsorbent effectively adsorbed Sr(II) in the presence of 2 M nitrate ion solution in the column operation. The loaded Sr(II) ions were successfully eluted with an eluent of distilled water. The separation of Y(III) from Sr(II)-Y(III) mixture was achieved successfully.

ACKNOWLEDGMENTS

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