

Extraction of the ^{131}Cs from neutron irradiated barium carbonate by using calcium alginate beads

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1. Introduction

^{131}Cs , which has a higher average photon energy than the more established ^{125}I and ^{103}Pd isotopes, is a relatively new radioisotope used for low-dose-rate prostate brachytherapy [1]. ^{131}Cs is obtained by the decay of ^{131}Ba , which generated by $^{130}\text{Ba} (n,\gamma)^{131}\text{Ba}$ reaction. The extraction of ^{131}Cs essentially leads to the separation of some tens of nanograms of ^{131}Cs from very high amount of Ba species including ^{131}Ba . In scientific literature, some separation methods of alkali and alkaline earth metals were described [2-4]. However, some of the those methods are not recommendable from the view point of process step.

In this regard, we try to simplify the extraction process of ^{131}Cs as the absorbent removes only Ba^{2+} ion under continuous flow conditions in a fixed mode. The calcium alginate (CA) beads can selectively adsorb Ba^{2+} ion in target solution because of an excellent divalent cation ion exchanger [5].

2. Methods and Results

2.1 Synthesis of CA beads

To synthesize CA beads, 2.0 wt % sodium alginate solution was added dropwise into 50 mL of 20 wt % $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ solution under stirring through a 18 gauge flat-tipped needle at room temperature, as shown in Fig. 1. After incubation for 12 h, the beads were filtered out, washed thoroughly with deionized water, and dried in oven at 40 °C.

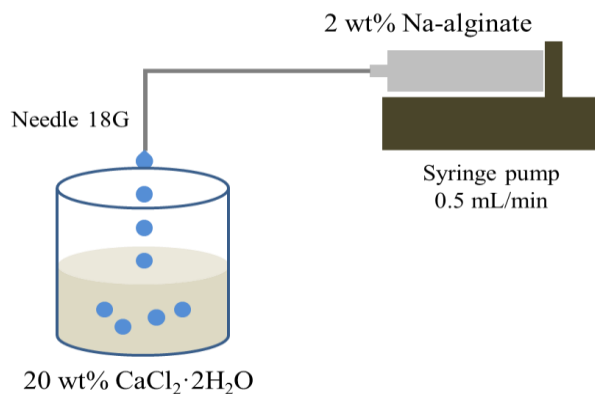


Fig. 1. Schematic diagram for preparing calcium alginate bead (CA).

2.2 The pH condition for the adsorption of Ba^{2+}

To determine the pH range at which maximum uptake of Ba^{2+} ions would take place on CA beads, 15 mL conical tubes each containing 0.2 g of CA beads were filled with 10 mL of 250 ppm Ba^{2+} solution. The pH was adjusted to values ranging from 1.0 to 6.5 (natural) by using dilute solution of HNO_3 . The tubes were shaken for 30 min to attain equilibrium. The Ba^{2+} concentrations of the liquid phases were determined by using Atomic Absorption Spectrophotometer (AAS).

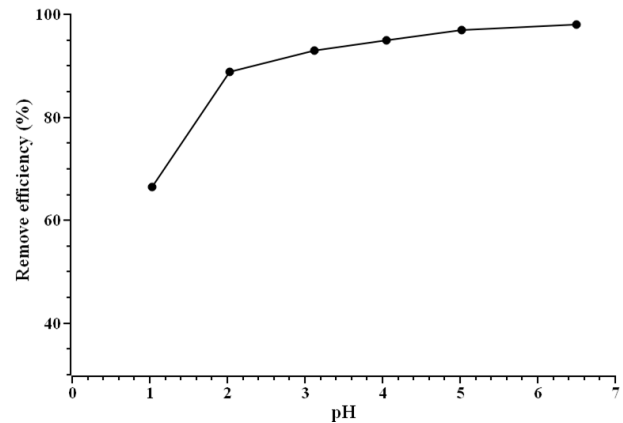


Fig. 2. Adsorption portion of Ba^{2+} ion on CA beads at different pH solution.

The adsorption curve of Ba^{2+} was showed a plateau from pH 2 to 6.5. At pH 6.5 the adsorption of Ba is maximum (99.5%), as shown in Fig. 2. Therefore, we confirm that the best adsorption pH condition of Ba^{2+} ion on CA beads is natural.

2.3 The flow rate for the adsorption of Ba^{2+} in fixed bed

Fixed bed adsorption test was conducted to evaluate the column performance for Ba^{2+} ions removal on CA beads. Experiments were performed by using the Supelco column of 1.0 cm inner diameter and 3.0 cm length packed with the prepared CA beads. The influent feed was supplied with the syringe pump in a vertical up flow. The influent solution was adjusted to 250 ppm Ba^{2+} in natural medium. Flow rate was controlled by 1.0, 3.0, and 5.0 mL min^{-1} so as to get steady state conditions in the column. Sampling of each effluents was done at every 5 mL intervals until their total volume become 45 mL. The results was shown in Fig. 3.

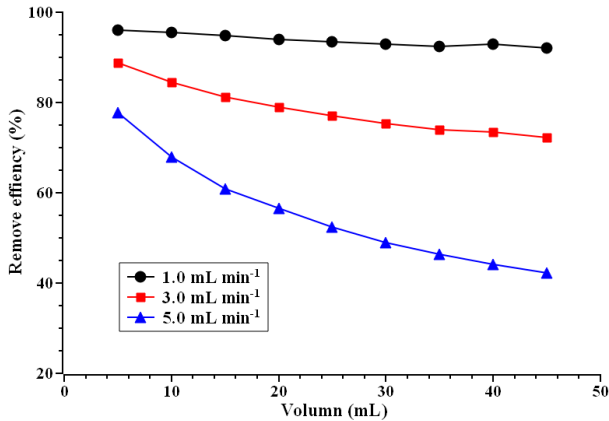


Fig. 3. Adsorption portion of Ba^{2+} ion on CA beads in fixed bed at different flow rate.

The adsorbed amount of Ba was considerably high and presented a plateau at 1.0 mL min^{-1} flow rate. In this test, the adsorption portions of first sample is maximum (96.5%). This value is similar to the result of batch mode under equilibrium condition. Also, the adsorption portion of final sample slightly decrease in comparison with other flow rates. These results suggest that 1.0 mL min^{-1} flow rate seems to be steady state condition for fixed bed mode.

2.4 Preparation of ^{131}Cs and ^{131}Ba mother solution

The 250 mg of $BaCO_3$ was sealed in quartz vessel and was located in aluminum block-containers. It was irradiated at a thermal neutron flux of $1.5 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ for 73 h at the Radioisotope Centre POLATOM. The irradiated sample was clearly dissolved by the 20.0 mL of 0.5 M HNO_3 . This solution was completely evaporated by IR Lamp. The recrystallized white powder was completely dissolved by the 10.0 mL of deionized water. This solution was used as the mother solution of ^{131}Cs and ^{131}Ba .

2.5 Fixed bed test in hot cell

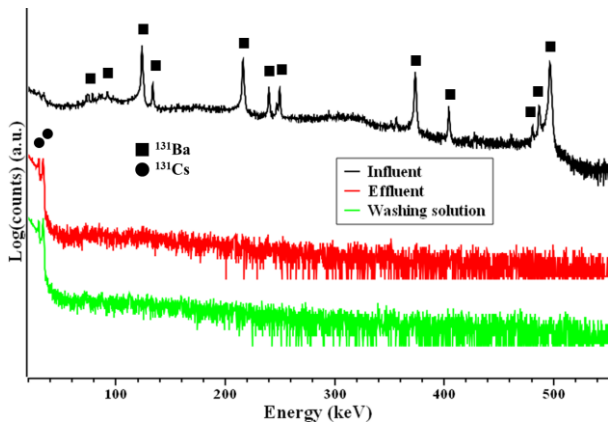


Fig. 4. MCA spectrum of influent, effluent, and washing solution.

The extraction of ^{131}Cs in mother solution was conducted by fixed bed mode. Test was performed by using stainless steel column of 1.0 cm inner diameter and 15.0 cm length packed with the prepared CA beads. The flow rate of influent was adjusted by the use of peristaltic pump under the flow rate of 1.0 mL min^{-1} . To match up the influent condition with cold test, The concentrations of Ba species was diluted by adding deionized water. The sampling was done at effluent and washing solution through deionized water. The ^{131}Ba and ^{131}Cs of each samples were determined by using HPGe gamma-ray spectroscopy (MCA).

Fig. 4 presents the MCA spectrum of influent, effluent, and washing solution through hot test. After adsorption process, the effluent disappear the spectra of ^{131}Ba . This result suggests that Ba species in influent was completely removed by CA beads. Additionally, washing solution only show the spectra of ^{131}Cs . From this result, we determine that the ^{131}Cs is able to extract to CA beads adsorbed Ba species under deionized water. Additionally, Ba species were stably adhered on CA beads.

3. Conclusions

An alternative production methodology of ^{131}Cs has been suggested for the separation of Cs-131 from its parent ^{131}Ba and other Ba species by using calcium alginate (CA) beads. From cold test of batch and fixed bed, we confirm the Ba^{2+} adsorption condition such as concentration, pH, and flow rate. Additionally, the results of hot test by using the diluted ^{131}Cs and ^{131}Ba mother solution show that ^{131}Cs was selectively extracted under deionized water and Ba species were well attached on CA beads. Based on these results, we believe that the fixed bed by using CA beads has potential as easy and simply method producing pure ^{131}Cs through more optimization.

REFERENCES

- [1] T. S. Kehwar, Use of Cesium-131radioactive seeds in prostate permanent implants. *Journal of Medical Physics*, Vol. 34, p.191, 2009.
- [2] S. Khujaev, A. Vasidov, E. A. Markelova, Extraction of the ^{131}Cs from neutron irradiated barium oxide under microwave radiation. *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 298, p. 435, 2013.
- [3] A. M. El-Kamash, Evaluation of zeolite A for the sorptive removal of Cs^+ and Sr^{2+} ions from aqueous solutions using batch and fixed bed column operations. *Journal of Hazardous Materials*, Vol 151, p. 432, 2008.
- [4] P. Vanura, E. Markrlik, Extraction of barium by using nitrobenzene solution of cesium dicarbollylcobaltate in the presence of 15-crown-5. *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 256, p. 373, 2003.
- [5] Y. Huang, Z. Wang, Preparation of composite aerogels based on sodium alginate, and its application in removal of Pb^{2+} and Cu^{2+} from water. *International Journal of Biological Macromolecules*, Vol. 107, p. 741, 2018.