

## High-throughput Preparation of Alginate Beads for the Separation of Barium from Cesium

In Won Choi <sup>a,b</sup>, Myunggoo Kang <sup>a</sup> and Seung-Kon Lee <sup>a\*</sup>

<sup>a</sup> Neutron and Radioisotope Application Research Division, Korea Atomic Energy Research Institute,  
111 Daedeok-daero 989 beon-gil, Yuseong-gu, Daejeon 34057, Republic of Korea

<sup>b</sup> Department of Physics, Chungnam National University,  
99 Daehak-ro 76 beon-gil, Yuseong-gu, Daejeon 34134, Republic of Korea

\*Corresponding author: seungkonlee@kaeri.re.kr

### 1. Introduction

Implementation of <sup>131</sup>Cs in clinical practice is considered as one of the most important innovations in brachytherapy for the past 20 years. The optimal combination of its half-life and radiation energy makes <sup>131</sup>Cs an attractive radioisotope for brachytherapy of malignant tumors. The list of organs include prostate gland, lungs, brain, mammary gland and etc. The half-life of <sup>131</sup>Cs is 9.7 days. It decays by electron capture emitting photon radiation in the X-ray range (29 keV). It can be produced by neutron irradiation of barium, followed by subsequent chemical separation from the 11.6day <sup>131</sup>Ba parent.

Sodium alginate is a natural hydrophilic polysaccharide derived isolated from marine brown algae.[1] It has been widely investigated in the field of drug delivery due to its biocompatible and biodegradable nature. It is an anionic polyelectrolyte with a backbone of (1-4) linked  $\beta$ -D-mannuronic acid (M units) and  $\alpha$ -L-guluronic acid (G units).

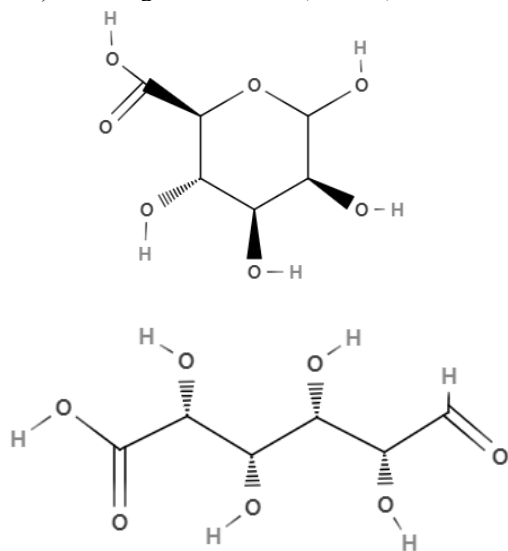


Fig. 1. Chemical structure of D-mannuronic acid (up) and L-guluronic acid (down).

The G-blocks of alginate are known to participate in intermolecular crosslinking with divalent calcium ions. Thus, the mechanical properties of alginate gels are enhanced by increasing the length of its G-block and molecular weight. Ca<sup>2+</sup> is most frequently employed for

alginate gel formation. Therefore, alginates can be considered as calcium-responsive polymers. [2]

Ca<sup>2+</sup> ions in the alginate gel can be exchanged by Ba<sup>2+</sup> ions due to the higher selectivity of Ba<sup>2+</sup>. Therefore, the alginate gel can be used for the separation of Ba<sup>2+</sup> ions and Cs<sup>+</sup> ions through the cation exchange. [3]

In this study, we proposed a co-current spray process using gas-liquid flow through co-axial nozzle for the high-throughput preparation of alginate microbeads. Additionally, co-current spray process gives better controllability on the bead size, compared to the conventional dripping method. Finally, adsorption behavior of the Ba and Cs ions was investigated through the batch experiments.

### 2. Experimental

#### 2.1. Preparation of Sodium Alginate Beads

Sodium alginate solution and co-flowing gas stream were injected through a co-axial needle to form a co-current flow. Flow rate of the sodium alginate solution was controlled by a syringe pump. And, the gas flow was controlled by pressure and flow meter. From the exit of the co-axial nozzle, sodium alginate solution breaks into series of droplets through the spray process. Then, the droplets were collected in the calcium chloride solution. Collected droplets were stirred for a sufficient time for gelation. [4]

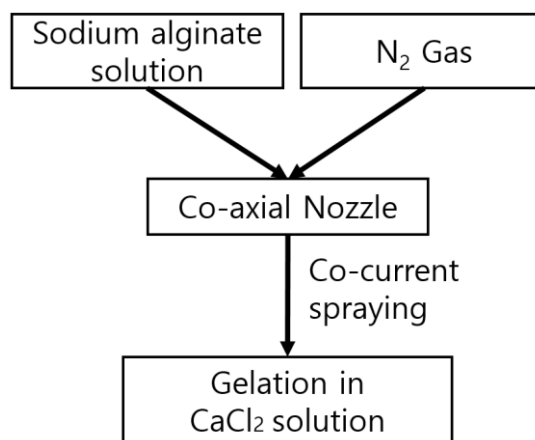


Fig. 2. Process scheme for the alginate bead production.

After gelation, the beads were collected and dried for storage. Drying is necessary to prevent the beads from contamination by bacteria.

## 2.2. Adsorption Experiments

Adsorption characteristics of alginate beads prepared from three different methods were compared. First beads were prepared by dripping of the sodium alginate solution without gas flow. Second and third beads were prepared by co-current spray process. The first and second beads were naturally dried. [5] And, the third beads were freeze dried. After drying, Ba and Cs adsorption experiments were performed in batch. Concentration of the remaining ions in the batch solution was measured by atomic adsorption spectroscopy.

## 3. Results and discussion

Prepared alginate beads through co-current spraying were presented in Fig. 3. Average diameter of the resulting beads was 220  $\mu\text{m}$ .

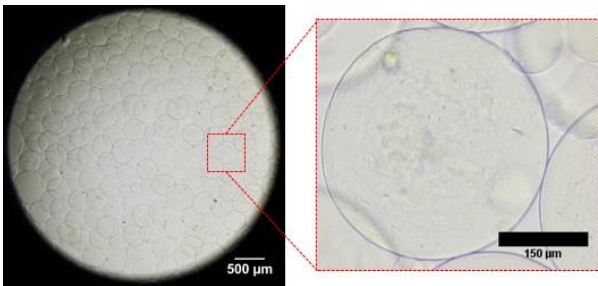


Fig. 3. Microscope images of prepared alginate beads through co-current spraying.

Fig. 4 and 5 show the atomic absorption spectroscopy results from barium and cesium adsorption tests. As we can see in the Fig. 4, alginate beads prepared by co-current spraying followed by freeze drying exhibited better barium adsorption performance.

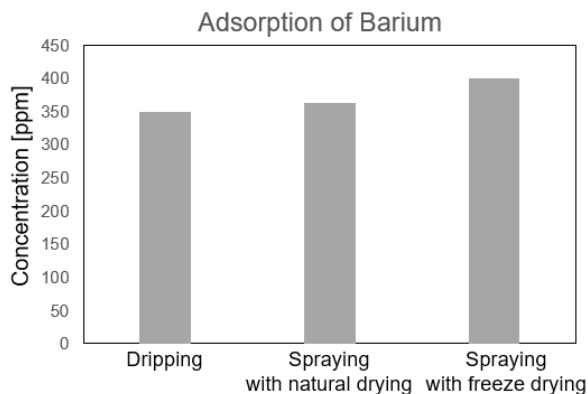


Fig. 4. Barium adsorption results for three different beads.

From the Fig. 5, the beads synthesized by spraying and natural drying showed minimum cesium adsorption. In conclusion, the beads made from co-current spray followed by natural drying exhibited high barium adsorption and low cesium adsorption, simultaneously. It is ideal characteristic for the selective separation of barium from the Ba/Cs mixture solution.

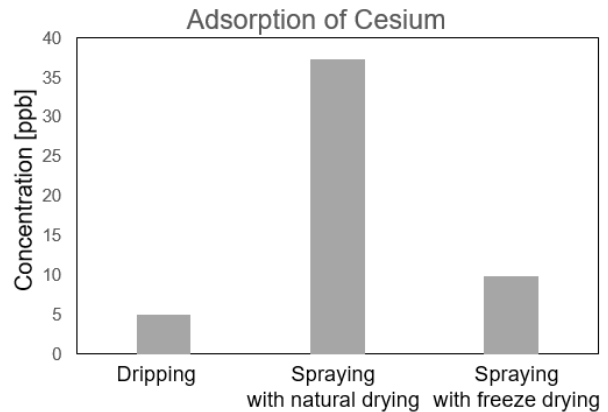


Fig. 5. Cesium adsorption results for three different beads.

## 4. Conclusions

In this study, we presented a co-current spraying process to prepare alginate microbeads for the effective separation of barium from cesium. The spray method exhibited higher throughput and controllability in size, in comparison with conventional dripping process. According to the drying process, adsorption yield of the alginate beads has been changed. The beads made from co-current spray followed by natural drying exhibited high Ba/Cs selectivity.

## REFERENCES

- [1] T. S. Kehwar, Use of Cesium-131 Radioactive Seeds in Prostate Permanent Implants, *Journal of Medical*, Vol. 34, No. 4, pp. 191-193, 2009.
- [2] S. K. Bajpai, S. Sharma, Investigation of Swelling/Degradation Behaviour of Alginate Beads Crosslinked with  $\text{Ca}^{2+}$  and  $\text{Ba}^{2+}$  Ions, *Reactive & Functional Polymers*, Vol. 59, pp. 129-140, 2004.
- [3] T. A. Davis, B. Volesky, A Review of the Biochemistry of Heavy Metal Biosorption by Brown Algae, *Water Research*, Vol. 37, pp. 4311-4330, 2003.
- [4] D. Rajmohan, D. Bellmer, Characterization of Spirulina-Alginate Beads Formed Using Ionic Gelation, *Hindawi International Journal of Food Science*, Vol. 2019, pp. 7, 2019.
- [5] N. Patel, D. Lalwani, S. Gollmer, E. Injeti, Y. Sari, J. Nesamony, Development and Evaluation of a Calcium Alginate Oral Ceftriaxone Sodium Formulation, Vol. 5, pp. 117-133, 2016.