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Alginate Beads-Based Cesium Adsorption in a Reservoir

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1. Introduction

Radioactive cesium based on its high radioactivity and relatively long half-life time (30.2 years) is a significant element of nuclear waste and nuclear fallout [1]. Cesium is water-soluble and behaves similarly to potassium and sodium in a biological behavior profile could be triggered negative health effects in humans [2]. Therefore, when absorbed into the body, it easily accumulates in organs and muscles. Since it takes a long time for water contaminated with radioactive cesium to be naturally purified due to the Fukushima nuclear accident, it is necessary to study quick and efficient cesium removal methods and adsorbents. Various matrixes such as several different polymers, zeolite-like materials, clay minerals, biomass, and metal oxides have already been reported [3]. Prussian blue is a material that selectively adsorbs representative radioactive cesium, and selectively adsorbs alkali cation. In this paper, we would like to investigate the adsorption degree and removal efficiency of cesium ions using beads of Prussian blue, which are known to have selective adsorption capacity for ^{137}Cs.

2. Material and Methods

To make an adsorption experiment using an adsorbent, a Prussian blue bead must be manufactured. In order to manufacture Prussian blue beads, Alginic acid sodium salt from brown algae (Sigma Aldrich, St. Louis, MO, USA), Iron (III) ferrocyanide (Sigma Aldrich, St. Louis, MO, USA), Calcium chloride, granular (SHOWA, Akasaka, Tokyo, Japan) were used in this study.

It has been commonly accepted that the exclusive abilities of PB to adsorb hydrated Cs^+ ions are caused by regular lattice spaces surrounded by cyanido-bridged metals. Cs^+ ions are trapped by simple physical adsorption in the regular lattice spaces of PB. Cs^+ ions are exclusively trapped by chemical adsorption via the hydrophilic lattice defect sites with proton-exchange from the coordination water [4].

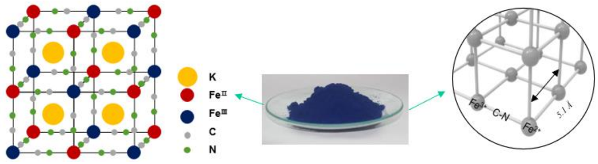

Fig. 1. Structure of ferric hexacyanoferrate

2.1 Alginate beads

To prepare a 4% sodium alginate solution and a 2.5% calcium chloride solution, 16 g of sodium alginate and 10 g of calcium chloride were added to 400 ml of DI water, respectively, and mixed overnight at 250 rpm using a magnetic bar. In addition, 10 g of iron ferrocyanide was added to synthesize 2.5% of Prussian blue alginate beads to prepare a solution.

Using 20 mL of a KOVAX-SYRINGE and a syringe pump (HARVARD APPARATUS 11 Plus), it was dropped one drop at a time into a 2.5% calcium chloride solution. At this time, the speed of the syringe pump was adjusted to 1.000 mL/min to adjust the sphericity of the bead. The bead thus made is washed and dried at room temperature as shown in Fig. 2. (a). Figure 2. (b) shows the size of one bead and is about 3 mm.

(a) (b)

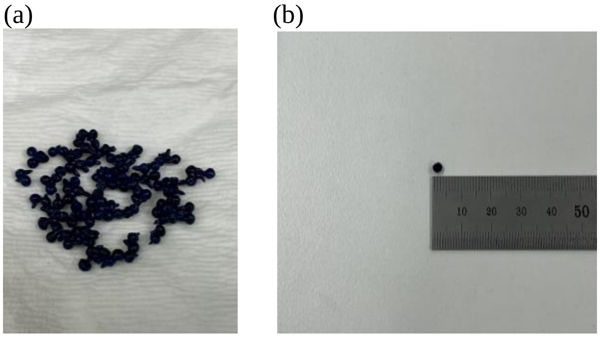

Fig. 2. (a) Prussian blue beads room-temperature drying, (b) Size of a bead

2.2 Cesium adsorption

An experiment was conducted to measure the adsorption amount of Standard Cesium over time at the same concentration of 5 ppm. 128 g of PB beads was put into a reservoir and 250 mL of a 5 ppm Cs solution was administered to collect a total of 7 samples for 0, 1, 3, 5, 7, 10, and 15 minutes. Samples collected are analyzed through ICP-MS.

Fig. 3. Prussian blue beads in a reservoir

3. Results

After cesium adsorption, the value of the concentration of cesium from Prussian blue beads in a reservoir by time scale was analyzed through ICP-MS. Based on the analyzed values, the cesium removal efficiency is shown in Figure 4.

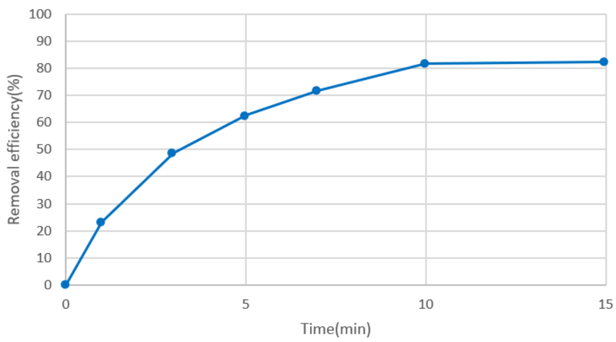

Fig. 4. Cesium removal efficiency

Considering that the concentration of remaining cesium is 0.798 ppm at 10 minutes and 0.767 ppm at 15 minutes, it is confirmed that most Prussian blue beads adsorb cesium within 10 minutes.

As shown in Figure 1, the cesium removal efficiency of about 82% was confirmed using Prussian blue beads.

When cesium adsorption reaches equilibrium, the coefficient representing the concentration ratio of substances present in each of the two media is called the distribution coefficient, and accordingly, the distribution coefficient (K_d) equation by experiment is as follows.

$$\frac{V_w (C_0 - C_i)}{M_{bead} C_i} \quad (1)$$

V_w is volume of water, C_0 is initial bulk concentration of contaminant, M_{bead} is mass of alginate bead from Prussian blue, and C_i is bulk concentration in solution remaining after sorption.

Through the adsorption experiment of Prussian blue beads, 518 mL per 1 g of Prussian blue was adsorbed. In addition, most Cs are believed to have been adsorbed in a short time (10 minutes), and it was confirmed that the Cs removal efficiency was 82% when the cesium solution added to the Prussian blue bead was shaken 5-6 times and left for about 10 minutes.

4. Conclusions

The adsorption experiment of Prussian blue beads showed that 518 mL per 1 g of Prussian blue was adsorbed, and most of the Cs were adsorbed in a short time. In addition, it was confirmed that the Cs removal efficiency of Prussian blue beads was 82%. Using alginate beads impregnated with Prussian blue like this will quickly adsorb radioactive cesium to help the environment and living things. As a result, alginate beads from Prussian blue show reinforcement properties compared to alginate beads gelled by traditional methods using Ca^{2+}. In the system, we designed a meaning method for purification of radioactive cesium contaminated seawater around nuclear power plants and/or after nuclear accidents. Therefore, this study is expected to provide insight into the fundamental structure-characteristic relationship of cesium removal and lay the foundation for various practical applications.

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