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Rice straw-based biochar beads for the removal of radioactive strontium from aqueous solution

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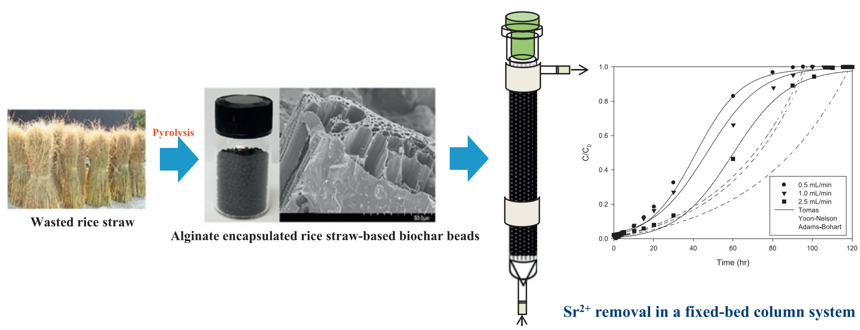
HIGHLIGHTS

- Rice straw-based biochar (RSBC) powder and beads were synthesized and characterized.
- Negatively charged RSBC beads showed a large surface area with high micro-porosity.
- The strontium adsorption capacity was high even in the presence of competing ions.
- A fixed-bed column reactor packed with RSBC beads was used for strontium removal.
- Both the Thomas and Yoon–Nelson models gave the best fit to the experimental data.

GRAPHICAL ABSTRACT



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ABSTRACT

Biochars from agricultural residues have recently attracted significant attention as adsorbents for purifying contaminated water and wastewater. In this study, the removal of strontium from aqueous solutions was investigated using rice straw-based biochar (RSBC) beads in both batch and continuous fixed-bed column systems. The RSBC beads had negatively charged surfaces and exhibited a large surface area (71.53 m^2/g) with high micro-porosity. The synthesized beads showed a maximum adsorption capacity of 175.95 mg/g at an initial strontium concentration of 10 g/L at 35 °C and pH 7. Furthermore, they showed a good selectivity toward strontium ions in the presence of competing ions such as Al^{3+}, Mg^{2+}, and K^{+}. The effects of different operating conditions like flow rate and initial strontium concentration were investigated in the fixed–bed column reactor. The Thomas, Adams–Bohart, and Yoon–Nelson models were applied to the experimental data to predict the breakthrough curves using non-linear regression. Both the Thomas and the Yoon–Nelson models were appropriate for describing entire breakthrough curves under different operating conditions. Overall, RSBC beads demonstrate great potential as efficient adsorbents for the treatment of wastewater polluted with strontium in a continuous operation mode.

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1. Introduction

Radioactive wastes generated during the operation and decommissioning of nuclear facilities require careful treatment and rigorous control due to hazardous radionuclides contained therein. Among various radionuclides (cesium, strontium, iodine, cobalt, plutonium, etc) radioactive strontium isotopes ^{90}Sr and ^{89}Sr, which have half-lives of 29 years and 50.53 days, respectively, are a major radioactive product of nuclear fission with high-energy beta emissions. The ^{90}Sr radionuclide is present at relatively low concentrations of 0.002–0.58 ppb in South Korea's low and intermediate level radioactive waste. However, the radioactive material in aqueous solution can be directly exposed to the human body at a higher concentration through the bioaccumulation in food chains or drinking water (Casacuberta et al., 2013; Hong et al., 2016; O'Donnell et al., 2016). Thus, developing an effective and economic waste management system for selectively removing strontium ions even at these low concentrations from aqueous solutions has recently received considerable attention. This work aims to develop such a system in order to eventually minimize the volume or weight of radioactive wastewater.

Various physicochemical methods for removing strontium ions from radioactive wastewater have been developed such as solvent extraction, separation membrane, evaporation, coagulation/sedimentation, electrodialysis, and adsorption. Among those available technologies, adsorption is a simple and economical technique, as it does not require further treatment after strontium ions are removed from the matrix. Several adsorbents such as zeolites, crystalline silicotitanate, and polyacrylonitrile have been developed for strontium ion removal from groundwater, seawater, and nuclear waste solutions. However, large-scale application of these materials is still limited due to their high cost, low strontium affinity, and potential risk of secondary contamination (Jang and Lee, 2016).

Biochar is the carbon-rich product obtained on heating biomass in a closed container with little or no available air (Lehmann and Joseph, 2015). Biomass for biochar production can be sourced from many abundant lignocellulosic wastes, including rice straw, spent mushroom substrate, and paprika plant wastes. Asia alone is responsible for ~90% of the 731 million tons of rice straw waste generated globally each year (Elumalai et al., 2016). In addition, about 30 million tons of spent mushroom substrate waste was generated worldwide in 2004, with South Korea accounting for about 1.7 million tons (Kwak et al., 2008). These wastes are conventionally burned or utilized as agricultural compost, giving rise to undesirable environmental implications and consequently very stringent environmental regulations. Therefore, biochar production from these waste biomasses employing pyrolysis has shown much promise. Several studies spanning the fields of agriculture and environment have been conducted on/with biochar owing to its uniquely abundant surface functional groups, porosity, surface area, sustainability, and attractive economic feasibilities (Mohan et al., 2014). Recently, biochar was shown as a potential adsorbent in the treatment of pollutants and for water purification. However, it is hard to retrieve or separate biochar powder from aqueous solutions after treatment because of their high dispersion property and small particle size. Alginate is a natural anionic polymer which is typically separated from marine algae. It has an affinity for multivalent cations and can be used for the entrapment of biochars, leading to an easy separation of the used adsorbents after the decontamination process. The alginate encapsulated biochar beads can be easily packed in a fixed-bed column reactor.

In this study, alginate encapsulated rice straw-based biochar (RSBC) beads were used to separate strontium ions from aqueous solutions. The equilibrium parameters, kinetics, and isotherms of the adsorption of strontium on the RSBC beads were studied. The effect of operational parameters such as flow rate and initial strontium concentration on the removal of strontium in a fixed-bed column reactor was also investigated. In addition, the Thomas, Adams–Bohart, and Yoon–Nelson models were applied to the experimental data to predict the breakthrough curves using non-linear regression.

2. Materials and methods

2.1. Materials

The biomass components derived from rice straw, paprika plant, and spent mushroom substrate were collected from Daejeon, South Korea. Activated carbon powder was purchased from the Duksan Pure Chemicals Co., Ltd., South Korea. Strontium nitrate ($Sr(NO_3)_2$), sodium alginate ($C_6H_9NaO_7$), calcium chloride ($CaCl_2$), sodium nitrate ($NaNO_3$), calcium nitrate tetrahydrate ($Ca(NO_3)_2 \cdot 4H_2O$), magnesium nitrate ($MgNO_3$), and aluminum nitrate ($Al(NO_3)_3$) were purchased from Sigma-Aldrich.

2.2. Preparation of biochar powder and beads

Three different biochars were derived from rice straw, paprika plant, and spent mushroom substrate under identical pyrolysis conditions and were named as RSBC, PPBC, and SMSBC, respectively. Each biomass feedstock was pyrolyzed using a fixed-bed continuous-stirred reactor setup, which involved a diesel-powered combustion unit around the reactor with a secondary reaction syngas chamber fitted with a cold-water circulation unit for controlling the temperature of the reactor and the exhaust gases for efficient energy utilization. A robust stainless-steel reactor chamber was charged with 20 kg of the feedstock and tightly capped in a screw-driven-mixer enclosing unit to prevent oxygen from entering the system during pyrolysis. The system temperature was increased steadily up to 450 °C, sustained for 1 h, and then immediately allowed to cool to room temperature with the entire operation lasting for ~5 h. The biochars produced were collected after an automated discharge from the reactor by means of a screw-shaft mechanism. The as-produced biochars were ground and sieved to pass particles of size 710 μm but retained on a 300-μm-sized sieve.

During preliminary experiments, RSBC powder showed superior strontium adsorption performance compared to the other biochar powders (Fig. 1). Based on these results, only alginate-encapsulated RSBC beads were prepared using the drop method. A mixture of RSBC powder (3 g) and sodium alginate (0.5% w/v) was dropped into a solution of $CaCl_2$ (2% w/v) using a syringe to form the beads and they were stirred

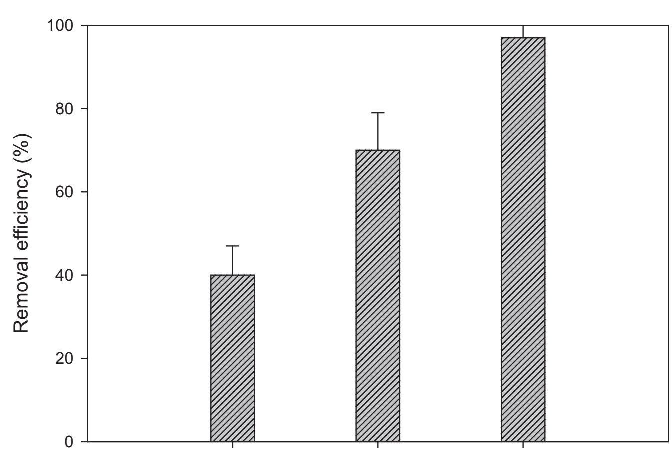

Fig. 1. Adsorption capacity of different biochar powders for strontium (conditions: weight of adsorbent = 1 g, initial strontium concentration = 1 mg/L, shaking time = 24 h, shaking speed = 150 rpm).