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Adsorptive removal of cesium from aqueous solution using oxidized bamboo charcoal

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ABSTRACT

In this study, the air oxidized bamboo charcoal (BC) was investigated for cesium (Cs) adsorption from aqueous solution. The physicochemical properties of the adsorbent were evaluated systematically using the different techniques including BET, FESEM, FTIR, XPS and also the pHpzc value. Batch adsorption experiments were conducted to determine the effect of contact time, solution pH, initial Cs concentrations, temperature and also the presence of competitive ions on adsorption. The adsorption kinetic parameters confirmed the better fitting of pseudo-second order kinetic model. The isotherm data could be well described by the Langmuir isotherm model and the maximum monolayer adsorption capacity was 55.25 mg g^{-1}. The high specific surface area and the porous structure with some acidic functional groups on the surface were obviously responsible for high Cs adsorption onto oxidized-BC. Thermodynamic parameters such as standard enthalpy, entropy, and Gibbs free energy were evaluated and it had been found that the adsorption process was favorable, spontaneous and endothermic in nature. In the competitive ions study, the presence of Na and K with their concentrations up to 12 mM did not strongly affect the removal of Cs by oxidized-BC. Therefore, the experimental results suggested that the oxidized-BC could be used as an effective adsorbent for significant Cs removal from aqueous solution considering the high adsorption capacity, short adsorption time and selective removal of Cs ions.

1. Introduction

In 2011, the massive nuclear power plant accident at Fukushima Daiichi in Japan was the second worst nuclear accident in the history of nuclear power generation and infamous for its devastating effect on the biological environment in the surrounding area. A Large amount of radioactive materials were released into the atmosphere, as a result of which the surface water and soil particles were severely contaminated. Among the several nuclides, especially cesium (^{137}Cs) released due to that nuclear accident, is considered one of the precarious elements of the environment. Moreover, the extended half-life (30.4 years), high solubility in aquatic media and transportability through the food chain has made it much hazardous for the human health. Cs can be easily assimilated into terrestrial and aquatic creatures because of its chemical properties identical to that of potassium (K) [1–3]. As a result, it can easily enter into the human body and causes thyroid cancer through potentially irradiating in living tissues [4]. Therefore, proper treatment and management are critically important before discharging the radioactive Cs to the environment.

The separation and removal of ^{137}Cs from the radioactive wastewater are commonly involved with the several approaches such as

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evaporation, solvent extraction, co-precipitation, membrane filtration, ion-exchange, and adsorption [5–7]. However, the use of the large quantity of chemicals and the associated cost, inefficient removal process, high energy consumption, problems related to the separated Cs disposal and management are considered as the fundamental drawbacks of these processes [8]. Particularly, the industrial scale application of the solvent extraction process is restricted due to the costly arrangements [9]. Moreover, in the case of ion exchange process, the performance of inorganic ion exchangers are generally superior compared to the organic ion exchangers because of their thermal stability and the specific ion selectivity [10,11]. Several ion exchangers have been developed for radioactive cesium treatment in the past few years. These included zeolites, metal hexacyanoferrate, polyphenol enrich ion exchange resins and different hybrid adsorbents [12,13]. Thus from the economical point of view, the development of the cost-effective Cs removal process still deserves strong attention.

Adsorption is one of the attractive methods for the removal of Cs in terms of the excellent removal performance in a safe manner, operation simplicity, suitability for low contaminated wastewater, availability of different low-cost adsorbents and scope of batch and continuous application processes [14]. Along with the inorganic, organic and hybrid ion exchangers, naturally available clay minerals (zeolites, bentonite, and montmorillonite) and adsorbents developed from waste materials (blast furnace slag and coal fly ash) have been used as low- cost materials for Cs removal. However, the Cs adsorption is seriously obstructed due to the competitive interaction with other monovalent cations; especially sodium and potassium [15,16]. Very few studies have been conducted for the removal of Cs using activated carbon in the previous years and the obtained results are not up to the satisfactory level. For example, Caccin et al. studied the Cs adsorption capacity of coconut shell activated carbon and found very poor adsorption capacity of this adsorbent (less than 1 mg g^{-1}) [17]. Almond shell was used as activated carbon for Cs sorption by Alarifi et al. and the removal capacity was 12.63 mg g^{-1} [18]. The commercially available activated carbon was employed for the practical radioactive cesium removal operation as reported by Vanderheyden et al. and they removed only 28% of Cs (adsorption capacity was 8.5 μg g^{-1}) from radioactive wastewater [19]. Therefore, in the recent years, a considerable attention has been employed to develop the low-cost activated carbon based adsorbents for the efficient and cost-effective removal of Cs from wastewater.

Bamboo is a renewable bioresource and abundantly found in different geographic areas of the world. Bamboo charcoal (BC) is one of the promising adsorbents enriched with microporous structure and higher surface area. The surface area of bamboo charcoal is generally 3–10 times higher than compared to the wood charcoal. The surface porous structure and heterogeneity of charcoal materials can be improved by modification process which has significant effects on adsorption performance. The physical and chemical modification of bamboo charcoal can enhance its affinity to adsorbate ions which is evidenced by the earlier studies [20]. However, the application of bamboo charcoal for cesium removal is not still widely explored. In the recent years, very few studies have been conducted for adsorption of heavy metals using bamboo charcoal [21,22].

In this work, we investigated the Cs removal performance of air oxidized-BC under the different experimental conditions such as contact time, initial Cs solution pH, initial Cs concentration, the presence of competitive ions and temperature in details. Moreover, adsorption kinetics, adsorption isotherm and thermodynamic studies were also conducted to understand the adsorption behavior.

2. Materials and methods

2.1. Materials

Non-radioactive cesium chloride (CsCl; FW:168.36 g) was purchased from Wako Pure Chemical Industries Ltd, Japan for using in this study as substitute of ^{137}Cs due to the similar chemical characteristics. Other chemicals such as sodium chloride (NaCl), potassium chloride (KCl), sodium hydroxide (NaOH) and hydrochloric acid (HCl) were obtained from the same supplier. All chemicals and reagents were of analytical grade purity. Ultrapure water (Elix Millipore, Japan) was used throughout the experiments. All working solutions were freshly prepared before using.

2.2. Preparation of adsorbent

Mosso bamboo (*Phyllostachys pubescens*) was collected from the forest near the Ito campus of Kyushu University in west Japan. Bamboo was cut into small pieces and dried properly. Before carbonization process, bamboo chips were washed with boiled water in order to remove impurities and dried at 110 °C for overnight. Carbonization was performed at 500 °C for 3 h with a heating rate of 5 °C/min in a muffle furnace (Yamato FP32, Japan) under nitrogen (N_2) atmosphere (0.15 m^3 min^{-1}). Carbonized bamboo chips were crushed by a ball mills machine (Mixer Mill MM 400, RETSCH, Japan) to make powder. The uniform size (75 μm) of the BC particles was confirmed by sieving.

2.3. Air oxidation

For air oxidation, the BC was placed into a muffle furnace and heated at 300 °C for 2 h under compressed air. After cooling down at room temperature into the muffle furnace, the BC was collected and preserved for experimental uses. The obtained BC was identified as oxidized-BC in this study.

2.4. Characterization of oxidized-BC

The porous properties of the adsorbent were determined by N_2 adsorption-desorption isotherms at −196 °C (77 K) by using