



Preparation of natural cation exchanger from persimmon waste and its application for the removal of cesium from water



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HIGHLIGHTS

- Novel bio-sorbent for cesium (Cs⁺) ion removal was fabricated from persimmon waste.
- The Cs⁺ was completely adsorbed onto cross-linked persimmon waste in fixed bed system.
- Maximum uptake capacity of persimmon waste was drastically improved after cross-linking.
- Mechanism of Cs⁺ adsorption is inferred to be ion exchange between proton and Cs⁺.
- 92% Toxic volume of Cs⁺ loaded cross-linked persimmon waste was reduced by simple incineration process.

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ABSTRACT

A novel bioadsorbent prepared from persimmon waste via cross-linking with concentrated sulphuric acid was investigated for its adsorption behavior towards Cs⁺. The adsorbent characterization included functional group analysis, elemental analysis, total organic carbon measurements and thermo-gravimetric analysis. The maximum adsorption capacity of PW (0.11 mmol/g) for Cs⁺ was drastically improved after cross-linking (0.76 mmol/g), furthermore the uptake capacity of the resulting cross-linked adsorbent appeared to increase with increasing temperature. The observed positive value of enthalpy change and negative value of Gibbs free energy change suggest that adsorption of Cs⁺ by this adsorbent is endothermic and spontaneous in nature. The mutual separation of Cs⁺ from Na⁺ can be successfully achieved with quantitative removal of Cs⁺ within 105 h using a packed column of the adsorbent. Thus, it can be expected that the CPW investigated in this study will contribute to the remediation of water polluted by radioactive cesium.

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1. Introduction

The existence of radioactive elements in water is of growing concern because of their toxic and carcinogenic effects on living organisms. The release of large amounts of radioactive elements into aquatic environment has occurred and the results have been confirmed as a direct threat to human health [1]. ¹³⁷Cs is one of such toxic radionuclides which had been generated during the nuclear fission reaction of ²³⁵U in nuclear weapons testing and some disasters that have occurred at atomic power stations such as the Fukushima incident on March 11, 2011, in particular [2–4]. Due to the emission of highly penetrating hazardous radiation (γ rays and β rays), high fission yield, long half life (30 year) and high water solubility, ¹³⁷Cs can easily enter into human body and persist for long period of time where it can potentially irradiate the living tissue,

thus possibly leading to serious diseases such as cancer and leukemia [5–7]. Thus, decontamination of aquatic environments polluted with Cs⁺ is a subject which should be urgently resolved [8].

Several physiochemical methods such as evaporation, liquid–liquid extraction, chemical treatment, micro-filtration and membrane processes have proven to be effective for the removal of radionuclide including ¹³⁷Cs from water and nuclear effluents [9–13]. However, disadvantages such as incomplete removal, high reagent cost and considerable energy requirements, in addition to issues related to the disposal of a considerable quantity of toxic waste resulting from such processes have all significantly hindered further adoption of such methods. Ion exchangers such as Prussian blue, zeolite, metal hexacyanoferrate and polyphenol rich ion exchange resins are all effective in removing radioactive cesium from water [14,15]. However, due to their high cost, the development of more cost-effective remediation system is strongly desired [16].

In recent years, research interest has been focused on the uses of biomass waste for the treatment of water polluted with

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radionuclides like ^{137}Cs . Several bio-sorbents such as those derived from coir pith, *Azolla filiculoides*, marine algae, pine cone powder, *Ocimum basilicum*, walnut shells and rice hulls were identified as effective biomaterials for the removal Cs^+ from water [16–22]. Other studies reported that polyphenol rich ion exchange resins are also effective for the treatment of Cs^+ polluted water [15,23,24].

Persimmon tannin extract is produced by juicing astringent persimmon rich in persimmon tannin and has been extensively used for various purposes including; natural dyes, paints as well as historically in leather tanning in Eastern Asia. In the previous study, an adsorption gel was prepared from commercially available persimmon tannin (PT) extract, produced and marketed by Persimmon-Kaki Technology Development Co. Ltd., Jincheng, China, for effective removal of Cs^+ from aqueous medium [25]. The commercially available powdered persimmon extract are produced from astringent persimmon in a sequence of juicing by mechanical crushing, fermentation and lastly spray drying, taking some costs. During this process, a large amount of juice residue is generated as waste. Thus, the development of adsorbents from such biomass waste containing high concentration of retained persimmon tannin for the removal and recovery of hazardous radionuclide such as ^{137}Cs appears highly promising from the view point of material costs and environmental concerns.

In the present work, an attempt has been made to prepare a more economical and environmentally benign adsorbent derived from persimmon waste (PW) generated after the extraction of tannin as a potential adsorbent for the adsorption of Cs^+ from water.

2. Materials and methods

2.1. Chemicals and analysis

The reagent grade cesium chloride (CsCl) and sodium chloride (NaCl) used in this study were purchased from Wako Chemical Co. Ltd., Japan. The stock solution (1000 mg/l) of Cs^+ was prepared by dissolving 1.26 g of CsCl in 1000 ml of deionized water, whereas that of Na^+ was prepared by dissolving 2.52 g of NaCl in 1000 ml of deionized water. The glassware and sample bottles were first washed with distilled water then rinsed with deionized water. The pH of the solution was adjusted using a small volume of 0.1 M HCl or 0.1 M NaOH ($M = \text{mol/l}$) solution. The concentration of metal ions in the sample solution before and after adsorption was measured using an atomic absorption spectrophotometer (AAS, Shimadzu model AA6800). The total organic carbon leaked from feed material, PW, before and after the cross linking was measured using a Shimadzu model TOC-VHS total organic carbon analyzer, whereas the incineration behavior of the adsorbent before and after Cs^+ adsorption was carried out by means of thermogravimetric analysis using a Shimadzu model DTG-60H DTA-TG apparatus.

2.2. Preparation of cross-linked persimmon waste gel

The sample of persimmon waste employed in this study was kindly provided from TOMIYAMA Co. Ltd., Kyoto, Japan. To avoid the partial dissolution of the adsorbent in aqueous solutions, it was cross-linked using concentrated sulphuric acid in similar manner to the persimmon tannin extract, CPT gel [25]. The sample was first dried and crushed into fine powder, yielding about 15 g of which was mixed together with 30 ml of concentrated sulphuric acid (96%) in a 250 ml eggplant flask, followed by refluxing the mixture for 24 h at 373 K in oil bath to achieve cross-linking via condensation reaction between hydroxyl groups of the polyphenol compounds contained in the persimmon waste. After completion of the reaction, the black product thus obtained was washed with

deionized water several times until a pH of approx. 7 was obtained for the washing water, after which the mixture was then dried in a convection oven at 343 K for 24 h. The final product thus obtained was designated as cross-linked persimmon waste and abbreviated as CPW gel, hereafter, while the crushed and dried sample of persimmon waste, the feed material, is abbreviated as PW, hereafter.

2.3. Batch wise adsorption tests

Batch wise adsorption tests were carried out in order to investigate the effect of pH, contact time, concentration and temperature on the adsorption of Cs^+ by the CPW gel. Fifteen mg of the CPW gel was combined and mixed with 10 ml of the sample solution containing 0.1 mmol/l of Cs^+ ions in a 50 ml glass bottle at various pH ranging from 1 to 8. The mixture was shaken for 24 h at 303 K to attain equilibrium, after which the gel was separated from the solution by filtration. The concentrations of Cs^+ before and after the adsorption were measured using the atomic absorption spectrophotometer. Kinetic measurements were performed by varying contact times for a solid liquid ratio of 1 g/l. Isotherm studies were carried out at a solid/liquid ratio of 1.5 g/l, a pH of 5.7 and using varying concentrations (1–10 mmol/l) of Cs^+ solutions at different temperatures. The % adsorption and the adsorption capacity were calculated according to the following mass balance equations.

$$\% \text{ adsorption } (\% A) = \frac{(C_i - C_e)}{C_i} \times 100 \quad (1)$$

$$\text{Adsorption capacity } (q) = \frac{(C_i - C_e)}{W} \times V \quad (2)$$

where C_i and C_e (mmol/l) are the initial and final concentration of Cs^+ , respectively. W (g) is dry weight of the CPW gel and V (l) is volume of the metal solution.

2.4. Continuous adsorption test of Cs^+ using a fixed bed column

Dynamic adsorption tests for Cs^+ and Na^+ ions onto the CPW gel was carried out in fixed bed system using a binary solution containing 0.015 mmol/l of Cs^+ and 0.30 mmol/l of Na^+ ions, within a glass column of 20 cm length and 0.8 cm internal diameter. Before column packing, 0.15 g of the CPW gel was soaked in deionized water for 6 h to allow for complete swelling and to avoid air entrapment in the bed. Conditioning of the gel was then achieved by flowing a metal free solution at the same pH as the metal solution (pH = 5.7) through the column overnight. Afterwards, the above-mentioned metal solution was percolated through the column at a constant flow rate of 5.1 ml/h in an upward flow mode using an IWAKI model 100 N peristaltic pump. The effluent samples were collected at various time intervals using a Bio-Rad model-2100 automatic fraction collector. The concentrations of Cs^+ and Na^+ in the effluent samples were measured using an atomic absorption spectrophotometer similar to that used during batch wise tests.

2.5. Incineration tests of CPW before and after Cs^+ loading

In order to evaluate the incineration behavior, thermogravimetric analysis of the dried sample of CPW gel was carried out in a TGA analyzer, before and after the adsorption of Cs^+ . For this experiment, 10 mg of Cs^+ loaded CPW and non loaded CPW were heated from 30 to 900 °C at a heating rate of 10 °C/min to record the percentage of the remaining weight at varying temperatures for both samples.

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