Journal of Water Process Engineering 10 (2016) 9–19

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Adsorption kinetic model of alginate gel beads synthesized micro particle-prussian blue to remove cesium ions from water

Yoshihiro Mihara[a,*], Md. Tajuddin Sikder[b,c], Hideaki Yamagishi[c], Takahiro Sasaki[d], Masaaki Kurasaki[c], Shinji Itoh[a], Shunitz Tanaka[c]

[a] Part of Medicinal Chemistry, School of Pharmacy, Hokkaido Pharmaceutical University, Sapporo, Hokkaido 006-8590, Japan
[b] Department of Public Health and Informatics, Jahangirnagar University, Dhaka 1209, Bangladesh
[c] Division of Environmental Materials Science, Graduate School of Environmental Science, Hokkaido University, Sapporo, Hokkaido 0600810, Japan
[d] Applied Chemistry Research Unit College of Environmental Technology, Graduate School of Engineering, Muroran Institute of Technology, Muroran, Hokkaido 050-8585, Japan

ARTICLE INFO

Article history:
Received 21 July 2015
Received in revised form 3 December 2015
Accepted 1 January 2016
Available online 6 February 2016

Keywords:
Alginate gel beads
Prussian blue
Cesium ion
Adsorption
Kinetics

ABSTRACT

Prussian blue (PB) impregnated in alginate gel (AG) beads were synthesized in order to test their viability for removing cesium (Cs) ions from wastewater. The impregnation yields PB-AG(syn.) beads containing very fine crystals of PB incorporated into the AG beads. The interaction between PB-AG(syn.) beads and Cs ions was investigated by adsorption experiments and analyzed using Langmuir and adsorption-kinetic models. The adsorption of Cs ions onto PB-AG(syn.) beads reached equilibrium after 12 h. The maximum adsorption capacities of the PB-AG(syn.) beads were 0.557 mg bead^{-1} and the kinetic rate was found to be $k_1 = 1.72 \times 10^{-1}$ min^{-1} at pH 6.0 for the pseudo first order, and 4.65×10^{-1} bead^{-1} mg^{-1} min^{-1} at pH 6.0 for the pseudo second order. The effects of pH, organic acids and salts such as potassium and sodium ions onto the adsorption of Cs ions by PB-AG(syn.) beads were found insignificant and low. Consequently, the PB-AG(syn.) beads showed a high capacity for the removal of Cs ions from water.
© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Over the last century, radioactivity and its toxicity attained significant attention all over the world. In the Great East Japan Earthquake on March 11, 2011, radioactive species like ^{134}Cs and ^{137}Cs ions were discharged into the surrounding environment of Fukushima Prefecture and thus contaminated the water and soil. The most abundant one is ^{137}Cs radionuclide having potent to cause detrimental effects on environment especially, human health can be affected due to its life-long existence and high solubility [1–3]. There is high possibility of ^{137}Cs to enter in various environments and easily assimilated by terrestrial and aquatic organisms where it deposited in the soft tissues that lead to cancer. Therefore, it's important to develop remediation options to remove Cs ions from waste water. Several technologies are now-a-days practicing all over the world for the decontamination of radionuclide Cs from wastewater including solvent extraction, chemical precipitation, membrane process, coagulation, electrochemical and ion-exchange lack effectiveness and selectiveness [4–7]. An adsorbent having novel attributes e.g. high adsorption capacity, to provide as an adsorbent having excellent durability even under severe using high acid and salinity condition, and easy post treatment management is now a time demanding quest for ^{134}Cs and ^{137}Cs ions removal from wastewater.

Recently, several researchers demonstrated Prussian blue (PB) as an effective absorbent for removing radioactive Cs ions from wastewater [8–10]. The basic structure of PB lies within a three-dimensional network with a face-centered cubic structure consisting Fe(III) (coordinated to nitrogen) and Fe(II) (coordinated to carbon atom) ions linked via the bridging of cyanide ligands [11,12]. Most adsorbed Cs is introduced into the lattice of PB and trapped there. Therefore, the fine crystals of PB are essential in order to gain high capacity and to speed up the adsorption of cesium ions in PB. However, the microcrystalline structure of PB are not reusable, recoverable through filtration or centrifugation, produced huge waste and make another problem for the environment. Hence, a possible solution is to entrapping the fine crystals of PB in supporting materials for preparation of new class conjugate adsorbent [13]. For example, several studies on magnetic separation have been reported about the treatment methods for heavy metal ions and organic pollutants by using modified Fe_3O_4. In particular, magnetic nanoparticles, such as Fe_3O_4, have been

* Corresponding author. Fax: +81 116768632.
E-mail address: mihara-y@hokuyakudai.ac.jp (Y. Mihara).

http://dx.doi.org/10.1016/j.jwpe.2016.01.001
2214-7144/© 2016 Elsevier Ltd. All rights reserved.

extensively studied for their utility in environmental remediation applications, magnetically guided drug delivery, due to their unique superparamagnetic and biocompatible properties [10,11]. And also, alginate is a promising and versatile immobilizer to use PB directly in water for treatment, so a powerful immobilizing approach is needed for safe use of PB. Alginate is a hetero polysaccharide composed of 1,4-linked α-L-gluronic acid and β-D-mannuronic acid, exhibits an excellent carrier for constructing powerful solid state adsorbent materials to trap metal ions through its carboxyl groups [14,15]. Vipin et al. developed Ca-alginate gel beads encapsulating PB powder and investigated their use in the removal of cesium ions from water [16]. However, the high dispersion of the PB powder in the alginate gel beads and produced huge sludge have been made it as major hurdle in the large-scale use for Cs separation. The key adsorbing materials such as prussian blue (PB) particulate in alginate gel beads was synthesized and purified systematically. The physical and chemical characterization of the prepared samples were scientifically completed using various techniques such as Scanning Electron Microscope (SEM) [17], X-ray diffraction (XRD) measurement [16], Fourier Transform Infrared Spectroscopy (FTIR) [18], zeta potential and size distribution by total volume [19]. Then the micro-particles of PB was encapsulated in hydrogels using sodium alginate as the immobilizer and ferric ions (Fe^{3+}) as the cross-linker. In each case two types of beads were prepared; one was a PB particulates synthesized in alginate gel beads method, PB-AG(syn.) and another with PB powder just mixed in alginate gel method PB-AG(mix.). The bead preparation was further optimized. The newly developed adsorbent beads could withstand in extreme pH, temperature and ionic strength. The developed beads were further examined by means of SEM and Brunauer–Emmett–Teller (BET) analysis to understand the morphological and porous peculiarities [20]. A series of experiments were performed to analyze the practical applications of high performing adsorbent for the removal of cesium from aqueous solutions under batch conditions. Various issues such as characterization, adsorbents preparations, effects of pH, equilibrium, kinetics, foreign ions effect, organic acid effect, salts effect were discussed systematically in detail. Therefore, encapsulate of PB as micro crystals in the gel beads of alginate through a novel method was attempted here in this study. Hence, the adsorption of Cs ions onto the PB entrapped alginate gel beads (PB-AG) can attain a more stable and much faster adsorption equilibrium than that of previous attempts. In this connection, the PBAG(syn.) beads were successfully synthesized and their effectiveness were compared with other PB derivatives.

2. Materials and methods

2.1. Materials

Sodium alginate (300–400 cp) and calcium chloride were used for the preparation of the AG gel beads, and potassium ferrocyanide, $K_4Fe(CN)_6{\cdot}3H_2O$ and Iron(III) chloride were used for the preparation of the prussian blue. All those materials were obtained from Wako Pure Chemicals Co. (Osaka, Japan). Cesium chloride was used to prepare model solution of cesium ions. Additives including sodium chloride, potassium chloride, hydrochloric acid, oxalic acid, nitric acid, acetic acid, citric acid, and ammonium acetate were purchased from Wako Pure Chemicals Co. (Osaka, Japan) and were used to investigate the effects of pH and coexisting ions on the adsorption of cesium ions onto PB-AG beads.

2.2. Preparation of PB powder

To a solution of 36.5 mg of pottadium ferrocyanide decahydrate (in deionized water 10 mL) was slowly added 0.15 M ferric chloride hexahydrate in deionized water 200 mL with mechanical stirring. The solution was mixed overnight. The colloidal product was a mixture of soluble and insoluble PB, with a high level of sodium chloride. The PB was separated via centrifugation at 10,000 rpm for 5 minutes and the separated sample was washed three times with deionized water. Finally, the PB powder was allowed to dry at 80 °C for 12 h in an oven.

2.3. Preparation of PB-AG beads

Prussian blue impregnated in alginate gel beads have been prepared by two different method (PB-AG(syn.), and PB-AG(mix.)). One is a synthesis, PB-AG(syn.) method; First, 10 mL of 0.5–3.0 wt% alginate solution containing 36.5 mg of $K_4Fe(CN)_6$ was prepared. This solution was added drop wise from a burette into a 0.1 M calcium chloride solution 200 mL to form Ca^{2+}-AG beads containing $K_4Fe(CN)_6$. And then, Ca^{2+}-AG beads containing $K_4Fe(CN)_6$ were immersed in a 0.15 M Fe(III) chloride solution 200 mL for three hours. After that, the PB particulate was synthesized by the reaction of $Fe(CN)_6^{4-}$ in the gel when ferric ions diffused from the Ca^{2+}-AG beads. Calcium ions in Ca^{2+}-AG beads was exchanged with ferric ions to form Fe(III)AG beads. After that, these gel beads were filtered and washed with distilled water and designated as PBAG(syn.) beads.

Another one is mix, PB-AG(mix.) method; To a powder of prussian blue (50.5 mg) was added sodium alginate solution (10 mL, 10.5–3.0%, w/v) under constant stirring. The solution was kept under magnetic stirring for 20 min. PB-encapsulated beads (PB-AG(mix.)) were prepared by the drop-wise addition of the aqueous PB particle-alginate colloidal solution into a beaker containing 0.1 M calcium chloride solution 200 mL under magnetic stirring. The PB-AG(mix.) beads were stirred in the same solution about 3 h for maturation. Then, calcium ions in gel beads were exchanged with ferric ions to form Fe(III)AG beads for 3 h. Finally, they were then collected by filtration, washed three times with deionized water, and stored in calcium chloride solution (1%, w/v).

For measuring the amount of the PB powder in alginate gel beads measured the calculated difference between the weight of PB powder synthesized in 125 number of alginate gel beads and the weight of the same number of plane type of Fe-AG (without PB powder). On the basis of the amount of the PB powder, the entrapping of PB powder in the PB-AG(mix.) adjusted an same quantity of the PB-AG(syn.), whereby the characteristic of the adsorption result is corrected.

2.4. Characterization of PBAG beads

Further chemical and physical characterization of PB powder, PB-AG(syn.) and PB-AG(mix.) was completed. The cross-sectional morphology of the alginate gel beads was observed using an Inverted Metallurgical optical microscope (CK2, OLYMPUS, Japan). Scanning electron microscopy was performed on the surface and cross-sectional morphology of PB particles in alginate gel beads by using scanning electron microscope (SEM) measurements (JSM-7400F, JEOL, Japan). The crystal structure of the PB in the alginate gel beads was measured by X-ray powder diffraction (XRD), using a Japan Rigaku D/MAX-cA X-ray diffractometer equipped with Cu Kα (0.1541841 nm) radiation over a 2θ range of 10–70° and X-ray power of 40 Kv/20 mA at a scan. The surface area of PB powder, PB-AG(syn.), PB-AG(mix.) and Ca^{2+}-AG were measured by the Autosorb (YUASA, Japan). The FT-IR spectrum of PB powder, PB-AG(syn.) were acquired with the Spectrum 100 (PerkinElmer, USA) in the range of 4000–500 cm^{-1} by preparing the Attenuated Total Reflection (ATR). SEM, FT-IR and BET studies performed on the synthesized beads, which were freezedried for maintain the exact structure. The zeta potential and the size distribution analysis of PB

Download English Version:

https://daneshyari.com/en/article/232445

Download Persian Version:

https://daneshyari.com/article/232445

Daneshyari.com