Bioresource Technology 100 (2009) 2995–3002

Contents lists available at ScienceDirect

Bioresource Technology

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

Removal of persistent organic pollutants from micro-polluted drinking water by triolein embedded absorbent

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ARTICLE INFO

Article history:
Received 5 October 2008
Received in revised form 19 January 2009
Accepted 20 January 2009
Available online 25 February 2009

Keywords:
Persistent organic pollutants
Absorbent
Triolein
Water treatment

ABSTRACT

A new biomimetic absorbent, cellulose acetate (CA) embedded with triolein (CA-triolein), was prepared and applied for the removal of persistent organic pollutants (POPs) from micro-polluted aqueous solution. The comparison of CA-triolein, CA and granular activated carbon (GAC) for dieldrin removal was investigated. Results showed that CA-triolein absorbent gave a lowest residual concentration after 24 h although GAC had high removal rate in the first 4 h adsorption. Then the removal efficiency of mixed POPs (e.g. aldrin, dieldrin, endrin and heptachlor epoxide), absorption isotherm, absorbent regeneration and initial column experiments of CA-triolein were studied in detail. The linear absorption isotherm and the independent absorption in binary isotherm indicated that the selected POPs are mainly absorbed onto CA-triolein absorbent by a partition mechanism. The absorption constant, *K*, was closely related to the hydrophobic property of the compound. Thermodynamic calculations showed that the absorption was spontaneous, with a high affinity and the absorption was an endothermic reaction. Rinsing with hexane the CA-triolein absorbent can be regenerated after absorption of POPs. No significant decrease in the dieldrin removal efficiency was observed even when the absorption–regeneration process was repeated for five times. The results of initial column experiments showed that the CA-triolein absorbent did not reach the breakthrough point at a breakthrough empty-bed volume (BV) of 3200 when the influent concentration was 1–1.5 μg/L and the empty-bed contact time (EBCT) was 20 min.

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1. Introduction

Persistent organic pollutants (POPs) are characterized by pronounced persistence against chemical/biological degradation, high environmental mobility, strong tendency for bioaccumulation in human and animal tissues and significant impacts on human health and the environment even at extremely low concentrations. POPs have been detected in rivers, soils, the atmosphere and often even in milk or human tissues. In China, POPs could be detected from the surface water, such as the Guanting reservoir, the Yongding river and so on (Kang et al. 2003; Guo et al., 2008). Most POPs are lipophilic chemicals and they are present at low concentrations in water, usually varying between nanogram and picogram per liter and sometimes hundreds of nanograms per liter in micro-polluted surface waters (Zhang et al., 2002; Guo et al., 2008). Their low concentration makes them refractory to the conventional treatment of drinking water.

Adsorption has been widely used as an effective method for removing organic compounds from water. Various adsorbents including activated carbon (Wang and Liu, 2001; Bembnowska et al., 2003; Matsui et al., 2003), natural materials such as sugar cane bagasse, green coconut shells, chitin, and chitosan (Crisafully et al., 2008; Li et al., 2009), organoclays (Lee et al., 2004) and DNA hydrogel beads (Liu et al., 2005) have been reported for POPs removal. However, it is difficult to remove trace level POPs near the environmental levels by these adsorbents.

Even at trace concentrations in water, lipophilic organics can be accumulated by organisms. Many studies have been carried out on bioaccumulation in diverse organisms such as white whales (Andersen et al., 2001), marketable fish (Ahmed and Aly, 2004), white seabream (Ferreira et al., 2008) and mussels (Azza et al., 2004). Semipermeable membrane devices (SPMD), which are based on the diffusion of hydrophobic substances from water to membrane bags filled with lipophilic phases, are widely used for monitoring organic contaminants in water (Huckins et al., 1990; Frank, 2005). It has also been reported that triolein embedded cellulose acetate (CA) membrane could quickly and efficiently accumulate hydrophobic POPs from water (Xu et al., 2005; Ke et al., 2006). In the present study, a novel composite absorbent

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doi:10.1016/j.biortech.2009.01.026

containing the lipid triolein was designed using the concept of bioaccumulation and was used to remove trace level lipophilic organics from water. The absorbent was prepared by embedding triolein into CA spheres. Earlier preliminary study showed that triolein was effectively embedded into CA spheres, and the CA-triolein absorbent was stable in water. This new absorbent are promising for the removal of non-polar compounds.

The purpose of this paper is to study the absorption properties of POPs on CA-triolein absorbent. Aldrin, dieldrin, endrin and heptachlor epoxide, which were detected from the surface water frequently, were selected as the model pollutants. The removal efficiency of four POPs, the absorption isotherm and the reusability of CA-triolein absorbents were studied. Special attention was on the absorption capacity in fixed-bed columns as an indicator of the viability to future implementation of a full-scale process.

2. Methods

2.1. Absorbent preparation

CA-triolein absorbent was prepared according to the procedures in our previous work (Liu et al., 2007) and was briefly described here. Homogeneous solution of CA (Chemical Reagent Corporation, Shanghai, China) in acetone was prepared and certain amount of triolein (99% purity, Sigma, USA), water, $Mg(ClO_4)_2$ and Tween 80 were then added to the solution. The mixtures were stirred at 35 ± 0.5 °C for at least two days in order to assure uniform mixing. According to the preliminary experiments, the optimum content of the distilled water in casting solution, which was used as a pore forming agent was about 12–14% (w/w). Thus, viscous syrups were prepared and homogeneous phases were obtained. Then, the viscous syrup was added dropwise to a glass tube containing the liquid olefin (about 2/3 of the volume). The temperature ranges of the CA solution and liquid olefin were kept at 30–35 °C and 15–20 °C, respectively. During the dripping process of the viscous syrup through the air to the column of liquid olefin and through the action of surface tension, a white, spherical absorbent was formed which was collected at the bottom of the glass tube. Finally, composite absorbents were washed with distilled water to remove all soluble residues. The TOC of the leachate was determined using a Phoenix 8000 Total Organic Carbon Analyser (Tekmar–Dohrmann Co., USA). After changing the water up to 10 times over a three day period, the TOC level of the leachate was near that of distilled water. CA spheres were also prepared using the above method but without triolein. All of the absorbents were stored in distilled water before use.

2.2. Absorption experiment

2.2.1. Adsorbates

Four selected POPs, namely, aldrin, dieldrin, endrin and heptachlor epoxide, were used as target pollutants. The physicochemical properties of these four pollutants are presented in Table 1. These compounds had similar molecular weights but different hydrophobicities, as indicated by their aqueous solubilities and octanol-water partitioning coefficients (log K_{ow}). The log K_{ow} of aldrin is 5.663, the highest of the four POPs. POPs aqueous solutions were prepared by directly adding the POPs standard solutions to deionized water in sealed conical flasks. The mixtures were then agitated for 8 h using a horizontal shaker at 170 rpm and 25 °C to dissolve the organics in water homogeneously.

2.2.2. Removal test

Comparing with GAC: The effectiveness of the novel absorbent was compared with GAC using dieldrin as model pollutant. Sorption experiments were carried out in triplicate with three sorbents, e.g. CA, CA-triolein and GAC in batch mode. The initial concentration of dieldrin was 10 μg/L. One gram of sorbent was employed in contact with a solution of 200 mL at 170 rpm at 25 °C. Experiments were continued for 24 h. Samples were withdrawn at regular intervals for analysis.

POPs removal by CA and CA-triolein absorbents: POPs removal efficiency by CA and CA-triolein absorbent were examined as follows: One gram of absorbent (CA-triolein and CA) was brought into contact with a solution of 200 mL which was spiked with mixed POPs standard and each compound concentration was 1 μg/L. Samples were withdrawn at regular intervals (e.g., 0.5, 1, 1.5, 2, 4, 6, 8, 12 and 24 h) for analyses.

2.2.3. Absorption equilibrium studies

A series of 500 mL conical flasks were filled with 200 mL distilled water, which was spiked with single or binary POPs (e.g., aldrin, dieldrin, endrin or heptachlor epoxide) and the mixtures were supplied with 1 g CA-triolein absorbent or 1 g CA absorbent. The spiking concentrations of POPs in water ranged from 1 to 20 μg/L. The solutions were sealed in conical flasks and shaken on a horizontal shaker at 170 rpm, and the temperature was controlled at 25 °C or 40 °C by air bath. Samples for analyses were taken after 72 h contact time. Preliminary kinetic tests were carried out and equilibrium was assumed when no further changes in POPs uptake were observed after 72 h.

2.2.4. Reusability of CA-triolein absorbent

After examining the POPs absorptivity of CA and CA-triolein absorbents, the absorbents were rinsed with distilled water, wiped up with clean filter paper, and dialyzed in 10 mL of hexane for 48 h. The absorption process was as follows: 1000 mL of mixed solution of four POPs (the concentration of aldrin was 20 μg/L and the dieldrin, endrin and heptachlor epoxide was 100 μg/L) contacted with 0.5 g absorbent for two days by electrical magnetic blender at 25 °C in sealed conical flasks. Dialysis solutions were evaporated under a gentle stream of nitrogen and analysis by GC. Then, the removal efficiency of the four POPs was examined again following the same procedures as described above. This experiment was repeated five times.

2.2.5. Column experiments

Since one of the objectives of the present work was to simulate the POPs removal in a full-scale tower, mini-column experiments

Table 1
Linear absorption isotherm parameters for the four POPs by CA and CA-triolein absorbents (temperature 25 ± 0.5 °C, pH 6.8, absorbent: 1 g) along with related physicochemical properties.

Compound	Molecular weight	Water solubility (mg L^{-1} @ 25 °C)	log K_{ow}	$K_{CA\text{-triolein}}$ [a]	r^2	K_{CA} [b]	r^2
Aldrin	365	0.027	5.663(BrÁs et al. (1999))	21114	0.990	9199	0.991
Deldrin	381	0.195	5.48 (Mackay (1982))	18681	0.991	8312	0.983
Endrin	381	0.25	4.56 (Mackay (1982)), 4.63 (Xu et al. (2005))	14269	0.987	7280	0.989
Heptachlor epoxide	389.2	0.35	4.51(Xu et al. (2005))	13178	0.982	6624	0.988

[a] The slope of linear regression, equilibrium partition coefficient between CA-triolein absorbent and water.
[b] The slope of linear regression, equilibrium partition coefficient between CA absorbent and water.

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