



Phenyl-functionalized magnetic palm-based powdered activated carbon for the effective removal of selected pharmaceutical and endocrine-disruptive compounds



Kien Tiek Wong^a, Yeomin Yoon^b, Shane A. Snyder^c, Min Jang^{a, d, *}

^a Department of Civil Engineering, Faculty of Engineering, University of Malaya, Kuala Lumpur, 50603, Malaysia

^b Department of Civil and Environmental Engineering, University of South Carolina, Columbia, SC, 29208, USA

^c Chemical and Environmental Engineering, University of Arizona, Tucson, AZ, USA

^d Department of Environmental Engineering, Kwangwoon University, 447-1 Wolgye-Dong Nowon-Gu, Seoul, Republic of Korea

HIGHLIGHTS

- Magnetite film was coated on the surface of PPAC via hydrothermal co-precipitation.
- Phenyl-organosilane was successfully silanized on magnetite film of MPPAC.
- Unique features of MPPAC-TEPS cause enhanced adsorption rates and capacities of POPs.
- Thermal regeneration effectively desorb POPs to from MPPAC-TEPS.

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ABSTRACT

Triethoxyphenylsilane (TEPS)-functionalized magnetic palm-based powdered activated carbon (MPPAC-TEPS) was prepared and characterized using various spectroscopic methods, and then tested for the removal of bisphenol A, carbamazepine, ibuprofen and clofibrac acid. Magnetite film on MPPAC-TEPS was homogeneously coated on the outer surface of palm-based powdered activated carbon (PPAC) through a hydrothermal co-precipitation technique. Followed by silanization of phenyl-functionalized organosilane on MPPAC's magnetic film. As results, micro/mesopore surface area and volume increased without significant pore clogging and iron (Fe) dissolution under the acidic conditions was greatly decreased. The unique structural and chemical features of MPPAC-TEPS were found to be the main reasons for the enhanced adsorption rates and removal capacities of POPs. The presence of electrolytes and different pH values greatly affected the sorption efficiencies. The dominant sorption mechanism of POPs by MPPAC-TEPS was determined to be π - π interaction (physisorption), based on thermodynamic (ΔG°) and differential scanning calorimetry (DSC). Thermal regeneration at a low temperature (350 °C) was an effective method to desorb the retained POPs and enabled to reactivate MPPAC-TEPS with sustained sorption rates and capacities, whereas PPAC was largely exhausted. As a new type of sorbent for POPs, MPPAC-TEPS has operational advantages, such as magnetic separation and stable regeneration.

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1. Introduction

As compounds within the class of persistent organic pollutants (POPs), endocrine-disrupting compounds (EDCs) and

pharmaceutically active compounds (PhACs) have been problematic in wastewater effluent, surface water, and groundwater worldwide (Yoon et al., 2010; Jurado et al., 2012). Many research has shown the presence of POPs in $\text{ng} - \mu\text{g L}^{-1}$ has been detected in finished drinking water (Zuccato et al., 2000; Stackelberg et al., 2004; Bao et al., 2012). Moreover, enormous concerns have been raised about removing these emerging organic micropollutants (Weyer and Riley, 2001; Snyder et al., 2003). Although present at trace-level, potential adverse effects was shown with continuous exposure towards human (Pomati et al., 2006) and the aquatic

* Corresponding author. Department of Civil Engineering, Faculty of Engineering, University of Malaya, Kuala Lumpur, 50603, Malaysia.

E-mail addresses: minjang@um.edu.my, minjang@kw.ac.kr, heejaejang@gmail.com (M. Jang).

environment (Watts et al., 2003; Kidd et al., 2007). The main reasons for PhACs such as clofibrac acid (CA), carbamazepine (CBZ), and ibuprofen (IBP) were commonly detected due to over usage of drugs and ineffective removal by conventional wastewater treatment plants (Ellis, 2006; Kim et al., 2007). One phenolic EDC, bisphenol A (BPA) is an important monomer used widely in the production of polycarbonate plastics, epoxy resins, and plastic containers (Ahmad et al., 2005), from which BPA can readily be leached (Desbrow et al., 1998; Laganà et al., 2004). This estrogen-mimicking compound poses negative health risks and may be carcinogenic to humans and animals by interfering with natural hormone receptors.

Numerous physicochemical techniques have been evaluated for treating secondary or tertiary effluents from water bodies containing PhACs and EDCs. Direct oxidation, including ultraviolet (UV) and UV coupled with hydrogen peroxide (Yuan et al., 2009) or the Fenton reaction (Sirtori et al., 2009) were found to achieve satisfactory removal. However, not only does a technique need to be inexpensive and simple, because of the huge volumes of water to be treated, concerns about any toxic by-products released from an oxidation process should also be eliminated. Compared with other treatments, adsorption on activated carbon (AC) is cheap, only requires simple equipment, and does not produce toxic by-products. Among sorption materials, especially, waste carbonaceous material-based AC has been studied extensively, and it significantly reduces production costs (Kang et al., 2013; Ozbay and Yargic, 2015; Saygılı et al., 2015).

To address the water purification problems outlined above, we prepared triethoxyphenylsilane (TEPS) functionalized magnetized palm-based powdered AC (MPPAC-TEPS) and studied its potential application in EDCs/PhACs removal. In detail, we developed a unique synthesis route to prepare magnetized PPAC (MPPAC) through coating magnetite film mostly on the outer surface of PPAC (Wong et al., 2015), so that the micropores were still available for adsorbing organic pollutants. However, magnetite in the medium may be released when exposed to a highly reducing environment. Additionally, the initial sorption rates and sorption capacities of organic pollutants could be reduced by magnetite loaded on the outer surface of the PPAC. Considering these issues, we sought to modify MPPAC by coating the magnetite film with phenyl-functionalized organosilane. Hypothetically, magnetite leaching could be prevented, and the sorption rate and capacity towards organic pollutants could increase via such an organosilane coating.

2. Experimental

2.1. Materials

Commercial PPAC (Bravo Green Sdn. Bhd., Malaysia, 100 × 200 mesh), activated by basic steam, was washed with 0.01 M HCl to remove metal impurities. Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), potassium nitrate (KNO_3), methanol (HPLC grade) and sodium chloride (NaCl) were purchased from R&M Chemical. Adsorbates (BPA, CBZ, CA, and IBP), reactants for the Boehm titration and triethoxyphenylsilane (TEPS; 99.9% purity) were obtained from Sigma–Aldrich. Physical and chemical properties of selected POPs are summarized in Table S1 in Electronic Supporting Information (ESI).

2.2. Preparation of magnetic palm-based powdered activated carbon (MPPAC)

Through a hydrothermal co-precipitation method, a magnetite film was coated on the outer surface of PPAC. The MPPAC synthesis method has been reported in previous paper (Wong et al., 2015)

with slight modification. Briefly, 72 g of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was dissolved in 200 mL of distilled water and 50 g PPAC was added into the solution. The suspension was transferred into a hydrothermal bottle and heated for 8 h at 80 °C after the alkaline solution (KNO_3 and NaOH) was added drop-wise. The MPPAC was separated using a magnet and washed thoroughly using distilled water to remove chemical residuals; the material obtained is referred to as MPPAC.

2.3. Silanization of MPPAC by TEPS

Surface silanization of PPAC or MPPAC was conducted by a typical silanization process (Liu et al., 2013; Saraji and Khaje, 2013). Briefly, 2 g medium was dispersed homogeneously into 100 mL ethanol (99.9%), followed by the addition of 0.2 mL TEPS into the suspension. Under mechanical stirring, the reaction was held at 70 °C for 7 h. The final composites were filtrated or separated using a magnet and washed thoroughly with ethanol prior to drying at 60 °C. PPAC and MPPAC coated with TEPS are referred to as PPAC-TEPS and MPPAC-TEPS, respectively.

2.4. Characterization of the adsorbents

Detail analysis and characterization of the adsorbents were describe in ESI, Section 2.1.

2.5. Analysis of surface functional groups

The surface properties of all samples were determined by Fourier-transform infrared spectroscopy (FTIR, Perkin Elmer, FTIR-Spectrum 400) using the KBr pellet method. Additionally, basic surface and oxygenated acid groups were quantified by Boehm titration (Boehm, 2002). The functional groups (carboxyl, lactone, phenol, carbonyl/quinone, and basic groups) present on the carbon surface could be determined by the different chemicals shown in Table S2. The procedure for the Boehm titration is described in the ESI, Section 2.2.

2.6. Sorption isotherms and kinetics

All experiments were carried out at room temperature. For kinetic tests, 0.01 g of each media was added into 100 mL of 10 mg L⁻¹ respective organic pollutants at pH 7 and then agitated in a thermostated shaker at 200 rpm for 6 h. Samples were obtained at predetermined time intervals and solute is collected after centrifuge at 100,00 rpm. The concentration of residual organic pollutants was measured using a spectrophotometer (Spectroquant Phoro 100, Merck) at different wavelengths (Table S1). With similar conditions, the pH effect was also investigated by adjusting the pH to 2 and 12, using 0.01 M HCl or NaOH.

For isotherms, the suspensions were shaken for 24 h with different concentrations of organic pollutants (10–50 mg L⁻¹), temperature (30–50 °C), and ionic strength (NaCl, 0.05–0.1 M). The amount of organic pollutants adsorbed (q_{eq}) and efficiency η (%) of the media applied were calculated as shown in ESI, Section 2.3. Adsorption isotherms were determined by Langmuir and Freundlich models. Whereas, pseudo-first order, pseudo-second order, and intraparticle diffusion were used for kinetic rate of reaction.

2.7. Thermal regeneration

Four different pollutants were adsorbed by PPAC and MPPAC-TEPS until saturated and analyzed by TGA-DSC. The energy changes during pollutant desorption as a function of temperature were analyzed with differential scanning calorimetry (DSC) graphs.

The selected POPs were adsorbed separately by PPAC and

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