Journal of Environmental Management 190 (2017) 274–282

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Review

Adsorption of non-steroidal anti-inflammatory drugs from aqueous solution using activated carbons: Review

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ARTICLE INFO

Article history:
Received 26 October 2016
Received in revised form
21 December 2016
Accepted 29 December 2016
Available online 4 January 2017

Keywords:
Activated carbon
Pharmaceuticals
Adsorption
Wastewater
Mechanism

ABSTRACT

Pharmaceutical pollutants are of significant effect on the environment, so that their treatments have been addressed in many studies. Activated carbon (AC) adsorbent shows best attraction for these compounds due to its unique characteristics represented by high capacity and porosity. In this article, the adsorption performance of AC towards non-steroidal anti-inflammatory drugs (NSAIDs) such as ibuprofen, ketoprofen, naproxen, and diclofenac were reviewed. According to collected data, maximum adsorption capacities of 417, 25, 290, and 372 mg/g were obtained from Langmuir isotherm for these drugs, respectively. The values of 1/n for Freundlich isotherm were lower than unity for all studied drugs, confirming the nonlinear and favorable adsorption. In addition, kinetics data were well represented by the pseudo-second-order model and mechanism was not controlled by the pore diffusion step alone. AC adsorption demonstrated superior performance for all selected NSAIDs, thus being efficient technology for treatment of these pharmaceutical pollutants.

Contents

1. Introduction

Pharmaceuticals have been classified as one of the most significant groups of environmental pollutants. Non-steroidal anti-inflammatory drugs (NSAIDs) represent one of the widely used pharmaceuticals. These drugs treat human and animal diseases in terms of analgesic, anti-inflammatory, and antipyretic actions (Rodríguez-Álvarez et al., 2013). Consequently, NSAIDs are among the most detected drugs in the aquatic environment. Due to their unique properties of hydrophilicity and stability, NSAIDs can remain in the aqueous phase. According to the literature, they exist in surface waters at concentrations up to μg/l (Manzo et al., 2014).

Several methods have been applied for treatment of NSAIDs pollutants like photocatalytic degradation (Kaur et al., 2015, 2016; Zhang et al., 2015a, 2015b), micro extraction (Manzo et al., 2014; D'Archivio et al., 2016), oxidation (Rodríguez-Álvarez et al., 2013), biodegrada tion (Yu et al., 2011; Koumaki et al., 2017), chlorination (Noutsopoulos et al., 2015), bio filtration (Binellia et al., 2014), electrocoagulation–flotation (Liu et al., 2015), electrochemical oxidation (Feng et al., 2013), and adsorption (Suriyanon et al., 2015; Cuerda-Correa et al., 2010; Jung et al., 2015). Among these

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http://dx.doi.org/10.1016/j.jenvman.2016.12.073
0301-4797/

techniques, a simple and low-cost adsorption process shows excellent removal efficiency towards pharmaceuticals (Vona et al., 2015).

In general, ACs present more stable adsorption properties as compared to clays, polymers, zeolites, and graphene-based adsorbents (Kyzas et al., 2015). Thus, the treatment of antibiotics by AC adsorption technology has been reviewed by many articles (Ahmed et al., 2015; Yu et al., 2016; Michael et al., 2013; Le-Minh et al., 2010; Homem and Santos, 2011). The majority of studied pharmaceutical pollutants are antibiotics because of their presence with relatively high concentrations in wastewaters (Kyzas et al., 2015). Rare reviews with poor informations are found in literature which include AC adsorption treatment of non-antibiotic pharmaceuticals (Rivera-Utrilla et al., 2013; Wang and Wang, 2016). Thus, the present review focuses on application of AC for adsorptive removal of non-antibiotic pollutants represented by NSAIDs such as ibuprofen, ketoprofen, naproxen, and diclofenac from aqueous solutions. It includes in detail the adsorption behaviors of these drugs in terms of isotherms, kinetics, thermodynamics, and mechanisms along with the effects of adsorption variables on capacity of each drug.

2. Activated carbon

Activated carbon (AC) is defined as a carbonaceous solid with high micropores volume, well developed surface area and high adsorptive capacity (Hesas et al., 2013; Pezoti et al., 2016). Therefore, AC has been classified as an efficient adsorbent for water treatment and air pollution control (Flores-Cano et al., 2016; Nor et al., 2013). The suitable application of AC depends on its properties which vary with used raw precursor and preparation technique (Ahmed and Theydan, 2012a; Torrellas et al., 2015). The common materials for the synthesis of AC are petroleum residue, natural coal and wood (Theydan and Ahmed, 2012). Recently, ACs are derived from agro-industrial wastes for a lower adsorption system cost (Kacan, 2016; Wong et al., 2016).

Activation of a carbonized precursor is the significant step in ACs production (Abbas and Ahmed, 2016; Ahmed, 2016b). The carbonized precursor or char is produced by pyrolytic decomposition of raw precursor at temperatures from 400 to 850 °C (Yahya et al., 2015). This char has favorable surface area and porosity which can be further enhanced by an activation step leading to a better porous product known as AC (Cha et al., 2016). The enhancement includes enlargement of char pores and creation of new pores (Chen et al., 2016). Physical, chemical, and physiochemical activations are the main techniques being applied for the production of ACs (Temdrara et al., 2015; Ahmed, 2016a).

Physical or thermal activation is conducted with the aid of an oxidant like carbon dioxide, air, and steam at a temperature range of 600–900 °C. Among these oxidants, CO_2 is preferred owing to its clean, ease, and flexible use (Ioannidou and Zabaniotou, 2007). In addition, uniform pores are created by CO_2 as compared to steam (Khezami et al., 2007). However, steam is more active than CO_2, thus producing ACs with high porosity. The smaller size of H_2O molecule facilitates effective diffusion of water cross the pores of char (Cagnon et al., 2009; Mak et al., 2009). Consequently, activation rate of steam is observed to be higher than CO_2 at constant activation conditions (Nowicki et al., 2010; Plaza et al., 2010).

Chemical activation includes impregnation of char with NaOH, H_3PO_4, $ZnCl_2$, and K_2CO_3 oxidants and activation at 300–500 °C. In this case, AC can be produced directly by a single step activation of precursor (Fig. 1) with developed porosity and high yield (Sudaryanto et al., 2006; Zuo et al., 2016). Activation by chemical oxidants is operated at low conditions as compared to physical activation (Ahmed, 2016c). Moreover, chemical AC has high microporosity and surface area (Ahmed and Theydan, 2013a, 2013b; Sumathi et al., 2010).

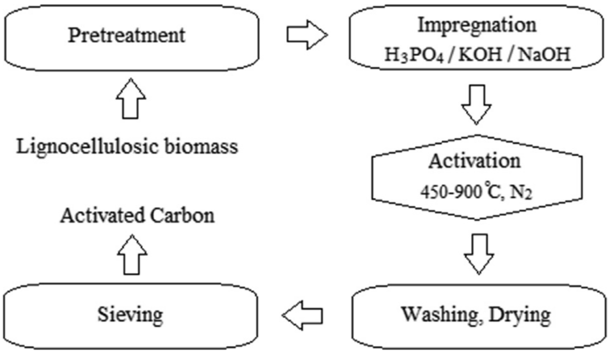

Fig. 1. Schematic diagram of chemical activation process for ACs production.

3. Anti-inflammatory drugs

Pharmaceuticals or drugs are medicinal compounds used for treatment of human and animal diseases (Deng et al., 2016; Lin and Li, 2016). The common drugs are antibiotics, analgesics and anti-inflammatories, painkillers, and hormones (Sun et al., 2015; Li, 2014). These chemicals appear in effluents of hospitals, drug factories, and landfills (Lu et al., 2016; Nazari et al., 2016). Although the concentration of drugs in wastewaters is low, their continuous release to aquatic system cause an emerging environmental problem (Caracciolo et al., 2015; Rakic et al., 2015).

Non-steroidal anti-inflammatory drugs (NSAIDs) constitute a group of pharmaceuticals with analgesic, antipyretic and anti-inflammatory effects (Schemeth et al., 2016). Considering the contamination level of NSAIDs in aquatic effluents, ibuprofen, ketoprofen, naproxen, and diclofenac can be considered as the most significant ones. These compounds are inefficiently treated by conventional processes according to results reported by application of advanced oxidation processes (Feng et al., 2013). The main physicochemical characteristics of studied pharmaceuticals are given in Table 1.

Ibuprofen (IBP) is a member of NSAIDs family found in water at concentration up to 24.6 μg/l, thus represents hazardous pollutant for human health (Guedidi et al., 2013). IBP has the properties of high consumption, slight water solubility, and high mobility in water (Mestre et al., 2007) Naproxen (NPX) belongs to the aryl acetic acid group of NSAIDs. It also presents in water at concentration with significant toxic effect towards ecosystems (Önal et al., 2007). NPX is commonly used for the reduction of pain, fever, and inflammation (Stancová et al., 2015). It is widely used in veterinary medicine with a chronic toxicity higher than its acute toxicity shown by bioassay tests. In addition, its photodegradation products are more toxic than NPX itself. Diclofenac (DCF), commonly used in ambulatory care, has a highest acute toxicity (Lonappan et al., 2016). It has been detected in wastewater, due to its high level of consumption and resistance for biodegradation (Bhadra et al., 2016). On the other hand, ketoprofen (KTP) is one of the propionic acid classes of NSAIDs with analgesic and antipyretic effects that exert its action by inhibiting the production of prostaglandin (Cuerda-Correa et al., 2010).

4. Adsorption isotherms

The adsorbate concentration as a function of adsorbed amount at equilibrium is an important relation in determination of attraction nature for a given adsorption system. In addition, the analysis