Contents lists available at ScienceDirect

Environmental Technology & Innovation

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

Removal of organic acids from water using biochar and petroleum coke

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HIGHLIGHTS

- Biochar (BC) and petroleum coke (PC) were tested for sorbing small organic acids.
- Lauric acid (LA) and 1-methylcyclohexenecarboxylic acid (MCA) sorption were tested.
- BC has greater binding site densities, and was generally a more effective sorbent.
- Admixtures of BC and PC showed synergy in removing lauric acid from solution.

ARTICLE INFO

Article history: Received 4 January 2016 Received in revised form 21 July 2016 Accepted 30 August 2016 Available online 4 September 2016

Keywords: Biochar Petroleum coke Organic acid Water Sorption

ABSTRACT

Alberta produces large volumes of oil sands process-affected water (OSPW) as a result of bitumen extraction and upgrading processes. Naphthenic acids (NAs) and other organic acids (OAs) comprise the main constituents of OSPW that can be acutely toxic to aquatic life. The recycling, safe return or storage of OSPW into the environment is a major challenge for the oil sands industry. Therefore, proper treatment technologies that are effective but inexpensive are needed. In this study, we tested the ability of a biochar (BC) produced from wheat straw and petroleum coke (PC) to remove two model organic acids (OAs) from aqueous solution: lauric acid (LA) and 1-methylcyclohexenecarboxylic acid (MCA). The results showed that BC was generally a more effective sorbent than PC, likely because BC has higher surface area and higher functional group densities than PC. More LA than MCA sorbed to both BC and PC due to the saturated 12-carbon chain of LA which renders it more hydrophobic than MCA. An admixture of BC and PC removed more LA from solution that was expected from its component parts, which may indicate a synergy between BC and PC in removing certain OAs from solution. This study shows that BC and PC might be useful materials for on-site treatment of organic acids. However, the use of PC may also be problematic due to release of significant heavy metals and sulfur to aqueous solution.

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http://dx.doi.org/10.1016/j.eti.2016.08.005 2352-1864/© 2016 Elsevier B.V. All rights reserved.

1. Introduction

The Alberta oil sands are composed of a mixture of water, sand and bitumen, with the latter being recovered by surface mining and by in-situ steam injection (Kelly et al., 2009). Bitumen extraction from surface-mined oil sands in Alberta, Canada is commonly achieved using the Clark hot water process (79–93 °C) that uses caustic soda to separate bitumen from other constituents such as clay, sand, dissolved metals and organic compounds including naphthenic acids (NAs), polycyclic aromatic hydrocarbons (PAHs), and other organic acids (OAs) (Giesy et al., 2010; Kelly et al., 2009, 2010). As a consequence, oil sands process-affected water (OSPW), generated on site during upgrading processes, produces tailings materials that contain high concentrations of OAs and PAHs (Ahad et al., 2013). Due to a zero-discharge policy by the province of Alberta, OSPW is stored in on-site tailing ponds, which cover an area of more than 130 km² (Giesy et al., 2010; Kelly et al., 2009). The Clark hot water process is water-intensive; one cubic meter of oil sands processing creates four cubic meters of OSPW and two to four barrels of water are required to extract one barrel of oil (Giesy et al., 2010). It is estimated that over a billion cubic meters of tailings water will be held in containment systems in the Athabasca region by 2025 as a result of the zero discharge policy.

NAs and other organics in OSPW are also persistent pollutants, and have been found in groundwater and surface waters near to oil sands tailings ponds (Frank et al., 2014; Kelly et al., 2010, 2009). Importantly, NAs and OAs in OSPW can accumulate in fish and other aquatic life forms, and pose risks to aquatic ecosystems and shallow freshwater aquifers (Ahad et al., 2013; Frank et al., 2008; Leung et al., 2001, 2003; Peters et al., 2007; Quagraine et al., 2005; Rogers et al., 2002). The proper handling of these large volumes of OSPW is a major challenge for the oil sands industry. To remove OAs from OSPW, several methods have been studied, including biodegradation, catalytic decarboxylation (using Ag₂O), membrane separation, and ozonation (Azad et al., 2013; Gamal El-Din et al., 2011; Iranmanesh et al., 2014; Quinlan and Tam, 2015; Scarlett et al., 2012; Zhang et al., 2006). These methods are effective in removal of OAs from wastewater, but are considered to be expensive, so their use is generally limited (Iranmanesh et al., 2014; Scott et al., 2008). Adsorption by activated carbon (AC) and nickel based alumina also showed promising results in removal of OAs from water, but are still expensive to implement (Azad et al., 2013; Gamal El-Din et al., 2011; Iranmanesh et al., 2014; Zubot et al., 2012). Two additional treatments materials that have received considerable recent attention are biochar (BC) and petroleum coke (PC). These materials are or can be produced near oil sand operations at low cost (Frankel et al., 2016; Gamal El-Din et al., 2011; Mohan et al., 2014; Ok et al., 2015; Pourrezaei et al., 2014). BCs are composed of a heterogeneous mix of carbonaceous materials with a range of surface functional groups that are activated by the thermal alteration of organic materials. BC surface functional groups are documented to be effective in the removal of organics (e.g., tricholoroethylene and sulfamethazine) and heavy metals contaminants from water (Ahmad et al., 2013, 2014; Rajapaksha et al., 2014; Uchimiya et al., 2012). The biochar pyrolysis temperature plays an important role in the stability, distribution and concentrations of surface functional groups, and thus the ability of the biochar to uptake pollutants from aqueous solution (Uchimiya et al., 2012; Vithanage et al., 2015). PC is a byproduct of the processing of oil sands, is abundantly available at mining sites, and may prove a good sorbent for OAs depending on its activation and the solution conditions (e.g., pH, water chemistry) (Gamal El-Din et al., 2011; Zubot et al., 2012). Due to its high sulfur content, PC is not generally considered useful for energy for heat generation, but may be used as an abundant and free-of-charge sorbent to treat OSPW (Zubot et al., 2012). A few previous studies have used PC as a pretreatment material before ozonation and have shown that PC can substantially remove acid extractable organics with carbon number (n) from 12 to 16, and in some cases for compounds with n > 18 and relatively high molecular weight (i.e., 200 amu or more). However there remains a gap in understanding the sorptive behavior of PC to lower molecular weight organics (n < 12) and molecular weight <200 amu. (Gamal El-Din et al., 2011; Mohammed et al., 2015). The smaller molecular weight OAs in OSPW generally exhibit greater toxicity to aquatic species than that larger molecular weight OAs, and have been overlooked in previous studies (Bauer, 2013).

In this study, we used the low molecular weight and low carbon number OAs methylcyclohexenecarboxylic acid (MCA) (142.1 amu; n = 8) lauric acid (LA) (200 amu; n = 12), and determined the effectiveness of BC, PC, and a combination of BC and PC to remove these aromatic (MCA) and aliphatic (LA) organic acids from aqueous solution. Overall, the objectives of this study were: (1) to assess the effectiveness and determine the feasibility of using BC and PC to remove OAs from water and (2) evaluate the use of admixtures of BC and PC for the removal of smaller molecular weight OAs.

2. Materials and methods

2.1. Chemicals and reagents

Distilled, deionized water (DDW) with 18.2 M Ω cm resistivity at 25 °C was used for preparing stock solutions and for all experiments. Hydrochloric acid (HCl), sodium bicarbonate (NaHCO₃), sodium carbonate (Na₂CO₃), sodium hydroxide (NaOH), calcium chloride (CaCl₂), barium chloride (BaCl₂) and 4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid (HEPES) were all of analytical grade and purchased from Fisher Scientific. Two organic acids: lauric (or dodecanoic) acid and 1-methylcyclohexenecarboxylic acid were purchased from Sigma Aldrich (Milwaukee, USA).

2.2. Biochar and petroleum coke production

BC produced from wheat straw was obtained from the Alberta Biochar Initiative (ABI). To produce the biochar, raw feedstocks were placed in a batch carbonizer and heated from 20 °C to 500 °C in 60 min under limited oxygen conditions.

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