



Granular activated carbon for simultaneous adsorption and biodegradation of toxic oil sands process-affected water organic compounds



Md. Shahinoor Islam, Yanyan Zhang, Kerry N. McPhedran, Yang Liu*,
Mohamed Gamal El-Din*

Department of Civil and Environmental Engineering, University of Alberta, Edmonton, Alberta T6G 2W2, Canada

ARTICLE INFO

Article history:

Received 29 October 2014
Received in revised form
8 January 2015
Accepted 14 January 2015
Available online

Keywords:

Biodegradation
Granular activated carbon
Naphthenic acids
Oil sands process-affected water
Ozonation

ABSTRACT

Naphthenic acids (NAs) released into oil sands process-affected water (OSPW) during bitumen processing in Northern Alberta are problematic for oil sands industries due to their toxicity in the environment and resistance to degradation during conventional wastewater treatment processes. Granular activated carbon (GAC) has shown to be an effective media in removing biopersistent organics from wastewater using a combination of adsorption and biodegradation removal mechanisms. A simultaneous GAC (0.4 g GAC/L) adsorption and biodegradation (combined treatment) study was used for the treatment of raw and ozonated OSPW. After 28 days of batch treatment, classical and oxidized NAs removals for raw OSPW were 93.3% and 73.7%, and for ozonated OSPW were 96.2% and 77.1%, respectively. Synergetic effects of the combined treatment process were observed in removals of COD, the acid extractable fraction, and oxidized NAs, which indicated enhanced biodegradation and bioregeneration in GAC biofilms. A bacteria copy number $>10^8$ copies/g GAC on GAC surfaces was found using quantitative real time polymerase chain reaction after treatment for both raw and ozonated OSPW. A Microtox[®] acute toxicity test (*Vibrio fischeri*) showed effective toxicity removal ($>95.3\%$) for the combined treatments. Therefore, the simultaneous GAC adsorption and biodegradation treatment process is a promising technology for the elimination of toxic OSPW NAs.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

Crude oil deposits in the form of bitumen are estimated to be around 176.8 billion barrels in the oil sands of Northern Alberta, Canada (Pereira et al., 2013). Extraction of bitumen from the oil sands using hot alkaline water generates a large volume of oil sands process-affected water (OSPW) (Anderson et al., 2012). The OSPW is currently being stored in cement-lined tailings ponds in compliance with Alberta's zero discharge policy for OSPW (Pereira et al., 2013) with the present volume of tailings ponds being greater than 10^9 cubic meters (Hagen et al., 2014). OSPW is toxic to bacteria, fish, zooplankton, phytoplankton, mammals, plants and birds (Pereira et al., 2013; Whitby, 2010) and creates an ecological imbalance in Northern Alberta (Johnson and Miyanishi, 2008). The major toxicity of OSPW to aquatic organisms is attributed to a group

of organic carboxylic surfactants known as naphthenic acids (NAs) (Anderson et al., 2012). The general chemical formula of NAs is $C_nH_{2n+Z}O_x$, where n is the number of carbon atoms, Z is either zero or a negative even integer representing the number of hydrogen atoms lost because of ring formation ($Z = 0$ to -12), and x represents the number of oxygen atoms ($x = 2$ for classical NAs; $x = 3$ to 5 for oxidized NAs, currently noted as O-NAs, O_2 -NAs, and O_3 -NAs, respectively) (Grewer et al., 2010). Given the large quantities of OSPW both in use and in storage, appropriate OSPW treatment technologies are currently being investigated to extend OSPW recycling to help reduce the need for fresh water intake from the Athabasca River. In addition, proper treatment of OSPW is needed to eventually permit the safe discharge of treated OSPW to the receiving environments, which helps to complete re-creation of landforms and ecosystems as suitable habitats for indigenous plants and animals (Johnson and Miyanishi, 2008).

A potentially environmentally sound and economical approach for OSPW treatment is the use of a biofilm approach which has been used successfully in industrial wastewater treatment (Baban

* Corresponding authors.

E-mail addresses: yang.liu@ualberta.ca (Y. Liu), mgamalel-din@ualberta.ca (M. Gamal El-Din).

et al., 2010). Despite the possible benefits of the biofilm process, few biofilm based OSPW treatment studies have been reported and it has been suggested that a biofilm process using a nonadsorptive support media is ineffective for OSPW NAs degradation (Headley et al., 2010; Hwang et al., 2013). The NAs resistance to degradation can be attributed to their extensive cyclical molecular structures being difficult for microorganisms to metabolize. Given this recalcitrance, it is clear that other processes should be considered to increase the biodegradability of NAs before or during the biofilm degradation treatment to improve their removal efficiency.

Granular activated carbon (GAC), as a biofilm substrate, may be useful in increasing the degradation of OSPW NAs. Previously, the GAC-biofilm combination has been applied successfully to treat industrial wastewaters (Baban et al., 2010; Rao et al., 2005) and oil field produced water (Zhao et al., 2006) that contain a large variety of recalcitrant organic compounds. Compared to previous studies, our recent study showed higher removals of raw OSPW NAs using the GAC-biofilm process for effectively degrading ~86% of NAs (Islam et al., 2014). This markedly improved treatment from previous studies may be attributed to the synergetic effects of the GAC adsorption and the attached biofilm biodegradation. Synergetic effects include the previously mentioned regenerative effects, as well as the concentration of organic compounds (including NAs) adsorbed on GAC surfaces that can trigger specific microbial metabolism mechanisms which can preferentially accelerate the compound-specific biodegradation efficiency. In addition, the biofilm growth on GAC surfaces increases the contact time between microorganisms and the adsorbed organic compounds, which may help to further improve the treatment efficiency. These synergistic removal mechanisms have not been investigated previously for OSPW, thus further study is clearly needed to help elucidate the potential benefits of these processes.

To determine the synergistic mechanisms of the combined GAC adsorption-biodegradation treatment process, this study investigated the following: (i) the role of GAC adsorption only using sterilized OSPW (no biodegradation); (ii) the impact of biodegradation only in the absence of GAC (planktonic biodegradation only); and (iii) the synergetic effect of a combined GAC adsorption-biodegradation system, on the removal of NAs in OSPW. To determine the combined treatment effectiveness, the impact of NAs chemical structure including carbon number (n) and cyclicity ($-Z$) were assessed. Previously, it has been shown that ozone can be useful for the complete degradation and/or biotransformation of classical NAs ($x = 2$) that exhibit high cyclization and long chains and, thus, increase their biodegradability (Martin et al., 2010; Wang et al., 2013a). Given this potential for increased biodegradability, the efficacy of the combined treatment using preliminary ozonated OSPW was also tested.

2. Materials and methods

2.1. Materials

Selected grade bituminous coal based steam-activated granular carbon (GAC) (SGL 8 × 30) (Calgon Carbon Corporation, Pittsburgh, PA, USA) with mean diameter of 1.5–1.7 mm was used to conduct the experiments. OSPW was sampled from an oil sands tailings pond in Northern Alberta, November, 2011, and stored in a cold room at 4 °C before use.

2.2. Methods

2.2.1. Ozonation of raw OSPW

Raw OSPW at room temperature was used for ozonation in a 4 L vacuum flask equipped with a ceramic fine bubble gas diffuser

located at the bottom. Ozone was generated by an ozone generator (PCI-WEDECO, GSO-40, Herford, Germany) using extra-dry, high-purity oxygen. The ozone gas mixture at 1 L/min containing an ozone concentration ~3 wt% ozone (~40 mg/L) was bubbled through the diffuser for 5 min. The utilized ozone dose was calculated by difference between the ozone concentrations at the reactor inlet and outlet as measured with identical ozone monitors (Model HC-500, PCI-WEDECO). The detailed ozonation procedure has been described previously by Wang et al. (2013a,b).

2.2.2. Simultaneous adsorption and biodegradation of OSPW

Four treatments were considered for both raw and ozonated OSPW (500 mL in each reactor) including: (1) 0.2 g GAC with OSPW (combined treatment); (2) OSPW without GAC (planktonic biodegradation only treatment); (3) sterilized OSPW with GAC (adsorption only treatment); and (4) sterilized OSPW without GAC (no treatment control). The GAC and sterilized OSPW were sterilized using a vacuum/gravity autoclave (Model 733LS, Getinge group Inc., NY, USA) at 121 °C for 30 min. Amber bottles containing triplicates of individual treatments were shaken at 150 rpm on a horizontal shaker (Innova™ 2100, Platform Shaker, New Brunswick Scientific, USA) for 28 days at 21 ± 1 °C. For biodegradation experiments, raw OSPW with endogenous bacteria was used without pretreatment, whereas ozonated OSPW was inoculated with endogenous bacteria extracted from raw OSPW by centrifugation using the following protocol. A 2.5 L volume of ozonated OSPW was centrifuged (Multifuge 3S/3S-R, Heraeus, Houston, USA) at 3700 rpm for 10 min to remove ozonated OSPW microbes, and the supernatant was collected. Raw OSPW microorganisms were then collected as pellets from 2.5 L of raw OSPW after centrifuging under the same conditions. Subsequently, raw OSPW microorganisms were inoculated in the bacteria-free ozonated OSPW supernatant.

2.3. Analysis of water chemistry

Chemical oxygen demand (COD), dissolved organic carbon (DOC), biochemical oxygen demand (BOD_5), and total solids (TS) were analyzed in triplicate in raw and ozonated OSPW according to standard methods (APHA, 2005). Fourier transform infrared (FT-IR) spectroscopy (PerkinElmer®, Woodbridge, ON, CA) was used to measure the acid extractable fraction (AEF) concentration in OSPW prior to and after treatment, as described elsewhere (Gamal El-Din et al., 2011) and briefly in Appendix A.

Aliquots (2 mL) from each replicate of an individual treatment were collected on days 5, 14, and 28 and pooled for NA analysis using UPLC-HRMS. Samples were pooled due to the high analytical cost of the UPLC-HRMS, however, concentrations are representative of each individual treatment. Quantification of classical NAs and oxidized NAs was performed using a Waters Acquity ultrahigh performance liquid chromatography (UPLC) system (Milford, MA, USA) and a high resolution Synapt G2 HDMS (40,000 FWHM) as described in a previous study (Wang et al., 2013a). The Synapt G2 HDMS system was integrated with an ion-mobility cell between the electrospray ionization source operating in negative ion mode and a TOF MS detector.

The acquisition of ion mobility spectra (IMS) to qualitatively assess classical NAs, oxidized NAs (O_x -NAs where $x \geq 3$) and potential heteroatom (sulfur and nitrogen) NAs has been described previously by Wang et al. (2013a). In brief, a Tri-Wave® ion-mobility cell of 15 cm long was used to conduct the ion mobility spectra (IMS) using nitrogen (purity > 99%) as the drift gas. A transfer cell in the IMS gathered a defined amount of ions (actual number of ions is not known) and a helium gate released the ions into the ion-mobility cell. Ions were moved counter-current to the gas flow

Download English Version:

<https://daneshyari.com/en/article/7482445>

Download Persian Version:

<https://daneshyari.com/article/7482445>

[Daneshyari.com](https://daneshyari.com)