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Research article

Evaluation of biochar-ultrafiltration membrane processes for humic acid removal under various hydrodynamic, pH, ionic strength, and pressure conditions

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ABSTRACT

The performance of an ultrafiltration (UF)-biochar process was evaluated in comparison with a UF membrane process for the removal of humic acid (HA). Bench-scale UF experiments were conducted to study the rejection and flux trends under various hydrodynamic, pH, ionic strength, and pressure conditions. The resistance-in-series model was used to evaluate the processes and it showed that unlike stirred conditions, where low fouling resistance was observed $(28.7 \times 10^{12} \text{ m}^{-1}$ to $32.5 \times 10^{12} \text{ m}^{-1})$, higher values and comparable trends were obtained for UF-biochar and UF alone for unstirred conditions $(28.7 \times 10^{12} \text{ m}^{-1}$ to 32.5 \times 10¹² m⁻¹). Thus, the processes were further evaluated under unstirred conditions. Additionally, total fouling resistance was decreased in the presence of biochar by 6%, indicating that HA adsorption by biochar could diminish adsorption fouling on the UF membrane and thus improve the efficiency of the UF-biochar process. The rejection trends of UF-biochar and UF alone were similar in most cases, whereas UF-biochar showed a noticeable increase in flux of around 18–25% under various experimental conditions due to reduced membrane fouling. Three-cycle filtration tests further demonstrated that UF-biochar showed better membrane recovery and antifouling capability by showing more HA rejection (3-5%) than UF membrane alone with each subsequent cycle of filtration. As a result of these findings, the UF-biochar process may potentially prove be a viable treatment option for the removal of HA from water.

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1. Introduction

Natural organic matter (NOM) is known to contribute to unpleasant odor and taste in water. NOM is also a widely known precursor to potentially carcinogenic disinfection byproducts ([Hua](#page--1-0) [et al., 2013\)](#page--1-0) and a significant contributor to membrane fouling ([Yoon et al., 2005](#page--1-0)). In particular, one of the major hydrophobic components of dissolved NOM (DOM) is humic acid (HA), consisting of anionic macromolecules having a wide range of molecular weights [\(Chu et al., 2016, 2017; Heo et al., 2012](#page--1-0)). It is often accountable for severe membrane fouling due to extensive pore

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adsorption, making the use of membrane filtration in water treatment more challenging ([Yu et al., 2008\)](#page--1-0).

DOM removal from water using conventional ultrafiltration (UF) membranes has been demonstrated to be largely ineffective due to membrane fouling, restricting its widespread application as a result of increases in hydraulic resistance and operational and maintenance costs, and deterioration in productivity due to the frequency of membrane regeneration problems [\(Zularisam et al., 2006\)](#page--1-0). UF membranes alone function poorly in removing color, DOM, and synthetic organic chemicals ([Lebeau et al., 1998](#page--1-0)). To overcome some of these shortcomings, UF membranes have been coupled with adsorbents to develop integrated processes that meet treatment requirements and improve process efficiencies. Coupling of UF membranes with powdered activated carbons (PACs) is one of the most widely studied integrated processes ([Campinas and Rosa,](#page--1-0) [2010; Li et al., 2003; Oh et al., 2006; Tomaszewska and Mozia,](#page--1-0)

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[2002\)](#page--1-0), which can effectively counter irregular and unpredictable increases in odor and taste compounds. It can also enhance the productivity of the membrane by reducing the foulant load toward the applied membrane [\(Oh et al., 2006\)](#page--1-0). However, if the PAC dose, membrane characteristics, and operating conditions are not monitored effectively, there can be a more severe flux decline with a PAC-UF membrane process than a UF process alone [\(Li et al., 2003](#page--1-0)).

To improve the efficiency of integrated processes, newer adsorbents such as biochars (e.g., a waste or byproduct of bio-oil production), have been studied ([Abdel-Fattah et al., 2015; Jung](#page--1-0) [et al., 2015a,b,c,d](#page--1-0)). The properties of biochar are dependent on the manufacturing process and vary with the pyrolysis conditions and the type of biomass used. Short-term pyrolytic conditions with low thermal treatment (<500 °C) are known to produce an enriched, carbon-based porous biochar with properties suitable for use as an adsorbent [\(Jung et al., 2015c\)](#page--1-0). Such biochar has favorable properties for adsorbing metal cations due to a large surface area with a high negative charge and charge density (Kołodyńska et al., [2017; Noraini et al., 2016; Yap et al., 2017](#page--1-0)) In addition, its hydrophobic characteristics provide high affinity to adsorb hydrophobic organic compounds such as 17a-ethinyl estradiol, diclofenac, ibuprofen ([Jung et al., 2013](#page--1-0)), and DOM [\(Hurtado et al., 2017; Kasozi](#page--1-0) [et al., 2010\)](#page--1-0). Biochar also exhibits superior adsorption performance to activated carbon in terms of organic removal, making it a potentially cost-effective adsorbent ([Jung et al., 2015a](#page--1-0)).

However, the effects of the adsorbent on membrane fouling need to be analyzed for any adsorbent used in an integrated process. The effects are a function of the physiochemical properties of the adsorbent (e.g., size, charge, and hydrophobicity), solution water chemistry (e.g., pH and ionic strength), and membrane characteristics (e.g., hydrophobicity, charge, and size) ([Campinas](#page--1-0) [and Rosa, 2010; Konieczny and Klomfas, 2002; Lin et al., 1999;](#page--1-0) [Tomaszewska and Mozia, 2002](#page--1-0)). Based on these properties, the adsorbent can remove HA from the solution, which would otherwise adsorb onto the membrane pores and cause fouling, or the adsorbent may itself act as a foulant, blocking membrane pores and preventing the efficient working of the integrated process. Extensive studies have been conducted on the effects of PAC on membrane fouling in an integrated process ([Lebeau et al., 1998; Li et al.,](#page--1-0) [2003; Oh et al., 2006\)](#page--1-0), whereas biochar, being relatively new in the field of water treatment, has been studied less in comparison.

A few studies have been reported previously on the adsorption mechanisms of DOM with biochar [\(Chintala et al., 2014; Jung et al.,](#page--1-0) [2015c; Kasozi et al., 2010\)](#page--1-0). Hydrophobic HA forms π bonds with biochar and occupies the π -rich sites on the surface, acting as a π acceptor, while biochar acts as an π donor [\(Jung et al., 2015c\)](#page--1-0). This leads to a higher adsorption affinity for HA on biochar when compared with other commercially available adsorbents, like PAC. Additionally, the presence of diverse functional groups on HA surface also promotes multi-layer adsorption with biochar due to increased adsorption via hydrogen bonding and polar interactions.

The objective of this study was to investigate the potential mechanisms for HA removal in an integrated process with a UF membrane in the presence of biochar prepared in the laboratory as an adsorbent. The effect of biochar on membrane fouling was also studied. To achieve this, flux decline and HA rejection in biochar-UF processes were investigated as a function of different solution and operating conditions, including pH, ionic strength, pressure, and biochar concentration.

2. Materials and methods

2.1. HA feed solution

HA (Sigma-Aldrich Co., St. Louis, MO, USA) was used as a

representative DOM, because it is one of the most dissolved hydrophobic components of NOM, thus making its treatment more challenging. HA, with a molecular weight distribution between 180 and 22,600 Da, is a mixture of complex polyelectrolytes, formed chemically with hydroxylic and carboxylic functional groups [\(Lin](#page--1-0) [et al., 1999\)](#page--1-0). To prepare the HA stock solution, powdered HA was dissolved in ultrapure deionized (DI) water and filtered consecutively through $GF/F(0.7 \mu m)$ glass microfiber filters (Whatman Inc., Piscataway, NJ, USA) and Durapore $(0.45 \mu m)$ membrane filters (Millipore Inc., Billerica, MA, USA) to remove any impurities and particulate matter. This HA stock solution was further diluted with DI water to obtain a concentration of 5 mg/L as dissolved organic carbon (DOC), which was used as the feed solution for rejection and flux decline experiments.

2.2. Characteristics of biochar

The biochar used in this study was produced under controlled laboratory conditions using the following process. Detailed production procedures were described in our previous report ([Jung](#page--1-0) [et al., 2013\)](#page--1-0). Briefly, torrefied loblolly pine chips (15 \times 6 mm) containing bark were treated thermally at 300 \degree C for 15 min in a laboratory-scale batch-tube furnace (OTF-1200X; MTI Corp., Richmond, CA, USA). The particular biochar used in this study was produced under a 7% oxygen +93% nitrogen atmosphere. Next, 3 g of this pyrolyzed biochar was activated with 40 mL of 4 M NaOH for 2 h and dried overnight at 105 \degree C. This was then heated for 2 h at 800 \degree C under a nitrogen gas flow (2 L/min) and subsequently cooled at a rate of 10° C/min following separation from the solution using a Buchner filter funnel. The characteristics of the biochar thus produced are described in [Table 1.](#page--1-0)

2.3. Biochar-UF experiments

A commercially available flat sheet polyethersulfone (PES) UF membrane (Sterlitech Corp., Kent, WA, USA), having a measured molecular weight cut-off of approximately 5 kDa, was used. The membranes were first cut to desired sizes and then washed thoroughly with DI water at least five times and preserved by soaking in DI water. To prevent microbial growth, the membranes were then stored away from direct light at 4 °C. A VCA Optima process (Optima XE; AST Products, Inc., Billerica, MA, USA) was used to measure the contact angle of the UF membrane. Contact angle is a measure of the hydrophilicity of the membrane. The water droplet size of $3-5$ μ L, the optimal volume for producing reproducible contact angle measurements with a nominal error of $\pm 2.3^{\circ}$, was dropped cautiously onto the UF membrane. The characteristics of the membrane are described in [Table 2.](#page--1-0)

Flux decline and HA rejection tests were used to evaluate performance of the biochar-UF membrane process. The bench-scale set up for the UF-biochar experiments was as shown in [Fig. 1.](#page--1-0) For all filtration tests, a dead-end stirred filtration process (Sterlitech Corp., Kent, WA, USA) was used, having a volume of 300 mL and an effective area of 14.2 cm^2 . The membrane filtration set-up was coupled with a biochar reactor. An initial concentration of 5 mg/L of HA was mixed with biochar (20 mg/L) for a contact time of 2 h to simulate realistic water treatment plants conditions, where adsorbent doses of $5-50$ mg/L are used over a contact period of $1-2$ h ([Heo et al., 2012](#page--1-0)). The majority of UF experiments were conducted under no-stirring conditions; transmembrane pressure (TMP) was kept constant at different levels (172, 345, and 689 kPa). Also, 210 mL of permeate and 90 mL of retentate was collected for each UF experiment. The flux decline was monitored for water recovery to show fouling-related flux decline in the membrane. The concentration of HA was determined by $UV-vis$ adsorption Download English Version:

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