



Removal of humic and tannic acids by adsorption–coagulation combined systems with activated biochar



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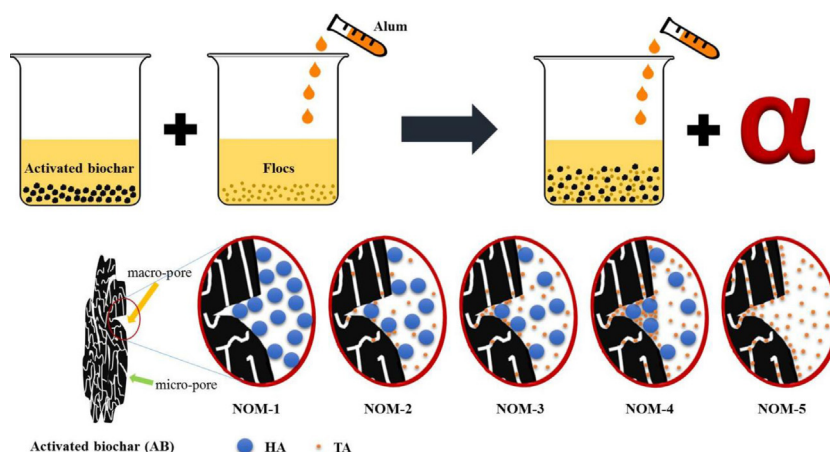
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HIGHLIGHTS

- A biochar was produced by fast pyrolysis under low temperature.
- Removal of NOM by biochar was significantly higher than that of commercial PAC.
- Synergistic effect in an adsorption–coagulation process was anticipated for NOM removal.

GRAPHICAL ABSTRACT



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ABSTRACT

Despite recent interest in transforming biomass into bio-oil and syngas, there is inadequate information on the compatibility of byproducts (e.g., biochar) with agriculture and water purification infrastructures. A pyrolysis at 300 °C yields efficient production of biochar, and its physicochemical properties can be improved by chemical activation, resulting in a suitable adsorbent for the removal of natural organic matter (NOM), including hydrophobic and hydrophilic substances, such as humic acids (HA) and tannic acids (TA), respectively. In this study, the adsorption affinities of different HA and TA combinations in NOM solutions were evaluated, and higher adsorption affinity of TA onto activated biochar (AB) produced in the laboratory was observed due to its superior chemisorption tendencies and size-exclusion effects compared with that of HA, whereas hydrophobic interactions between adsorbent and adsorbate were deficient. Assessment of the AB role in an adsorption–coagulation hybrid system as nuclei for coagulation in the presence of aluminum sulfate (alum) showed a synergistic effect in a HA-dominated NOM solution. An AB–alum hybrid system with a high proportion of HA in the NOM solution may be applicable as an end-of-pipe solution.

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1. Introduction

Interest in bio-oil production to serve as an alternative to fossil fuels and to address global climate change has increased. Thermal treatment of biomass (plants, domestic and industrial wastes, sewage sludge, animal manure) such as in pyrolysis processes under limited oxygen conditions yields syngas and biochar as well as bio-oil [1]. The yield and quality of these products are sensitive to pyrolysis conditions (temperature, contact time) and type of the biomass, while biochar has been treated as a waste or byproduct from such energy production. However, short-term pyrolytic conditions under lower thermal treatment (<500 °C) resulted in low oxygen and hydrogen contents in an enriched carbon-based porous biochar [2]. These properties enable it to replace pulverized coal and activated carbon in the fields of soil amendment [3] and water purification [4], respectively.

The properties of a large surface area with a high negative surface charge and charge density in carbonized biochar provide the ability to adsorb metal cations [5,6], and the hydrophobic characteristics provide affinity to adsorb hydrophobic organic compounds, such as hydrophobic pharmaceuticals [7,8] and natural organic matter (NOM) [9,10]. Additionally, superior adsorption performance of biochar compared with activated carbon has been reported in terms of organic removal, suggesting the use of biochar as a cost-effective adsorbent [11].

The aquatic environment is complex and consists of a wide variety of organic compounds, including those from the decomposition of plant and animal materials and NOM [12]. The presence of NOM in various water sources is a major concern in water treatment, and achieving better purification has been a focus for environmental scientists and engineers. NOM often contributes not only to offensive taste and odors but also to various complexation processes with harmful (emerging) organic/inorganic chemicals (e.g., endocrine-disrupting compounds, pharmaceuticals, heavy metals) [13]. Additionally, NOM is a potential precursor to carcinogenic by-products, such as trihalomethanes and halo acetic acids, that can form during chlorination and contribute to bacterial regrowth and biofilm formation in drinking water distribution systems [14] as well as to the fouling of microfiltration, ultra-filtration, and nanofiltration membranes [15]. Natural surface-active materials such as humic acids (HA) [16] and tannic acids (TA) [17] are widely distributed in the environment, and studies on their removal by various treatments and processes have been conducted to purify water contaminated with NOM. However, there is a lack of information on the different contributions of HA and TA in surface water and wastewater in purification, and particularly their adsorption onto activated biochar.

The primary objective of this study was to investigate the removal of HA and TA through a combination of adsorption and coagulation with activated biochar (AB) and aluminum sulfate (alum). The respective removal efficiencies of HA and TA were evaluated in terms of adsorption capacity, as well as in a coagulation system with alum. Moreover, the NOM removability was compared with the performance of commercially available powdered activated carbon (PAC). The target NOM (HA and TA) solutions used in this study had five different HA/TA ratios, and removal mechanisms and 'ideal' water chemistry were discussed in terms of the physicochemical properties of the adsorbent and adsorbate.

2. Materials and methods

2.1. Adsorbents and coagulant

Pinus taeda, known as loblolly pine chip, and bark were pyrolyzed at 300 °C for 15 min under pure nitrogen gas in a

laboratory-scale batch tube furnace (OTF-1200X, MTI Corp., USA) to increase the yield of biochar (42.3%). The biochar was produced with less than 3% difference in the weight loss from each batch pyrolysis and was then ground and sieved to a No. 200 mesh (<74 μm) fraction after chemical activation, as described elsewhere [7]. In brief, pyrolyzed biochar samples were impregnated with a NaOH solution, heated under nitrogen gas at 800 °C, and cooled down. The dried samples were rinsed with 0.1 M HCl followed by deionized water and then dried at 105 °C. Commercially available coal-derived powdered activated carbon (PAC), WPM (Calgon Carbon Corp., USA) was used for comparison. Aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$; Fisher Scientific, USA) was used to coagulate/flocculate the NOM. Concentrated alum stock solution was prepared, and then different volumes of the stock solution were transferred to the jar-test apparatus to yield the desired concentration.

2.2. Chemicals

HA and TA were purchased from Sigma–Aldrich (St. Louis, MO, USA). HA stock solution (800 mg/L as dissolved organic carbon, DOC) was prepared by adding pre-dried (105 °C) HA (2 g) to 1 L of deionized water (18.2 MΩ/cm), and the dissolvability was increased by increasing the pH, to ~10, with 1 M NaOH solution. The solution was then filtered through a 0.22-μm Durapore membrane filter (Millipore, Cork, Ireland) to remove particulate matter, while 100 mg/L as DOC of TA stock solution was prepared for all experiments without further filtration.

2.3. Water sources

Volumes of the HA and TA stock solutions were added to achieve total concentrations of 10 mg/L as a DOC value for each type of NOM with different combinations of HA and TA. Table 1 shows the compositions of the different NOM solutions. The compositions of these solutions were determined based on a review of several studies related to the treatment and characterization of drinking water sources [18,19]. Ranges of water chemistry values were adopted for drinking water sources, and different HA/TA ratios mimicked the hydrophobic and hydrophilic properties. NOM-1 corresponds to a more hydrophobic NOM solution, and NOM-5 to a more hydrophilic NOM solution. The pH of each NOM solution was adjusted to neutral using 1 M HCl and 1 M NaOH, and conductivity was measured to verify the characteristics of the NOM solutions, accounting for the contributions of ionic strength.

2.4. Adsorption experiments

Isotherm batch experiments were conducted under fixed initial NOM conditions at 10 mg/L DOC for both HA and TA solutions under various adsorbent doses at pH 7. Stock suspensions of 10,000 mg/L of the adsorbents (AB and PAC) were prepared and then stirred at 500 rpm for the homogeneous transfer to the sample of 0–100 mg/L in 40-mL amber vials with a polytetrafluoroethylene-lined screw cap; they were then placed in a rotator (14 rpm) for 7 d to achieve equilibrium. The adsorption of mixed NOM solutions under different combinations of HA and TA with both adsorbents was conducted in a similar manner to the isotherm experiment. Various adsorbent doses (20–100 mg/L) were applied to evaluate the effects of adsorbent dosage. Kinetic batch experiments were performed with a 1-L jar-test apparatus with the five different NOM solutions at pH 7. All NOM solutions were placed in cylindrical jars for fixed contact times (0, 1, 2.5, 5, 15, 30, 45, and 60 min), and each kinetic constant was compared with the coagulation results conducted under short contact conditions (37 min). Each sample was then filtered through a membrane filter (0.45 μm) and pre-

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