

Removal of total organic carbon from peat solution by hybrid method—Electrocoagulation combined with adsorption

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ABSTRACT

Humic substances end up in water from the drainage basin causing eutrophication and the spread of algae. These natural organic substances have an influence on the physical, chemical and biological properties of the water system. Adsorption and electrocoagulation (EC) are commonly used purification methods in the water and wastewater treatment. Both methods are used by themselves for removing a wide range of impurities. In this research, the novelty was to study the effect of combined activated carbon (AC) adsorption and electrocoagulation method on the removal of organic substances as total organic carbon (TOC) from the peat solution. With the hybrid method, TOC content was efficiently removed (~95%) by using first adsorption treatment followed by electrocoagulation with Al- or Fe -electrode. Instead, when using the adsorption and electrocoagulation separately, the removal of TOC was 79–89%. Based on this study, the hybrid method could be efficiently used for removing organic substances from the water system.

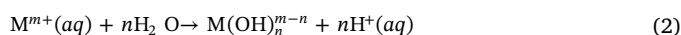
1. Introduction

Humic substances are organic, high molecular weight, natural polymers and originate from the decomposition of organic substances. Organic substances come to the water from drainage basins and from water system itself. The toxic algae and microorganisms utilize inorganic and organic nutrients of humic substances in the water. Humic substances have an impact on the toxicity of heavy metals and organic pollutants forming complexes with many toxic substances [1–4].

Humic substances occur in water as insoluble form, colloidal form and solid form. Humic substances are complex organic substances that vary from light brown (fulvic acids) to black (humins) in color. They consist of three groups of heterogeneous organic polymers: humic acids, fulvic acids and humin substances. Humic acids are alkali-soluble and fulvic acids are both alkali and acid soluble whereas humin substances are either alkali or acid soluble [5]. Humic acids are the most abundant fraction of humic substances in soil. Humic acids have various functional groups, such as carboxylic, alcoholic and phenolic groups and in a random way arranged aromatic rings [6]. These groups provide a number of potential binding sites for metal ions, for example, chlorinated organic compounds, which are toxic disinfection byproducts [7–9].

Electrocoagulation (EC) is an old but growing water and wastewater treatment method. Contaminants that can be removed by

electrocoagulation are for example heavy metals [10,11], pharmaceuticals and antibiotics [12,13] and organic pollutants e.g. herbicides, phenols, textile dyes [14–17]. The electrocoagulation reactor is an electrochemical cell, which has two electrodes, an anode as a sacrificial metal, and cathode. Aluminum and iron metals are typically used as anode metals. Electrocoagulation generates metal ions from a sacrificial anode (Eq. (1)). The metal ions from the anode spontaneously hydrolyze in water forming various coagulant species (Eq. (2)). These coagulant species agglomerate together forming larger particles by coagulation [18,19]. The purpose of coagulation is to destabilize particles by coagulant chemicals and enable them to become attached to other particles. Aluminum and iron salts hydrolyze in the water forming insoluble precipitates, which destabilize the charge of particles by adsorbing onto their surface. There are repulsive forces between the particles because they have similar electric charge, usually negative. The hydrolyzed products have positive electric charge.



Electrolytically formed gases, mainly hydrogen, are generated on

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the surface of the cathode (Eq. (3)). During beneficial side reactions, gas bubbles are generated which promotes flotation. These agglomerated pollutants form larger agglomerates, which rise upwards to be removed in the subsequent process. The electrochemical reduction may take place on the cathode (Eq. (4)). [20,21]

Activated carbons (AC) are widely used in different industrial water purification processes as adsorbents because of its high surface area and well-developed porous structure [22–28]. Adsorption has been considered as one of the most economically viable techniques for water treatments and AC is the most widely used adsorbent. Typically ACs have been prepared from carbonaceous materials e.g. coal. As an alternative eco-friendly and low-cost raw material, AC can be prepared from residual or waste biomass materials [29–33]. For the preparation of AC, thermal procedures including carbonization and activation are used. AC has been proven as an effective adsorbent for the removal of a wide range of organic and inorganic pollutants dissolved in aqueous media. During the adsorption, dissolved species diffuse into the pores of adsorbent. Because adsorption takes place on the surface, a large surface area and a high number of pores are necessary for adsorbent. With high porosity, adsorbents can have a pore volume of $0.1\text{--}0.8\text{ mL g}^{-1}$ and a surface area ranging from ~ 700 to $1500\text{ m}^2\text{ g}^{-1}$. As a result, the adsorption capacity can be as high as 0.2 g of adsorbate per gram of adsorbent, depending on the adsorbate concentration and type [21,34]. The size of the adsorbate that can enter a pore is limited by the size of the pore. In the case of humic acids when adsorbate is a large organic molecule, meso- and macroporous (pore diameter $> 2\text{ nm}$) structure of activated carbon is favored [35]. Even though AC has a large market as an adsorbent in water treatment processes, the specific adsorption mechanisms for solutes containing organic and inorganic material are still not clear [36].

In this paper, the removal of organic substances has been studied by a hybrid method: electrocoagulation combined with adsorption. This kind of experiment setup hasn't been studied before in removal of organic molecules from waste waters. As an electrocoagulation material, iron and aluminum have been used and the process was optimized for the reaction. As an adsorbent material activated carbon (forest-residue-based spruce from Finland) has been used. The removal of total organic carbon has been studied with separate adsorption or electrocoagulation methods and compared to the hybrid method. In the hybrid method, a combination of adsorption and electrocoagulation has been tested in the order a) electrocoagulation followed by adsorption and b) adsorption followed by electrocoagulation.

2. Materials and methods

2.1. Raw materials

Peat obtained from Northern Finland has been used in the preparation of an aqueous solution containing humic material, activated carbon derived from residue fractions of lignocellulosic biomass; sawdust of spruce.

2.2. Methods

2.2.1. Preparation of activated carbons

The dried and sieved lignocellulosic biomass was carbonized and steam activated in a one-step process in a rotating quartz reactor (Nabertherm GmbH RSRB 80). The thermal process was divided into two parts: the first carbonization step, in which the temperature was raised to $800\text{ }^{\circ}\text{C}$, followed by the activation step. During the activation, the temperature was kept at $800\text{ }^{\circ}\text{C}$ for 120 min with a stream of steam, for the proper surface activation. During the whole process, the reactor was flushed with an inert gas, N_2 . Prepared AC was sieved to particle size to $0.4\text{--}2.0\text{ mm}$. By using Micromeritics ASAP 2020, the resulting AC was characterized by the specific surface area and pore size distribution under isothermal conditions. Specific surface areas were

calculated from adsorption isotherms according to the BET method [37] and pore size distributions were calculated using the BJH algorithm [38].

2.2.2. Total carbon determination

The content of carbon present in AC, given as total carbon (TC) percent, was measured using a solid phase carbon analyzer (Skalar Primacs MCS). Dried samples were weighted in quartz crucibles, combusted at $1100\text{ }^{\circ}\text{C}$ in an atmosphere of pure oxygen and the formed CO_2 was analyzed by an IR analyzer. Carbon content values were obtained by reading the signal of IR analyzer from a calibration curve derived from known masses of a standard substance, oxalic acid. The total mass of carbon was calculated as a percent of the mass initially weighted and was measured after the carbonization and activation step. The content of organic carbon present in each sample ($\pm 5\%$), given as total carbon (TC) mg L^{-1} , was measured using a liquid phase carbon analyzer (Skalar Primacs MCS). TC (Total carbon), TOC (Total organic carbon) and IC (Inorganic carbon) could be determined with the method.

2.2.3. Adsorptive properties

The formed AC's adsorption properties were tested with the adsorption capacity of total organic carbon. A solution containing a known amount of TOC was prepared, 50 mL of this solution was transferred into Erlenmeyer flask with a defined amount of activated carbon and the solution was continuously agitated for 24 h in order to achieve equilibrium between adsorption and desorption of the organic material. Portions of each solution were filtered and, if needed, diluted, and the concentration of TC, TOC and IC was measured with TOC analyzer. The adsorbed mass was calculated using Eq. (5) and the percent removed was calculated using Eq. (6)

$$(ads) = (C_0 - C_t) \cdot \frac{V}{m} \quad (5)$$

$$\%removed = \frac{C_0 - C_t}{C_0} \quad (6)$$

Where C_0 is initial concentration (mg L^{-1}), C_t is measured concentration after 24 h , V is the volume of the solution used and m is a mass of the activated carbon used.

2.2.4. Metal content

Metal contents of samples were measured by inductively coupled optical emission spectrometry (ICP-OES) using a Perkin Elmer Optima 5300 DV ICP-OES instrument. $0.10\text{--}0.12\text{ g}$ samples were added to 63% nitric acid and hydrogen peroxide, then digesting in a microwave oven (MARS, CEM Corporation) at $200\text{ }^{\circ}\text{C}$ for 10 min . After digestion, the solution was diluted to 50 mL and measured by ICP-OES.

2.3. Optimization of distance between anode and cathode

Aluminum and iron metals were used as an anode and stainless steel (SS) was used as the cathode. Electrode plate dimensions were $50 \times 50 \times 10\text{ mm}$. Electrodes were connected a laboratory electrical source (Tektronix DC Power Supply; 32 V , 6 A) and adjusted at 2 V and 1 A . The voltage was kept constant during the experiments. The distance between electrodes varies from 5 mm to 20 mm . During the coagulation experiments, the electrodes were completely immersed in the sample solution. Prior to experiments, both electrodes were cleaned with 1 M nitric acid, tap water and distilled water and finally, dried at $105\text{ }^{\circ}\text{C}$. Anode plates were weighted before and after EC experiments, which led to the discovery of the mass of dissolved anode metal. The number of metal cations dissolved during the electrocoagulation can be calculated according to Faraday's law. The experiments were carried out using 5 mm , 10 mm and 20 mm distance between anode and cathode. Fig. 1 illustrates a schematic diagram of the electrocoagulation.

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