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Energy efficient electrocoagulation using an air-breathing cathode to remove nutrients from wastewater

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HIGHLIGHTS

- First application of an air-breathing cathode for electrocoagulation (EC).
- Energy reduced for treatment due to lack of a need for wastewater aeration.
- High efficiency in the removal of nutrients from wastewater.
- Design allows for a compact, stackable and modular configuration.

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ABSTRACT

Electrocoagulation (EC) can be used to remove nutrients as well as suspended solids and organic carbon from wastewaters, but the high energy requirements (pumping air) and the use of precious metals in some processes has hindered widespread application of EC technologies. The use of a thermodynamically favorable activated carbon air cathode and a sacrificial aluminum anode was examined to reduce the energy needed for EC for nutrient removal. Performance of the air cathode electrocoagulation (ACEC) process was tested using raw wastewater, and a carbon-free synthetic solution (nitrogen:phosphorus ratio of 1:10 in deionized water) to simulate nutrient removal of a wastewater treated for organic matter removal. ACEC treatment of wastewater removed up to 99% of both ammonia and phosphorus, along with 72–81% COD and 78–89% TSS in 4 h (1.5 cm electrode spacing, current density of 8 A m^{-2}). Nearly total removal of the nutrients required 1.8 kWh m^{-3}, which is lower than that previously reported for some EC processes for this application (0.4–22 kWh m^{-3}). Nutrient removals using synthetic solutions (no organic matter) ranged from 74% to 93% for nitrogen (47–370 mg-N/L) and 44–76% for phosphorus. These results indicate that the ACEC can achieve good levels of nutrient removal with reduced energy demands compared to previous EC systems.

1. Introduction

In recent years, there has been great interest in the development of completely anaerobic treatment processes, such as anaerobic fluidized bed reactors, anaerobic membrane bioreactors [1–3] and microbial fuel cells (MFCs) [4]. While these processes can be used to remove organic matter, there is little nutrient (nitrogen and phosphorus) removal in these processes. Therefore, effective processes that can be coupled to these anaerobic systems are needed in order to achieve nutrient removal [5].

Electrocoagulation (EC) is an electrochemical process that can be used to treat wastewaters in terms of both organic matter and nutrient removal [6,7]. The process is based on the use of sacrificial electrodes such as aluminum, iron, and zinc [8–11] that are oxidized to produce metal ions that can be used to coagulate and react with organic matter and nutrients in the wastewater. When current is applied, the metal ions that are produced react with primary hydroxides and produce polyhyroxides and polyhydroxy-metallic flocs [12]. When aluminum is used as the sacrificial electrode material, the primary chemical species that are produced under typical alkaline (pH of 7.5–9) or acidic (pH 3.5–6) conditions at the anode (20 °C) are:

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$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3 \quad \text{(Alkaline pH)} \quad (1)$$

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad \text{(Acidic pH)} \quad (2)$$

where applied voltages are typically <0.81 V, and the temperatures are usually in the range of 25–50 °C reported. The flocs that form have a large surface area that can adsorb soluble organic compounds and trap colloidal particles in the wastewater [10]. Various metals have been examined to improve process performance [13], but commonly aluminum or iron is used [8,14–18] due to the good performance of these metals for domestic and textile wastewaters, as well as for pesticides and arsenic [18–20].

While EC processes are effective for treatment of many types of wastewaters, applications have been limited due to high energy demands and high current densities. Examples include: oil in wastewater at 25 mA cm^{-2} [21] and 4–30 mA cm^{-2} [22], berberine hydrochloride removal from a pharmaceutical factory wastewater at 19 mA cm^{-2} [23], food wastes from restaurants at 30–80 A m^{-2} [24], 50–200 A m^{-2} for hexavalent chromium treatment in an industrial wastewater [25], heavy metal ions removal from a metal plating wastewater at 4 mA cm^{-2} with 6.3 kWh m^{-3} energy consumption [26]. EC has also been used for drinking water treatment to remove hardness and fluoride, but at very high applied voltages of 24 V, with 22 A m^{-2} and a treatment time of 1 h [27,28]. The advantage of the EC process is that treatment can be accomplished without the need for additional chemicals or pre-treatment, but energy costs need to be reduced for EC processes to become more applicable.

The cathodic reaction in an EC process is usually hydrogen evolution, or oxygen reduction supported by wastewater aeration. Both of these approaches require energy that is not intrinsically part of the treatment process. Hydrogen evolution is thermodynamically unfavorable, and requires a cathode potential of −0.414 V under standard conditions [29]. While oxygen reduction is thermodynamically favorable, the production of dissolved oxygen requires aeration, which is an energy consuming process. For example, wastewater aeration in an activated sludge process consumes about 0.6 kWh m^{-3} [30]. Air cathodes are commonly used in many types of electrochemical systems such as hydrogen fuel cells and microbial fuel cells (MFCs) [31,32], to provide oxygen to the cathode. The use of an air cathode avoids energy losses due to aeration, while providing a favorable reaction at the cathode. However, air cathodes have not been previously examined for nutrient removal from wastewater using EC systems.

Air cathodes were examined here as a method to reduce electrochemical energy requirements for nutrient removal from wastewater using an air cathode electrocoagulation (ACEC process). Air cathodes developed for MFC applications do not need to use precious metal catalysts. Instead, inexpensive activated carbon can be used, with the cathode made using various press or rolling methods [33,34]. The impact of the reactor configuration, such as electrode spacing, operational conditions, and treatment time, were examined using two solutions: domestic wastewater, which contains high concentrations of residual organic matter; and a simulated treated wastewater, prepared by adding ammonia and phosphorus to deionized water, to examine the impact on nutrient removal in the absence of organic matter. Aluminum mesh anodes were used to produce a higher active surface than flat plate electrodes. Power requirements were examined for different operational conditions to achieve high nutrient removals while minimizing power consumption.

2. Materials and methods

2.1. Reactor construction

Cube-shaped ACEC reactors were constructed based on a previous design [35] and contained a 3 cm diameter electrolyte chamber and an air cathode. Reactor lengths of 0.5, 1, 1.5, and 2 cm were used to examine the impact of electrode spacing on performance, with the electrodes placed on opposite ends of the chamber (Fig. 1). The reactive surface area of each electrode was 7 cm^2. The anode was a single piece of aluminum mesh (mesh size 200 per 2.54 cm, wire diameter 0.05334 mm, opening 0.07366 mm; TWP Corporation). Cathodes contained an activated carbon catalyst, and were made by a continuous rolling and press process, using polytetrafluoroethylene (PTFE) as the binder, and a PTFE/Carbon black diffusion layer to avoid water leakage [36].

2.2. Solutions

Wastewater was collected from the primary clarifier of the Penn State Wastewater Treatment Plant and stored at 4 °C. The wastewater had a pH of 7–7.2, and contained 490 ± 10 mg L^{-1} COD, 200 ± 6 mg L^{-1} TSS, an ammonia concentration of 52 ± 4 mg-N L^{-1}, and a total phosphorus concentration of 3 ± 0.1 mg-P L^{-1}. The ratio of N:P in this wastewater was 17:1, and many wastewaters often have a total nitrogen concentration about ten times that of phosphorus. The solution used to simulate treated wastewater was prepared by adding NH_4Cl and $NaH_2PO_4{\cdot}H_2O$ (Sigma Aldrich) to deionized water at an initial N:P ratio of 10:1, at four different

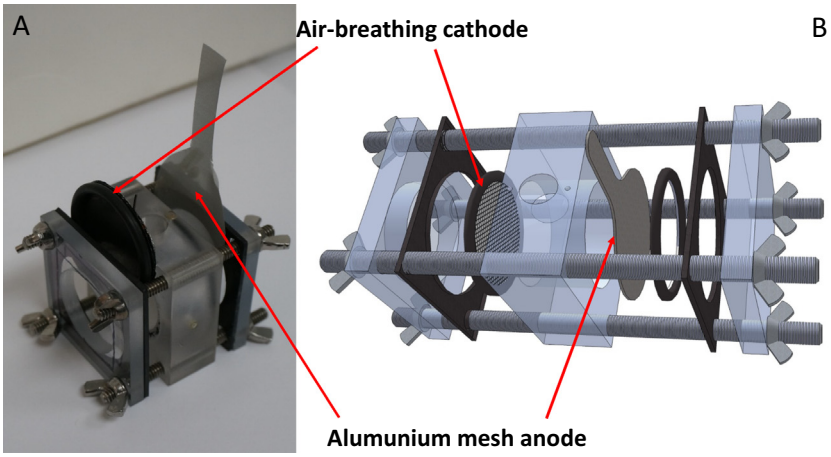

Fig. 1. (A) Schematic diagram and (B) photo of the electrocoagulation reactor with an air cathode.