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A novel bio-electrochemical system with sand/activated carbon separator, Al anode and bio-anode integrated micro-electrolysis/electro-flocculation cost effectively treated high load wastewater with energy recovery

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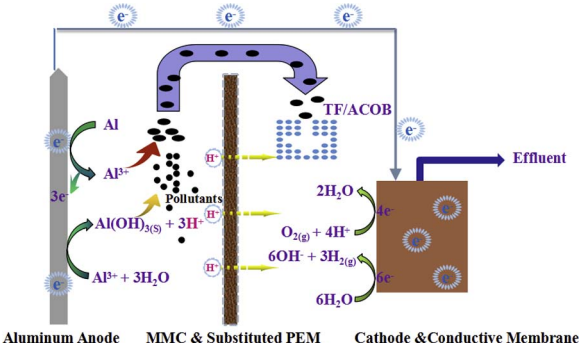
GRAPHICAL ABSTRACT



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ABSTRACT

A novel bio-electrochemical system (BES) was developed by integrating micro-electrolysis/electro-flocculation from attaching a sacrificing Al anode to the bio-anode, it effectively treated high load wastewater with energy recovery (maximum power density of 365.1 mW/m^3 and a maximum cell voltage of 0.97 V), and achieving high removals of COD (> 99.4%), NH_4^+-N (> 98.7%) and TP (> 98.6%). The anode chamber contains microbes, activated carbon (AC)/graphite granules and Al anode. It was separated from the cathode chamber containing bifunctional catalytic and filtration membrane cathode (loaded with Fe/Mn/C/F/O catalyst) by a multi-medium chamber (MMC) filled with manganese sand and activated carbon granules, which replaced expensive PEM and reduced cost. An air contact oxidation bed for aeration was still adopted before liquid entering the cathode chamber. micro-electrolysis/electro-flocculation helps in achieving high removal efficiencies and contributes to membrane fouling migration. The increase of activated carbon in the separator MMC increased power generation and reduced system electric resistance.

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1. Introduction

Micro-electrolysis is an electrochemical wastewater treatment technique based on metal corrosion. Being a simple process with small footprint, with low equipment investment and low operating cost, but good treatment performance, it thus has attracted more and more attentions (Han et al., 2016). Recently, micro-electrolysis tests had been extensively conducted in treating various industrial waste waters, from nonferrous and ferrous metallurgy to electronics and electroplating, from chemical and pharmaceutical to petroleum processing, printing and dyeing and so on (Wang et al., 2017).

Micro-electrolysis does not require external power supply, in wastewater, the Al/carbon (or iron/carbon) particles form electrode pairs and micro-batteries, achieving catalytic wastewater treatment (Xing et al., 2016). The pollutants are removed by mechanisms involving galvanic cell and redox reactions, flocculation, adsorption and co-precipitation. The electrode reactions produce high activity species, react and remove pollutants in wastewater, decompose macromolecular components into intermediates or small molecules for enhanced biochemical conversion and enhanced biodegradability (Deng et al., 2017).

Another widely used electrochemical process in wastewater treatment is electro-flocculation (EF). Since its concept was put forward for the first time in 1901. Without adding chemicals, it is advantageous in easy operation and combination with other technology. Its main industrial applications are in treating heavy metal wastewater, inorganic and organic wastewater, and smelting/mining wastewater, electroplating and oilfield waste waters (Ben Sasson and Adin, 2010).

During EF, fresh species with flocculation characteristics are produced from the sacrificial anode such as Fe or Al, also polynuclear hydroxyl complex are formed after hydrolysis and polymerization (Lakshmanan et al., 2009). Anodic removal of pollutants in EF can be realized by adsorption, coagulation and precipitation. Cathodic reduction reactions produce small hydrogen bubbles with good adhesion properties, thus help up-flow of air flotation suspensions to water surface, and remove refractory pollutants. Also hydrophobic colloids, as well as hydrophilic colloidal substances, which are difficult to remove by traditional flocculation methods, can be removed. It is effective for natural organic matter (NOM) removal (Matilainen et al., 2010).

But, EF alone cannot achieve high quality water effluent (Bocos et al., 2016). Recent focus is combining EF with membrane filtration. Pretreatment by EF and followed membrane filtration can efficiently remove bacteria, heavy metals and colloidal silicon. EF positively reduces membrane fouling (Ben Sasson et al., 2011). However, required external power supply limits its application, especially in areas with power shortage, such as remote mountainous areas where electricity is not readily available (Kim et al., 2017).

MFC output power was lower than other fuel cells (Logan et al., 2015). But most promising and green technology for lowering energy consumption in wastewater treatment, microbial fuel cells (MFCs) were popular with researchers, for economic and efficient energy production, if only significant reduction of equipment and electrode costs is possible, which is also crucial for application of MFC.

Bio-electrochemical system (BES) integrating microbial fuel cell with electric membrane bioreactor can not only convert biomass into electrical energy, but also optimize effluent quality (Akamatsu et al., 2010; Gao et al., 2017; Khalid Bani-Melhem, 2011). Separator use in BES is important for separating anode chamber and cathode chamber, transferring protons from the anode chamber into the cathode chamber, and preventing the permeation of dissolved oxygen (DO) from the cathode chamber to the anode chamber (Zhang et al., 2016). Proton exchange membrane (PEM) has been widely used for these purposes. Compared with other separation materials, e.g., cation exchange membrane, ultrafiltration membrane and/or ceramic membrane, PEM such as Nafion had become indispensable because of its low internal resistance and high ionic conductivity. However, its use is limited because the PEMs made of Nafion are very expensive, have very common biological pollutions accompanying long-term operation, the consequences of pollution can lead to a dramatic deterioration in the performance of BES, as well as a substantial increase in operating costs (Xu et al., 2012).

The development of materials replacing PEM can not only greatly reduce the cost, but also can promote the scale-up and application (Logan, 2010). Previous researches from our team have shown that PEM can be replaced by cheap materials such as quartz sand chamber (QCS) (Gao et al., 2017), without compromising the electrochemical performance.

Activated carbon and other cost-effective materials have physical and chemical characteristics, such as better conductivity and active electrochemical properties. Using activated carbon and sand replacing PEM may lower internal electric resistance. However, this has not been attempted or reported in any previous reports. This assumption need to be tested and validated.

In this study, we developed BES integrating micro-electrolysis and electro-flocculation by using an attached Al anode in integrated MFC and MBR, kept an air contact oxidation bed (ACOB) and trickling filter (TF) with gravitational flow. We used activated carbon and graphite as bio-anode, and a dual function conductive membrane previously developed as cathode, which simultaneously functioned as filter media in MBR. Replacing PEM, low cost quartz sand or Mn-sand and inexpensive activated carbon was used in a separator called multi media chamber (MMC). The purpose, aiming at solving the environmental problems economically and efficiently, was realized achieving efficient treatment of high load wastewater, by integrating micro-electrolysis, electro-flocculation and bioelectrochemistry.

2. Materials and methods

2.1. MFC with multi media chamber separator

In order to facilitate scale-up and practical applications, low-cost and cheap materials were adopted. The PEM-free MFC has three chambers, the anode chamber, the multi medium chamber (MMC) and the cathodic chamber. A pop can was connected to the bio-anode of the MFC. Before being used, the aluminum anode was polished to remove the oxide layer and liner layer. The anode chamber was filled with graphite and activated carbon mixed particles (particle size 3–5 mm, volume ratio 1:1). The net volume of the anode chamber was 4 L (10 cm × 10 cm × 40 cm), the filling rate of particles was 100%. To ensure effective contact between the particles and the aluminum anode, the bottom of the Al can was cut and inserted into the particle, the top of Al can was also partly cut and exposed above the liquid surface. The reference electrode and the aluminum anode were respectively connected to the data logger (PISO-813, Taiwan) using copper wires.

The top of the anode chamber was sealed. For plug flow operation, high load synthetic wastewater entered the anode chamber from the bottom, then overflowed at the top and dropped through nozzles into the ACOB and TF, then again overflowed into the cathode chamber (Gao et al., 2017). This design has enhanced removal of the organic compounds and reduced the cathodic aeration cost.

Traditional expensive PEM was not used in this experiment. One MMC was designed replacing PEM and separating the anode chamber and the cathode chamber. The volume of MMC was 1 L (5 cm × 10 cm × 20 cm) filled with mixed manganese sand/quartz sand and activated carbon (particle size 0.5–1 mm) with different ratios (100% of filling rate). This proton channel (21 cm^2 in area, 7 cm × 3 cm) allowing effective proton transfer was maintained. To prevent the leakage of mixing particles from this channel, two sealing flanges and non-woven fabrics were fixed across the channel.

In the cathodic chamber, a novel bifunctional conductive membrane was used (with Fe/Mn/C/F/O catalyst, 2–7 nm membrane pore size) as cathode of MFC and the membrane of MBR. The total surface area of the