



# Enhanced separation performance of coal-based carbon membranes coupled with an electric field for oily wastewater treatment



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## ABSTRACT

Coal-based carbon membrane coupled with an electric field is designed to achieve enhanced separation performance for oily wastewater treatment in this study. Effect of electric field intensity, concentration and pH of oily wastewater, rotate speed of peristaltic pump, electrolyte concentration, and electrode distance on separation performance of carbon membrane are investigated. The morphologies of carbon membranes are examined using scanning electron microscope (SEM). Fouling analysis is also carried out for further evaluating the antifouling ability of coal-based carbon membrane. The results demonstrate that coal-based carbon membranes integrated with an electric field show improved permeate flux and removal efficiency for oily wastewater treatment due to anodic oxidation. No obvious oil foulants are observed on carbon membrane by SEM images. Low total fouling ratio (TFR) and high flux recovery (FR) imply that exerting an electric field can significantly improve antifouling ability of carbon membrane. Acidic condition is benefit for carbon membrane to possess good fouling resistance to oil droplets. A decrease in electrode distance improves the separation performance of the treatment system. The optimum operation conditions of 0.31 V/cm electric field intensity, 7.5 r/min pump rotate speed, and 5 g/L electrolyte concentration are recommended. After cleaning, carbon membrane coupled with an electric field still demonstrates great potential in oily wastewater treatment.

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## 1. Introduction

Nowadays, large quantities of oily wastewater are generated from various industries including petrochemical, steel, leather, textile, and transportation, etc., which are considered as the major threat to aquatic environment [1–4]. This ecologically hazardous oily wastewater must be treated before being discharged according to related environmental regulations [5–7]. Up to date, several conventional techniques, including gravity separation, air flotation, chemical de-emulsification, coagulation, flocculation, and biological treatment, etc., have been used for oily wastewater treatment [8]. However, these techniques have several limitations such as more time consumption, low efficiency, high operation costs, and not effective on treating tiny oil droplets. These disadvantages prompt many researchers to turn to develop novel treatment technology for oily wastewater [9,10].

Among large numbers of emerging methods on oily wastewater treatment, membrane separation gets extensive concern as a

promising technology due to the advantages of compact design, small space occupancy, simple operation process, low energy consuming, high retention ratio and easy control of membrane properties [11,12]. Also, another important reason to choose the technology is that it can purify the oily wastewater containing oil droplets smaller than 20  $\mu\text{m}$  with acceptable discharge quality [13]. To my knowledge, investigation on the treatment of oily wastewaters using membrane separation processes has been started since 1976 [14]. After that, a lot of related works using various membranes processes, such as nanofiltration (NF), ultrafiltration (UF) and microfiltration (MF), etc., are reported [15–17]. These studies reveal membrane separation technology possesses high oil removal efficiency compared to conventional treatment methods. However, as approved by many researchers, membrane separation performance is often deteriorated by membrane fouling, resulting from deposition or adsorption of foulants on membrane surface and/or inside pores during filtration, which has become one of the major obstacles greatly limiting the commercialization of membrane technology [18].

To minimize membrane fouling, various approaches have been adopted in previous literatures, which were classified into four categories: boundary layer (or velocity) control; turbulence inducers/generators; membrane modification and materials, and combined

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(external) fields [19–21]. Among them, applying an electric field to reduce membrane fouling is found to be quite effective due to the electrophoretic forces induced by the electric field can prevent foulants from depositing on the membrane [22,23]. Moreover, some adsorbed substances may be electrochemically degraded. The idea inspires many researchers to use an external electric field to improve separation efficiency during membrane process. In their previous works, two different configurations have been reported. One is that an electric field is applied across the membrane. This configuration usually suffers from low efficiency due to the placement of an anode and a cathode located on opposite sides of the membrane. Also, extra electrodes will increase commercial implementation and reactor volume. The other is that the electric field is applied between the membrane (as an electrode) and another electrode. The configuration thus requires less energy in order to obtain the same electric field intensity. Therefore, developing novel materials which not only behave as membranes but also will have the ability to conduct electricity will favor the successful application of the electric field enhanced antifouling technology. Huotari et al. used a carbon fiber-carbon composite membrane as a cathode to treat oily wastewater, and demonstrates significant improvement in flux and permeate quality [24]. Dudchenko et al. synthesized a conductive thin film made of cross-linked poly(vinylalcohol) and carboxylated multi-walled carbon nanotubes as a cathode to treat alginate acid. Significant fouling inhibition was observed due to electrostatic repulsion, which prevented alginate acid from interacting with the membrane surface [25]. Akamatsu et al. developed a novel membrane-carbon cloth assembly for submerged MBRs by applying an electric field intermittently to suppress fouling, in which low cost carbon cloth was adopted as electrodes [26]. These studies demonstrate that carbon materials have great potential as electrodes on electric field enhanced antifouling system due to their electrical conductivities.

Carbon membranes are novel porous inorganic membranes, which are usually prepared by pyrolysis of carbonaceous materials, such as polyimide and derivatives, polyacrylonitrile (PAN), phenol formaldehyde (PF), and poly(furfuryl alcohol) (PFA) [27]. In our previous work, we developed novel carbon membrane derived from coal, which exhibit great potential on wastewater treatment [28–30]. Moreover, coal-based carbon membrane possesses good electrical conductivity, which makes it be a good candidate as

electrode materials on the antifouling system for wastewater treatment. In this work, we design an antifouling treatment system, where coal-based carbon membrane is adopted as the anode and coupled with an electric field for oily wastewater treatment. The influence of an electric field on the improved separation performance of carbon membrane is studied in detail. Fouling analysis is also carried out to further approve the enhanced antifouling ability of carbon membrane under the electric field. Moreover, other operating conditions such as concentration and pH of oily wastewater, rotate speed of peristaltic pump and electrolyte concentration are investigated and optimized.

## 2. Materials and methods

### 2.1. Properties of coal-based carbon membrane

Tubular coal-based carbon membrane with average pore diameter of  $0.382\ \mu\text{m}$  and porosity of 49.56% is used in this study. The inner diameter and membrane area are 8.5 mm and  $0.0024\ \text{m}^2$ , respectively. The preparation process of coal-based carbon membrane refers to our previous literatures [29].

### 2.2. Treatment of oily wastewater

Oily wastewater is obtained by mixing 180# fuel oil, distilled water and surfactant (sodium dodecylbenzene sulfonate (SDS)) for 8 h at a speed of 5000 rpm using a homogenizer. Sodium sulfate ( $\text{Na}_2\text{SO}_4$ ) is added into oily wastewater as electrolyte. Sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and sodium hydroxide (NaOH) are used to adjust the pH value of the oily wastewater [31].

Fig. 1 shows the schematic of the treatment system. The tubular carbon membrane connected with a DC power is placed in the center of the container as the anode, and the titanium plate surrounding the membrane as the cathode. The electric field intensities are set to 0.15, 0.23, 0.31, 0.38 and 0.42 V/cm, respectively. During treatment process, permeate samples are collected at different time for further analysis.

The permeate flux is an essential parameter to evaluate the fouling status of carbon membrane, which is calculated according to the following equation:

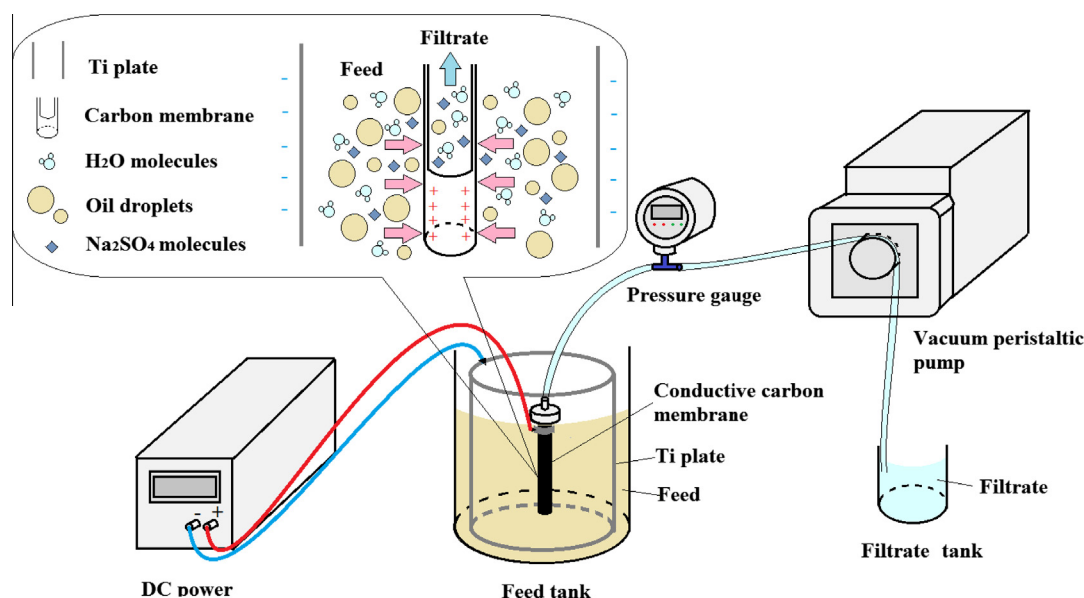


Fig. 1. Flow schematic diagram of carbon membrane coupling with an electric field.

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