



Designing fouling-resistant clay-embedded polyelectrolyte multilayer membranes for wastewater effluent treatment[☆]



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ABSTRACT

This work explores the applicability of clay-polyelectrolyte based hybrid thin films to develop fouling resistant membrane surfaces for wastewater treatment applications. Clay nanoplatelets were layered on a commercial polyethersulfone membrane in conjunction with two oppositely charged polyelectrolytes via the aqueous-based layer-by-layer (LbL) assembly technique. These hybrid nanostructured membranes showed a high degree of fouling resistance as compared to other commercial membranes and the pure polyelectrolyte multilayer (PEM) membranes, when tested against an electrocoagulation-treated high strength wastewater. With the deposition of just 2.25 quadlayers, the clay-PEM (c-PEM) membranes demonstrated good anti-fouling properties. On crosslinking the polyelectrolytes, the c-PEM hybrid membranes showed higher reduction in the chemical oxygen demand (COD) value and enhanced fouling resistance as compared to their uncrosslinked counterparts, the pure PEM membranes (both uncross linked and crosslinked) and the bare membrane. However, the high fouling resistance of the c-PEM membranes was attained at the cost of compromising the high initial flux value of the underlying membrane. Several possible optimization strategies have therefore been suggested in this paper, which can potentially increase the flux of the modified membranes. This work, for the first time, demonstrated an attempt to evaluate the performance of clay-polyelectrolyte nanocomposite membranes against a real wastewater effluent.

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1. Introduction

The shortage of drinkable water has been one of the most pressing issues of the 21st century. The worldwide depletion in fresh water resources has created the need to purify other sources of water (like sea water) which are present in huge abundance. While desalination has grown to be a big area, the recycling and reuse of wastewater has also gained considerable attention. Various membrane based processes like ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have been proposed and used for treating many different types of water sources [1–3]. While membrane technology has witnessed tremendous development over the past few decades, its application in many cases is still challenged by the issue of membrane fouling. Fouling accounts for a significant flux decline over a period of time that leads to higher pressure requirements as well as higher costs involved in

membrane cleaning, replacement etc. Several research efforts have been directed towards mitigating membrane fouling like introducing changes in the process [4], bulk modification [5] and surface modification [6,7]. Surface modification techniques have garnered considerable interest among researchers because of economical merits and ease of processing. In this work, layer-by-layer (LbL) assembly was explored as the surface modification process. It is a thin film deposition technique which involves the layering of polyelectrolytes /colloidal particles/nanomaterials via secondary molecular interactions like ionic attraction, hydrogen bonding, hydrophobic-hydrophobic interaction etc. [8]. LbL provides an effective way to tune the surface by manipulation of a number of parameters in order to satisfy the application requirements [9]. It is of general consensus that changing the physico-chemical properties of the membrane surface like increasing the hydrophilicity and surface charge help in reducing the fouling propensity of that membrane [10]. Polyelectrolyte multilayer (PEM) membranes fabricated by the LbL assembly of polyelectrolytes on commercial membrane surfaces, therefore can well satisfy the criteria for being an effective modification process related to water treatment applications.

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PEM membranes have been widely applied as high flux alternatives to commercial salt rejection membranes and have shown their potential in removing many different types of ions [11–15]. However, most of these studies were confined to some simple ion-rejection tests. While most of these studies have focused on the aspects of ion rejection and ion selectivity, only a couple of studies were done on real wastewater effluents [16,17]. Very recently we published our work on extending the application of PEM membranes to electrocoagulation-treated high-strength wastewater effluent [18]. This work showed that PEM membranes are capable of reducing the chemical oxygen demand (COD) value of the effluent to the same level as commercial RO membranes but with much higher flux values. These membranes also exhibited superior anti-fouling properties than commercial membranes, when tested under dead end conditions. In this work, we focus on evaluating the anti-fouling property of these membranes with a similar effluent under cross flow conditions.

In addition to the PEM membranes, the effect of introducing nanomaterials within the LbL films was also studied. In previous studies, nanomaterials like graphene [19,20], carbon nanotubes [21,22] and clay [23–26] have been applied to fabricate or modify membranes. Among the many different nanomaterials, clay is easily dispersible in water and therefore can be incorporated within the LbL assembly without any additional functionalization. It is cheap, easily available and imparts hydrophilicity to the surface. It therefore suits the requirement of a typical surface modifier which can be used for developing large scale anti-fouling membranes. The use of clay within PEM films has been studied in great details by Grunlan et al., to fabricate gas barrier films [27–29]. The introduction of clay increased the tortuosity of the gas under consideration [30, 31]. The application of the clay-PEM hybrid films to water treatment membranes, has been comparatively rare. A couple of studies reported depositing such films on UF membrane surfaces to develop ion-rejecting membranes. Jungkyu et al., used these hybrid films for salt rejection and the incorporation of clay nanoplatelets rendered the membrane more chlorine resistant [23]. Very recently, Kovacs et al. also worked on the spray assembly of clay-polyelectrolyte quadlayers on polyethersulfone membranes for fabricating desalination membranes with higher flux than commercial RO membranes, albeit with equal salt rejection [24,25,30]. In fact, their work encompassed the tuning of several factors to optimize the hybrid film structure as selective layers for ion rejection and provides an excellent methodology to identify the best suited system. However, the films developed by them were only tested with a certain ion and under dead end conditions which represent markedly different conditions than testing a real effluent under tangential flow conditions. So far, these clay-PEM modified membranes have not been used with any wastewater effluent and thereby not evaluated based on their anti-fouling property. However, Hang et al., developed a composite clay-PES membrane whereby they attained higher fouling resistance with the addition of clay [26]. With this background, we believed it would be interesting to co-deposit clay nanoplatelets with polyelectrolytes using LbL and evaluate the performance of the modified membranes against the wastewater effluent.

In this work, the effluent from an Anaerobic Digestion (AD) reactor was fed to an electrocoagulation (EC) unit. The EC step served as a pretreatment stage for the membrane separation process. Based on our knowledge about the nature of this effluent, as reported in our previous publication [18], it contains humic compounds which are responsible for the organic fouling. To counter the fouling propensity of commercially available membranes, a polyethersulfone membrane was chosen as the substrate for the deposition of PEM films and clay-PEM nanocomposite films. While the main focus of this work was evaluating the flux and fouling behavior of the modified membranes, we have also

shown some results related to the COD removal capability of these membranes. Poly allylamine hydrochloride (PAH) and poly acrylic acid (PAA) were chosen as the polyelectrolytes and montmorillonite (MMT), as a member of the smectite group of clay minerals, was used to further modify the PEM membranes.

2. Experimental section

2.1. Materials

Poly (allylamine hydrochloride) (MW 900,000) and poly (acrylic acid sodium salt) (MW 15,000, 35 wt% in water) were procured from Sigma Aldrich. Sodium-Montmorillonite (MMT- Na^+) clay (commercial name: Cloisite- Na^+) was obtained from BYK Additives Inc. (Gonzales, TX). 2-(N-Morpholino) ethanesulfonic acid (MES) Buffer was also purchased from Sigma Aldrich. N-(3-Dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride (EDC) was purchased from Fisher Scientific. The commercial membranes NF 270 and BW 30 were purchased from Dow Filmtec (Midland, MI). The polyethersulfone membranes (MWCO 10,000 UE-10, Tri Sep) were provided by Sterlitech (Kent, WA). All aqueous solutions were prepared using deionized (DI) water ($> 18.2 \text{ M}\Omega$) supplied by a Barnstead Nanopure Diamond-UV purification unit equipped with a UV source and a final $0.2 \mu\text{m}$ filter. Unless specified otherwise all procedures were carried out at room temperature.

2.2. Cross-flow filtration process setup

A CF 042 cross flow unit (Sterlitech, Kent, WA) with an effective surface area of 42 cm^2 was employed as the membrane module. The cross flow module has a rectangular geometry, the flow channel being 3.62" in length, 1.8" in width and 0.09" in depth. A positive displacement pump (Hydra-cell M03, Wanner Engineering, Minneapolis, MN) was used to deliver the feed from a conical 5-gallon feed tank. A variable speed drive (Emerson, St Louis, MO) was attached to the pump which controlled its speed. A part of the feed stream was directed back to the feed tank via a bypass valve. A back-pressure regulator was used to control the transmembrane pressure across the membrane module. The retentate stream was recycled back to the feed tank and its flow rate was measured by a Site Read Panel Mount Flowmeter (Blue-White, Huntington Beach, CA). The permeate stream was also recycled back to the feed tank. The temperature of the solution inside the feed tank was maintained at room temperature by a digital chiller (Polysciences, Warrington, PA). All the components of the cross-flow setup were obtained from Sterlitech (Kent, WA) and assembled in the lab. A detailed diagram of the setup can be found in our previous publication [15], the only difference in this case being the recycling of the permeate stream back to the feed tank.

2.3. Effluent characterization

The COD measurements are done following the standard procedure using COD kits purchased from HACH (Loveland, CO). All the three types of kits (ultra-low range, low range and high range) were employed based on the COD level of the sample. A HACH reactor was used to digest the samples and the values were read using a direct reading spectrophotometer.

2.4. Experimental protocols

2.4.1. Electrocoagulation (EC) protocol and preparation of the feed solution for the membrane processes

AD effluent was obtained from the Anaerobic Digestion Research and Education Center (ADREC) at Michigan State University.

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