



Thermodynamic assessment of adsorptive fouling with the membranes modified via layer-by-layer self-assembly technique

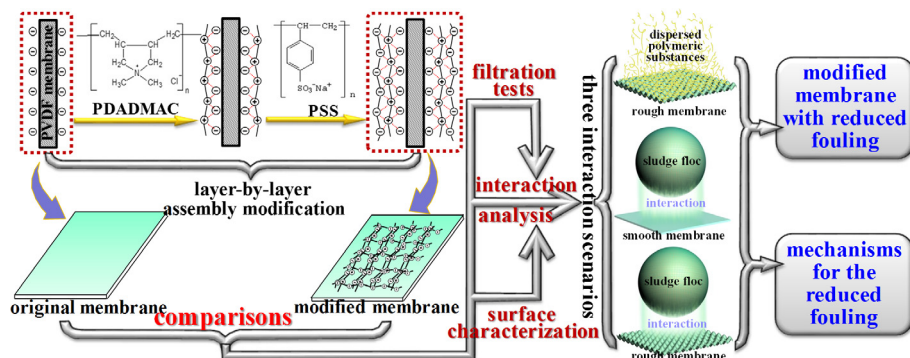


Liguo Shen^a, Xia Cui^a, Genying Yu^a, Fengquan Li^a, Liang Li^b, Shushu Feng^a, Hongjun Lin^{a,*}, Jianrong Chen^{a,*}

^a College of Geography and Environmental Sciences, Zhejiang Normal University, Jinhua 321004, PR China

^b Department of Civil and Environmental Engineering, University of South Carolina, 300 Main Street, Columbia, SC 29208, USA

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 30 November 2016

Revised 8 January 2017

Accepted 16 January 2017

Available online 18 January 2017

Keywords:

Membrane fouling
Membrane modification
Interfacial interaction
Layer-by-layer assembly
Membrane bioreactor

ABSTRACT

In this study, polyvinylidene fluoride (PVDF) microfiltration membrane was coated by dipping the membrane alternately in solutions of the polyelectrolytes (poly-diallyldimethylammonium chloride (PDADMAC) and polystyrenesulfonate (PSS)) via layer-by-layer (LBL) self-assembly technique to improve the membrane antifouling ability. Filtration experiments showed that, sludge cake layer on the coated membrane could be more easily washed off, and moreover, the remained flux ratio (RFR) of the coated membrane was obviously improved as compared with the control membrane. Characterization of the membranes showed that a polyelectrolyte layer was successfully coated on the membrane surfaces, and the hydrophilicity, surface charge and surface morphology of the coated membrane were changed. Based on the extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) approaches, quantification of interfacial interactions between foulants and membranes in three different scenarios was achieved. It was revealed that there existed a repulsive energy barrier when a particle foulant adhered to membrane surface, and the enhanced electrostatic double layer (EL) interaction and energy barrier should be responsible for the improved antifouling ability of the coated membrane. This study provided a combined solution to membrane modification and interaction energy evaluation related with membrane fouling simultaneously.

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* Corresponding authors.

E-mail addresses: jaylin1978@163.com (H. Lin), cjr@zjnu.cn (J. Chen).

1. Introduction

Membrane fouling contributes to enormous cost, and hence becomes the bottleneck for the large-scale industrial application of membrane separation technology [1–3]. In order to attenuate the membrane fouling problem, significant efforts have been executed to fabricate excellent performance membranes [3–5]. The approaches which have been explored include optimizing the membrane pore or surface structures by controlling the membrane preparation conditions [6,7], chemical grafting of hydrophilic functional monomers [8,9], and organic-inorganic or organic-organic blending [10,11]. Among them, layer-by-layer (LBL) self-assembly technique has been frequently used to coat advantageous polyelectrolytes on membrane surface for membrane fouling mitigation [12–14] due to its high efficiency and the ability to regulate the coated layer structure and composition [15,16]. It was reported that, the electronegative and hydrophilic polyelectrolyte networks could be easily coated on membrane surfaces by this technique [17,18]. For example, Su et al. [19] prepared a polyelectrolyte ultrafiltration (UF) membrane with the materials of poly(acrylonitrile and 2-dimethylaminoethyl methacrylate) (PAN-DMAEMA). The modified membrane showed excellent antifouling property to bovine serum albumin (BSA), and lysozyme in certain range of pH and ionic strength. Zhao et al. [20] prepared poly(vinylidene fluoride) (PVDF) UF membranes by assembling chitosan (CS) and sodium alginate (SA) polyelectrolytes. Under the optimized experimental conditions, BSA adsorption on the membrane surface is only $4 \mu\text{g cm}^{-2}$ and the flux recovery ratio after washing reaches to 89%, 99% and 98% for the three typical pollutants of BSA, SA and humic acid (HA), respectively. Ilyas et al. [21] modified the UF membrane by alternately assembling Poly(allylamine hydrochloride) (PAH) and poly(acrylic acid) (PAA). The modified membranes were much easily recovered by washing after fouled. These results strongly suggested that membrane surface properties could be tailored by coating different polyelectrolytes with LBL self-assembly technique, which is flexible for membrane modification to meet various demands. The polyelectrolytes containing $-\text{SO}_3^-$ functional group have been proved to be an efficient hydrophilic and negatively charged functional group in lots of literature studies [22–24]. While the experimental results in literature confirmed the improved antifouling ability of the coated membranes, its underlying causes have not been well investigated. A better understanding of mechanisms regarding antifouling ability is essential to optimize LBL self-assembly technique as well as prepare membranes with high antifouling property.

Previous studies generally reported that the reduced fouling of the coated membrane was mainly caused by the reduced adsorption of foulants on membrane surface [19–21,25]. Meanwhile, it is widely accepted that thermodynamic/interfacial interactions between foulants and membrane surface are critical predictors for the susceptibility of a membrane to foulants adsorption [26–28]. Interfacial interactions between two solid substances in water can be described by the extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory [29–31]. In this context, XDLVO theory may provide a probable way to explore the underlying mechanisms of the improved antifouling performance of the coated membranes by LBL self-assembly technique. Nevertheless, no specific study has been conducted to investigate this issue for LBL self-assembly technique.

The objective of this study is, therefore, to modify PVDF membrane by coating PDADMAC and PSS polyelectrolytes alternatively through LBL self-assembly technique, and to assess the interfacial interactions between foulants and the modified PVDF membrane. The interfacial mechanisms for the antifouling property of the coated membranes were investigated based on XDLVO theory and its extensions.

2. Materials and methods

2.1. Modification of membranes

The PVDF membrane (Pore size $0.1 \mu\text{m}$, Jiangsu Dafu Co. Ltd.) was alternately immersed into polyelectrolyte solutions for LBL assembling. It is known that PDADMAC and PSS are the typical positively charged and negatively charged polyelectrolytes, respectively. Since the PVDF membrane was negatively charged ($-21.19 \pm 1.67 \text{ mV}$) in this study, the assembling was initiated by the positively charged PDADMAC solution (1.00 g L^{-1} , MW = 100,000–200,000 Da, Aldrich Chemical Co. Inc). The PVDF membranes were immersed into the PDADMAC solution for 1 min to obtain the positively charged PVDF-PDADMAC membranes. After 5 min rinsing by water to remove the superfluous PDADMAC, the positively charged PVDF-PDADMAC membrane was immersed into the negatively charged PSS solution (1.00 g L^{-1} , MW = 70,000 Da, Aldrich Chemical Co.) for 1 min to obtain the negatively charged PVDF-PDADMAC-PSS membrane. By the electrostatic interactions and van der Waals forces, PSS was adhered onto the surface to form the negatively charged PVDF-PDADMAC-PSS membrane. Another rinsing by water was lasted for 5 min to remove the superfluous PSS. To minimize the flux decline, the number of bilayers was limited to 1 via the reported strategy [32]. In this study, the negatively charged membrane was alternately assembled with positively charged PDADMAC and then negatively charged PSS. The coated PDADMAC together with PSS means one bilayer. The negatively charged PVDF-PDADMAC-PSS membrane was then used for characterization, filtration, and antifouling experiments. The coating process is shown in Fig. 1.

2.2. Analytical methods

Surface of the membranes was measured using a scanning electron microscope (LEO1530vp SEM (Germany)). Previous to observation, samples were attached by carbon tape to the sample stage and sputtered with gold. The voltage was set at 25 kV, and the current was set at 10 mA. Chemical components of the membranes were measured by an Energy dispersive X-ray spectroscopy (7426 EDX OXFORD). The measurement voltage was 20 kV.

A commercially available atomic force microscopy (AFM) instrument (Nanoscope 8, Bruker) equipped with a J scanner was used to observe the morphology of the membrane surface. AFM imaging of the membranes was conducted in tapping mode with silicon nitride cantilevers (NP-S, Bruker).

The contact angles of the sludge foulant and membrane samples were measured by a contact angle meter (Kino industry Co., Ltb, USA). Ultrapure water, glycerol and diiodomethane were chosen as the probe liquids in the experiments. A drop ($5 \mu\text{L}$) was lowered onto the membrane's surface from a needle tip. A magnified image of the droplet was recorded by a digital camera. Static contact angles were determined from these images with a calculation software. The contact-angle measurement was taken as the mean value of 5 different points on each membrane. For measurement of the contact angles on the sludge foulant surface, the sludge liquor obtained from a membrane bioreactor (MBR) was firstly filtered through the membrane to form a cake layer. The formed cake layer was fixed within two glass slides to form a relatively flat surface, and then kept in an oven for 24 h at $40 \text{ }^\circ\text{C}$ to get rid of water. The foulant samples were used for contact angle measurement.

The zeta potential of the membrane was analyzed by a zeta potential analyzer (Zeta 90 Plus, Brookhaven, UK). The zeta potential of the sludge foulants was determined by a microelectrophoresis (Zetasizer Nano ZS, Malvern, UK). The measurement method

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