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Highly efficient anti-fouling electro-conductive membranes fabricated by reduced Graphene Oxide-Polyaniline (rGO-PANI) laminates

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Abstract

As an obstacle against the widespread application of membranes in larger scales, fouling via different materials could cause a severe decline in water permeation. Herein, we fabricated a novel mechanically-stable electro-conductive membrane by simple pressure-assisted laminating of reduced Graphene Oxide-Polyaniline (rGO-PANI) suspension on the commercial polyethersulfone (PES) support layers. Field emission scanning electron microscopy (FESEM) and water contact angle measurements were used to characterize rGO-PANI membranes. Using PANI improved the mechanical stability of laminated film and reduced the water contact angle, making a more hydrophilic surface. Organic fouling behaviors of newly fabricated membranes were also investigated by applying a DC voltage to membranes. The experimental results showed a significant improvement in water flux recovery when 2 V DC potential is applied on the surface of the rGO-PANI membrane.

Keywords: Electro-conductive membrane, Reduced graphene oxide-polyaniline nanocomposite, Anti-fouling, Electro-oxidation, Microfiltration

1. Introduction

In recent years, graphene-based membranes have demonstrated very promising performance in water desalination (Karkooti et al., 2018). Two neighboring graphene sheets can create special interconnected channels, which can selectively allow water to permeate, while rejecting all other solutes. Among different methodologies to fabricate graphene-based membranes, laminating graphene layers over porous supports such as polyethersulfone (PES) is a versatile technique and can be easily scaled up. Using strong oxidants, graphite is exfoliated into graphene oxide (GO) sheets. To prepare reduced graphene oxide (rGO), the oxygen containing functional groups on the surface of GO should be partially removed. Earlier studies showed that GO laminated membranes lose their selectivity over time due to the swelling effect in in aqueous environment (Zheng et al., 2017). On the other hand, rGO possesses a very hydrophobic surface which is potentially prone to fouling by various kinds of contaminants in water.

2. Materials and Methods

Reduced graphene oxide (rGO) was synthesized by modified Hummer's method followed by a thermal reduction in 1000 °C under pure N_2 atmosphere (Marcano et al., 2010). Stock solutions of PANI (Sigma Aldrich, Emeraldine Base, MW~65,000 Da) in N-methyl-2-pyrrolidone (NMP, Fisher Scientific Co)/DI water ($W_{NMP}/W_{DI\ water}$ = ¼) and rGO in DI water were prepared after 1 h sonication. Using a dead-end filtration cell, a certain amount of each solution was forced to pass through a microporous PES (0.1 µm pore size, Sterlitech) substrate. The recipe that was used to prepare three membranes (M1-M3) is shown in Table 1.

Table 1. Compositions of different membranes

Membrane	Substrate	Top layer	
		PANI Sol. (50 ppm)	rGO Sol. (20 ppm)
M1	PES	10	-
M2	PES	10	100 ml
M3	PES	5	100 ml

To verify antifouling performances of the fabricated membranes, 0.1 g of sodium alginate (ACROS organics) and 0.58 g of NaCl (as an electrolyte) were added to 2 L DI water. This solution was then circulated in a custom-made cross-flow filtration setup equipped with a plastic membrane cell. A lab-scale diaphragm pump was used to apply 40 psi transmembrane pressure. In some experiments, membrane's top layer was connected to the cathode output of a DC power supply. Using a piece of stainless-steel anode (radius = 2 cm), 2 V electric potential was employed to study the electroactivity of the fabricated membranes. The distance between anode and cathode was adjusted to be 0.5 cm. The mass of permeate water was measured using a weighing balance (ME4002, Mettler Toledo, USA) and was automatically recorded on a computer via a data acquisition system. Water flux (J) was finally obtained by measuring the volume change of water

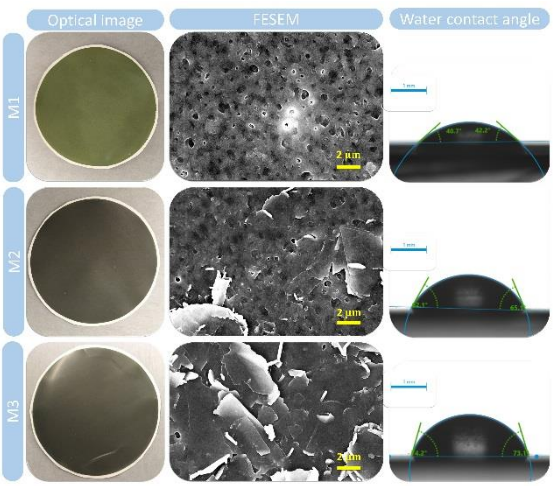
(ΔV) being passed through the effective membrane's surface area (A) during time of sample collection (Δt):

$$\boldsymbol{J} = \frac{\Delta \boldsymbol{V}}{\boldsymbol{A}\Delta \boldsymbol{t}} \qquad (1)$$

3. Results and Discussion

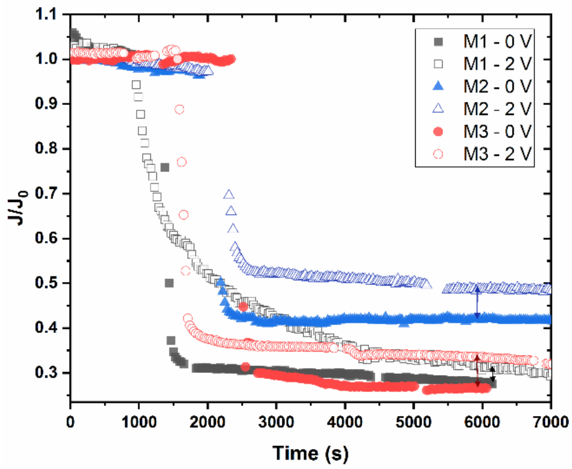
Optical images, FESEM and water contact angles of the top surfaces of the synthesized membranes are shown in Fig. 1. As can be seen, the PANI and rGO were uniformly coated on the surface of all three membranes. They were mechanically stable and after cross-flow filtration tests no detachment from the surface of membranes was observed. The top FESEM image of M1 shows large pores which are mainly related to open spaces between linear PANI chains. By increasing the rGO concentration, more planer structure could be seen in FESEM image of M3 membrane. Furthermore, the top surface of M1 membrane had the highest hydrophilicity with average water contact angle of 41.5°. As mentioned before, to synthesize rGO, GO was initially synthesized through Hummer's method. Due to the presence of abundant oxygen-containing functional groups, GO is very hydrophilic and can be easily dispersed in water. However, after thermal reduction, most of these functional groups are removed that renders rGO hydrophobic property (Rastgar et al., 2019). M2 and M3 membranes contained hydrophobic rGO nanosheets which enhanced their hydrophobicity or water contact angles.

Fig. 1. Optical images, FESEM and water contact angles from top surfaces of the fabricated membranes.si hydraulic pressure.



Antifouling performance of membranes was assessed by applying either 0 or 2 V DC electric potential between membrane's surface (as a cathode) and a stainless steel plate (as an anode). As can be seen in Fig. 2, by applying 2 V, all three membranes demonstrated better antifouling performance as compared to the tests with no electric potential. This improvement for M1, M2, and M3 was 2.1%, 6.8%, and 7.3%, respectively. Since the membranes were connected to cathode output of a DC power supply, their surface possessed more negative charges. Therefore, electrostatic repulsion between membrane surface and negatively charged alginate foulants could be presumed as a main reason for such observation. The existing rGO on the surfaces of M2 and M3 is more electroactive than the PANI and can improve kinetics of electron transfer. This effect could be manifested in more improvement of relative water flux which was achieved by M2 and M3 membranes.

Fig. 2. Fouling behaviors of different fabricated membranes.



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