



Novel conductive membranes breaking through the selectivity-permeability trade-off for Congo red removal

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ABSTRACT

The permeability-selectivity trade-off remains the major drawback limiting application of membrane technology. This study provided a new strategy to break through this trade-off. A novel conductive membrane prepared by electroless Ni plating of the polyvinylidene fluoride (PVDF) membrane was used for dyeing wastewater treatment by applying an electric field. Series analyses demonstrated success of immobilizing a Ni/NiO layer on the pristine PVDF membrane. The conductive membrane possessed improved hydrophilicity indicated by the water contact angle decrease from 75.5° to 45.0°. Moreover, the conductive PVDF membrane integrating electric field voltage of 20 V simultaneously possessed high selectivity (97.93% rejection) and extremely high permeability ($202.51 \pm 9.63 \text{ L m}^{-2} \text{ h}^{-1}$ at 0.1 MPa) for filtration of Congo red (CR) solution, significantly breaking through the permeability-selectivity trade-off. The filtration performance was quite stable even after 6 filtration cycles, indicating the feasibility of the conductive membrane for long-term treatment of CR wastewater. In addition, the conductive membrane showed high antifouling ability to methylene blue (MB), rhodamine B (RB), CR and microorganisms. The conductive membranes, together with the new strategy, offered in-depth insights into membrane fabrication.

1. Introduction

The whole world is suffering increasing water stresses due to the continuous discharge of dyeing wastewater. According to the literature, the textile industry annually produces more than 700,000 tons of dyes and 100,000 commercial synthetic dyes [1–4]. Presence of dyes even at a rather low concentration is significantly visible and toxic to aquatic life by damaging the esthetic nature of water and reducing the photosynthetic activity of aquatic organisms [5–7]. Among dyes, Congo red (CR) has drawn special attention due to its extensive usage in textile industries and high resistance to fading and biological degradation [8]. Therefore, removal dyes, especially CR, from dyeing wastewater before discharging into the water systems is of great importance.

Pressure-driven membrane technology, especially microfiltration (MF) and ultrafiltration (UF) process, becomes more and more attractive for wastewater treatment due to its advantages such as low investment, low energy consumption, efficient rejection and environment friendly property [9–12]. However, in practice, it has to generally reduce membrane flux so as to guarantee high rejection, and vice versa

[13,14]. The inherent trade-off between membrane permeability and selectivity remains the major drawback limiting its application [13,15,16]. Extensive efforts have been devoted into overcome the permeability-selectivity trade-off limitation [13,16–18]. Whereas, almost all these efforts focused on adjusting pore structure including skin layer thickness, porosity and pore size. As adjusting pore structure is quite complex and involves varied disciplines, there is lack of a universally effective adjusting strategy. Novel strategies based on other principles have yet to be developed to break through this trade-off limitation.

Permeability decrease during filtration process is mainly caused by the foulant adhesion and/or pore blocking [19–24]. It is, therefore, envisaged that, the permeability decrease can be avoided to some extent if there is an extra force provided to repel the foulant adhesion and/or blocking, and meanwhile, the repulsive force can help to retain foulants and improve the selectivity, facilitating to break through the trade-off limitation. Since most of the dyes in solution are positively or negatively charged [25–27], exerting an electric field on membrane surface would provide an electrostatic repel force against the dyes

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[28,29]. Moreover, the relatively large pore size of MF/UF membrane is beneficial to obtain relatively high flux. This hypothesis essentially requires a conductive membrane involved in the filtration. Development of electrically conductive MF/UF membranes have recently drawn significant attention [30,31]. However, conductive membranes are generally used in electro dialysis and/or capacitive deionization process to separate ions from water [32], or used as a critical unit in catalysts, fuel cells and sensors [33]. To our knowledge, no one has considered them as a potential strategy to break through the permeability-selectivity trade-off limitation for filtration of dyes.

Electroless plating has been reported to be effective to endow a polymeric materials with conductivity [34–36]. Although no attempt has been made to fabricate conductive membranes by the electroless plating technique, it should be feasible to make up a conductive separation membrane because of the same electroless plating mechanism between them. However, while this hypothesis is obviously reasonable, to our knowledge, there is no study conducted to test this hypothesis. Poly(vinylidene fluoride) (PVDF) represents one of the most popular materials used for fabricating pressure-driven membranes [37–39]. Meanwhile, electroless nickel plating is one of the most promising technique to create a conductive layer on an objective [40,41]. Therefore, fabricating PVDF-Ni conductive membranes should be very helpful for examining this hypothesis.

In this study, in order to test the above-mentioned hypothesis, PVDF-Ni conductive membranes were prepared by the combined process of polydopamine (PDA) coating, Ag catalytic activation and electroless Ni plating. The prepared membrane together with the pristine membrane were characterized by series techniques including scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDX), X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and thermogravimetric analysis (TGA). Thereafter, the prepared PVDF-Ni conductive membranes were used to treat dyeing wastewater by applying an electric field. Performance of the prepared membranes was assessed. This study would provide a novel strategy to break through the permeability-selectivity trade-off limitation of membranes.

2. Materials and methods

2.1. Materials

The PVDF membrane supported by nonwoven was purchased from Jiangsu Qingquan Co. Ltd. Dopamine (DA) was provided by Aladdin Holdings Group. Silver nitrate (AgNO_3), nickel sulfate ($\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$), dimethylaminoborane ($\text{C}_2\text{H}_{10}\text{BN}$), sodium pyrophosphate ($\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$), ammonia ($\text{NH}_3 \cdot \text{H}_2\text{O}$, 28%), methylene blue (MB), rhodamine B (RB) and CR were provided by Sinopharm Chemical Regagent Co., Ltd. All the chemicals were used without pretreatment.

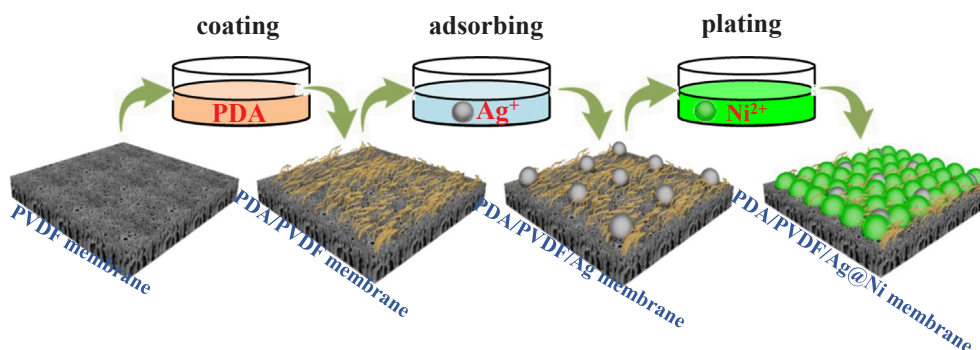


Fig. 1. Diagram of the membrane modification process adopted in this study.

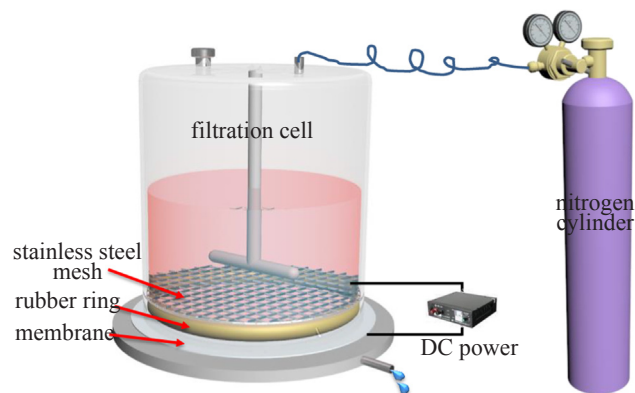


Fig. 2. The schematic of the filtration cell integrated with electric field.

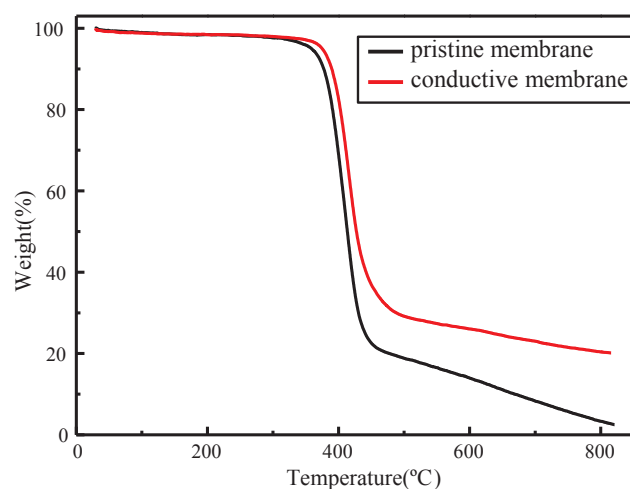


Fig. 3. TGA curves of the pristine and conductive membranes.

2.2. Preparation of the conductive membranes

Fig. 1 shows the diagram of the membrane preparation process adopted in this study. The preparation process can be described as follows: The PVDF membrane was rinsed with pure water for cleaning. The clean membrane was immersed into a dopamine hydrochloride solution (2 mg/L) and shaken for 24 h at $30 \pm 1^\circ\text{C}$. In this step, the PDA was coated onto the PVDF membrane. The PDA coated membrane was immersed into AgNO_3 solution (1.597 g/L) for 10 min to absorb silver ions (Ag^+) at room temperature ($25 \pm 1^\circ\text{C}$). The membrane was rinsed again to remove the extra Ag^+ . The adsorbed Ag^+ will be reduced to micro Ag cores which facilitate reduction of Ni^{2+} . After that, the membrane was immersed into plating solution (25 g $\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$, 25 g $\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$, 50 g $\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$, 45 ml $\text{NH}_3 \cdot \text{H}_2\text{O}$ and 1.5 g

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