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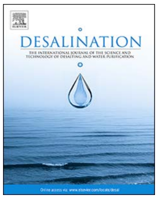

Fabrication and properties of graphene oxide-embedded cellulose triacetate RO composite membrane via melting method

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ABSTRACT

Graphene oxide (GO) has attracted significant attention on account of its unique structure and superior performance, deriving a new research field for materials science. Herein, cellulose triacetate (CTA) flat-sheet RO composite membrane was fabricated with GO embedding in its layer via melting method, which exhibited higher permeate flux as compared to pristine CTA RO membrane. The morphologies and structural arrangements of the obtained composite membranes were analyzed by X-ray diffraction (XRD), Raman spectroscopy, Confocal scanning microscopy (CSM), and Scanning electron microscopy (SEM), respectively. It was found that thermogravimetric (TG) spectrograms revealed GO generating the degradation centres and changing degradation process with the increase addition of GO. Moreover, the mechanical property indicated that the incorporation of GO could contribute to the improvement of tensile strength from 10.2 MPa to 23.1 MPa because of the dominant interactions between C═O groups from CTA and −OH and −COOH groups from GO. Finally, the RO membranes' performance showed that the permeate flux of membrane increased from 1.67 $L \cdot m^{-2} \cdot h^{-1}$ to 4.74 $L \cdot m^{-2} \cdot h^{-1}$ with the increasing GO content. However, the salt rejection compromised at the same time, perhaps due to the formation of channels which might offer the passageway of water accompanying by salt with the increasing GO content.

1. Introduction

Due to the effects of climate change and fresh water resource pollution, water shortage has further accelerated since 1995, is becoming a very urgent problem [1]. Providing hygienic and affordable portable water to meet the ever-rising demand is an on-going global requirement [2,3]. One of the most pressing challenges is saline water desalination, which can achieve clean portable water from the most abundant global water resource. Especially, the technology of desalination can transform sea water into fresh water, and act as a kind of important method to solve clean water shortage problems [4]. In recent years, RO is generally accepted as the most important desalination technology. Also, it has slowly replaced traditional evaporation technology such as multi-stage flash [5], and can combine with some new technologies such as capacitive deionization [6], electrodialysis [7], membrane distillation [8] and forward osmosis [9], is expected to lead the membrane industry in the near future. Over the past few decades, the developments of RO membrane materials, membrane process, feed solution pre-treatment, and reduction in energy consumption were studied as remarkable advances.

There is no denying that one of the key issues is to improve the permeate flux of membranes with higher operation efficiency in RO filtration process. The permeate flux closely associates with the membrane thickness according to molecular-transport models [10,11]. Up to now, most of water treatment plants employ polyamide (PA) or its derivative RO membranes can provide satisfied permeability and salt rejection [12]. However, PA RO membranes deserve poor chemical stability to oxidation system, for example, free chlorine which has a good performance to resist biofouling, can weaken the PA RO membrane performance [13,14]. Cellulose triacetate (CTA) has emerged to prepare RO membranes, particularly for wastewater with severe biofouling, because of its good chlorine resistance. Generally speaking, CTA RO membrane exhibits temperature stabilization in a wider temperature range, while it deserves poor permeability compared to PA membranes [15,16]. One of the best methods is adding polymer or nano particle in CTA membrane, which can offer better permeability and selectivity [17,18].

It is generally accepted that various nanostructured carbons have been used for enhancing the properties of materials [19–21]. For example, carbon nanotubes (CNTs) have been widely studied because of their unique atomic structure, including excellent mechanical, thermal, and chemical properties as well as high surface area-to-volume ratio

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[22,23]. Especially, nanoparticle-coated and polymer- multi-walled carbon nanotubes (MWCNTs) have attracted some attention in order to overcome low surface energy of the outer walls of MWCNTs [24–26]. Moreover, graphene, a monatomic two-dimensional carbon nanomaterial, have been intensively investigated in terms of its high specific surface area, excellent electrical, thermal, optical and mechanical properties [27]. Similarly, graphene can effectively enhance the performance of the matrix polymer even at a very low loading [28,29]. Specially, graphene can be regarded as a king of novel functional separation materials, which can generate immense interest for water purification among the membrane field due to its honeycomb lattice structure [30,31]. It is commonly known that GO is an important graphene derivative which exhibits excellent hydrophilicity because of its abundant oxygen functional groups on the surface of atomic-layer thick nano sheet [32–34]. On the basis of these advantages, several excellent filtration membranes constructed from GO, including chemically converted graphene nanosheets [35–36], have been reported very recently and opened up an exciting direction in separation membranes because of their facile and large-scale production in solution [37–38]. Moreover, the performance of anti-fouling properties [39–40] and chlorine resistance [41] also can be improved when GO is applied in separation membranes. Recently, Chae et al. [42]. have studied that GO-embedded PA RO membrane by interfacial polymerization, which exhibited higher permeate flux, chlorine resistance and anti-fouling property accompanying by little loss of salt rejection. Perreault et al. [43]. reported that antimicrobial properties could be enhanced by enriching GO on the surface of membrane, because of an immediate contact between GO and bacterial cells. In addition, Nair et al. [30]. has verified that layered arrangement GO sheets could offer abnormal rapid permeation of water vapor on account of its unique water transport nature. To sum up, GO can be regarded as modified additive to improve the permeate performance of RO membrane because of its unique mass transfer performance.

As we all known, CTA exhibits higher crystallinity and poor solubility in solvents miscible with water, which have become the main obstacles to employ it for the membrane preparation by the phase inversion method. The melting method (M-M) process is another way of preparing membranes. In the M-M process, a polymer mixed with plasticiser is melted at high temperature, and then moulding in the moderate coagulating bath. Fabrication of polypropylene and poly(vinylidene fluoride) microporous membranes are the most common materials by the M-M process. As far as we know, about the attempts of fabricating CTA RO membranes by M-M process has rarely been reported in literature except for few patent documentations [44–45].

This work utilized the unique properties of GO, and the common CTA RO membrane material to fabricate GO-embedded CTA RO composite membrane with higher permeate flux via M-M process. The objective of this study was to research the generating channel via the interaction between CTA and GO so as to achieve higher permeate flux of GO-embedded CTA RO composite membrane as compared to pristine CTA RO membrane. However, there were some limitations that the salt rejection compromised, due to the difficulties of precise controlling the size of the channel, which might offer the passageway of water accompanying by salt with the increasing GO content.

2. Experiments

2.1. Materials and preparation process

Before membrane fabrication, CTA resins (LT35, average $M_n \approx 50{,}000$ g/mol, kindly provided by Daicel.), and benzoic acid (BA) (Tianjin Fengchuan Chemical Reagent Science and Technology Co., Ltd.) were desiccated to remove moisture in a vacuum oven (24 h, 80 ± 2 °C, 2 mbar), GO (Nanjing Yoshikura Nano Technology Co., Ltd.) was dispersed in Tetramethylenesulfone (TMS) (Tianjin Kermel Chemical Reagent Co., Ltd.) by sonication for 90 min at three different mass ratio related to polymer, 0.02, 0.05, and 0.1 wt%, respectively. Firstly, CTA, BA, GO, TMS and ethylene glycol(EG) (Tianjin Guangfu Chemical Reagent Co., Ltd.) with different mass ratio (Table 1) were homogeneously mixed under vigorous mechanical stirring. Secondly, the mixture (10 g) was placed in plate vulcanization machine with a 300 μm thickness stainless steel mold. Then, the pressure between the mold was pre-compressed to 2 MPa with the 160 °C heating temperature, and when the temperature was up to 180 °C, the pressure was compressed to 15 MPa for 10 min. Thirdly, the nascent CTA flat sheet RO membranes were prepared after the mold was cooled to room temperature by circulating water system (25 °C). Finally, GO-embedded CTA (CG) flat-sheet RO composite membranes with different mass ratio of GO were fabricated after TMS and additives was extracted through dipping in coagulating bath of 10 °C for 24 h, which was comprised TMS/EG/Water in 24/1/75. All the preparation parameters were tabulated in Table 2. The obtained CG RO composite membranes were dipped in water of 60 °C, and subjected to an annealing treatment for 30 min. Four different RO membrane with GO (0, 0.02, 0.05, and 0.1 wt%) were named CG-0, CG-1, CG-2, and CG-3, respectively. Fig. 1 showed the plate vulcanization machine and fabrication process of CTA RO composite membranes, while the parameters and the compositions were tabulated in Table 1.

Table 1
The composition of the prepared CG RO membranes.

Sample	CG-0	CG-1	CG-2	CG-3
CTA	42 wt%	42 wt%	42 wt%	42 wt%
TMS	50 wt%	50 wt%	50 wt%	50 wt%
BA	1 wt%	1 wt%	1 wt%	1 wt%
EG	7 wt%	7 wt%	7 wt%	7 wt%
GO	–	0.02 wt%	0.05 wt%	0.1 wt%

2.2. Characterization of CG RO membrane

2.2.1. Functional group analysis

Thermo Fisher Scientific Nicolet iS50 instrument was utilized to acquire Fourier transform infrared spectroscopy (FTIR) spectrum with the attenuated total reflectance (ATR) technique. Testing the infrared range was logged in 550–4000 cm^{-1} interval with 1 cm^{-1} resolution. Moreover, Horiba Xplora Plus Raman Microscope was utilized to measure the Raman spectrum. A 532 nm laser line was used accompanying by total number of 10 scans.

2.2.2. X-ray diffraction

Bruker D8 Discover instrument was utilized to perform the X-ray diffraction (XRD) investigation of GO and CG RO composite membranes at room temperature by employing Cu K_α radiation.

2.2.3. Thermal performance

Thermogravimetric (TG) equipment (STA449F3, Germany) was utilized to assess the thermal performance of the GO and CG RO composite membranes. The temperature of measurements was from 25 °C to 600 °C with a heating rate of 10 $°C\cdot min^{-1}$. Above all, all of the testing

Table 2
Fabrication parameters of CG RO membrane.

	Parameters
Compressed temperature (°C)	180
Compressed time (min)	20
Compressed pressure (MPa)	15
Coagulation bath (TMS/EG/water)	24/1/75
Annealing treatment temperature (°C)	60
Annealing treatment time (min)	30
Compressed membrane size (μm)	300

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