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FABRICATION OF NANOFILTRATION MEMBRANE UTILISING SURFACTANT VIA NON-SOLVENT INDUCED PHASE SEPARATION METHOD

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Abstract

In this work, graphene oxide (GO) and titanium dioxide (TiO_2) were used as an additive to fabricate the nanofiltration (NF) membrane. GO was synthesised via electrochemical exfoliation method utilising sodium bis(3,5,5-trimethyl-1-hexyl) sulphosuccinate (AOT4) surfactant. The synthesised GO was then used to fabricate PVDF-based NF membrane namely PVDF/GO_TiO_2 via non-solvent induced phase separation (NIPS) method. The effects of embedded GO and TiO_2 on the morphology and structural properties of PVDF/GO_TiO_2 were investigated by using field emission scanning electron microscopy (FESEM) and micro-Raman spectroscopy. The finding shows that PVDF/GO_TiO_2 present thin and dense top layer supported by macro-voids sub-layer with sponge-like layer at the bottom. Based on its morphology, the fabricated PVDF/GO_TiO_2 membrane is potential to be applied as membrane filtration for water treatment application.

Key Words: Graphene oxide; Electrochemical exfoliation; Nanofiltration; Titanium dioxide

INTRODUCTION

Nowadays, dye contamination becomes a crucial issue due to the lack of clean water to be consumed [1]. Since water is essential to human being, numerous studies have been conducted in order to overcome this problem and provide fresh water. Membrane separation as an effective technology for dye removal has become a good selection for water treatment and plays a significant rule in major industrial [2]. Membrane separation technology can be classified according to its separation principle and membrane properties such as microfiltration, ultrafiltration, nanofiltration (NF) and reverse osmosis. Among them, NF membrane has been applied to treat dye pollution due to its advantages; low operating pressure, can retain ions and small pore size [3].

Poly(vinylidene fluoride) (PVDF) is a synthetic polymer with outstanding physical and chemical properties. PVDF is extremely stable and good chemical resistance. Compatible with process-ability, PVDF membrane can be fabricated in form of flat sheet, rolls of hollow fiber and tubular membrane with different technique such as stretching, sintering and phase inversion [4, 5]. Therefore, due to its properties, PVDF have been extensively used as a membrane material for industrial application [6]. However, hydrophobic nature of PVDF has increase the tendency of membrane fouling during separation process thus affecting its performance. Recently, there are several approaches has been introduced to overcome this problem such as physical and chemical modification with inorganic material [7, 8]. Among them, physical modification attracted more attention due to it simple preparation. In particular, blending polymer with inorganic additives could enhance membrane performance [9] by increasing the water permeability, hydrophilicity and antifouling of the membrane.

Titanium oxide (TiO_2) is considered as the potential candidate to improve membrane performance as it is hydrophilic, has chemical stability, low toxicity and commercial availability [10]. On the other hand, graphene oxide (GO) also offers a great improvement on membrane performance as it contains abundance number of oxygen-functional group [11] which make GO strongly hydrophilic [12-13]. Some studies indicated that blending TiO_2 and GO directly into polymeric membrane resulted in increasing the hydrophilicity, higher dye rejection and excellent antifouling properties. Wu et al. (2018) reported water flux of the membrane was increases as higher porosity of the hybrid membrane was achieved when TiO_2 and GO were embedded in the polymeric membrane [10]. This result was in a good agreement with other reports [14] which shows the performance of the membrane were improved as incorporating TiO_2 and GO in polymeric membrane.

GO was usually synthesised via Hummer's method since it can produce GO with high quality. However, this method involves hazardous chemical consumption and complicated procedure [15]. Currently, chemical exfoliation method assisted by surfactants has been introduced as greener and low cost productions to synthesise GO [16-19].

In this work, the fabrication of NF membrane; PVDF/GO_TiO_2 was done by utilising sodium bis(3,5,5-trimethyl-1-hexyl) sulphosuccinate (AOT4) surfactant to assist exfoliation process and study its effect on the morphology of the fabricated NF membrane.

METHOD

The first step in fabrication of NF membrane was preparing the GO solution. GO is synthesised via electrochemical exfoliation method in electrolyte solution assisted by a surfactant based on previous study conducted by Md Disa et al. and Suriani et al. [17, 18, 20]. 0.1 M concentration of electrolyte is made from dimethylacetamide

(DMAc) with AOT4 surfactant. Two graphite rods were immersed in the prepared electrolyte solution and connected to 7 V power supply for 24 hours.

Next, the obtained GO solution was further used to fabricate the membrane solution. PVDF (20 wt%) and TiO_2 (1 wt%) were added to the GO solution. The membrane solution was stirred under 70°C of temperature for 48 hours. For a day, membrane solution was kept at room temperature to release the air bubbles. The NF membrane was prepared by non-solvent induced phase separation (NIPS) method. The membrane solution was casted on the glass plate using a casting knife with 200 μm thickness. After that, fabricated NF membrane (PVDF/GO_TiO_2) was immersed in deionised (DI) water for overnight. The fabricated membrane was immersed in DI water until further characterisation. The morphological and structural properties of the fabricated NF membrane were determined by field emission scanning electron (FESEM) and micro-Raman spectroscopy.

RESULTS AND DISCUSSIONS

FESEM images of the cross section of fabricated membrane, PVDF/GO_TiO_2 membrane are presented in Figure 1. Based on Figure 1 (a), PVDF/GO_TiO_2 membrane shows a smooth surface with the visible pore in the range of 23.8-55.6 nm. Moreover, the fabricated membrane also consists of three layers as depicted in Figure 1 (b). The upper layer presents a thin and dense skin layer with macro-voids sub-structure. The formed macro-voids structure is believed due to the rapid exchange rate between solvent and non-solvent into the casting solution during NIPS process [21, 22]. Large macro-voids structure presented by PVDF/GO_TiO_2 was believed to give an advantage if they were applied as a membrane filtration in water treatment application since it can increase the permeability of the membrane. The lowest layer was observed to be a sponge-like layer structure.

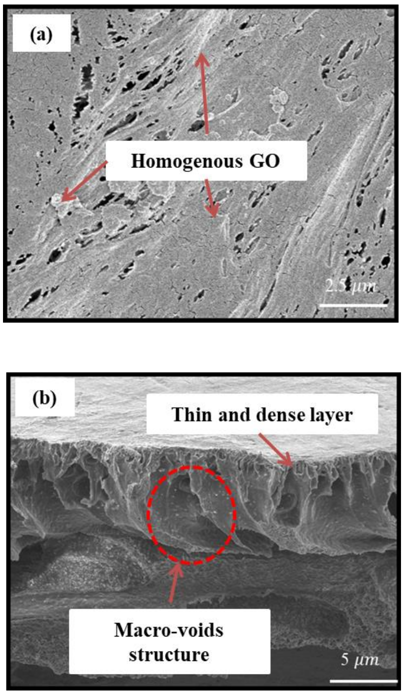

Figure 1. FESEM images of PVDF/AOT4_GO/TiO_2; (a) Surface morphology and (b) cross-section of membrane

The structural properties of the fabricated PVDF/GO_TiO_2 membrane are presented in Figure 2. The D- and G-peaks of PVDF/GO_TiO_2 were observed at 1331.49 and 1568.8 cm^{-1}, respectively. The shifted G-peak of PVDF/GO_TiO_2 and graphite (1581 cm^{-1}) confirmed the successful oxidation process during electrochemical exfoliation. The defect level of the fabricated NF membrane was then determined based on I_D/I_G ratio. The I_D/I_G ratio presented by PVDF/GO_TiO_2 is higher than graphite (0.84) which is 1.03. This result shows that PVDF/GO_TiO_2 membrane possesses higher defect level than graphite. In addition, there were several TiO_2 peaks spotted at 147, 392, 510 and

635 cm^{-1}. The spotted peaks were confirmed the modes of the anatase phase of TiO_2.

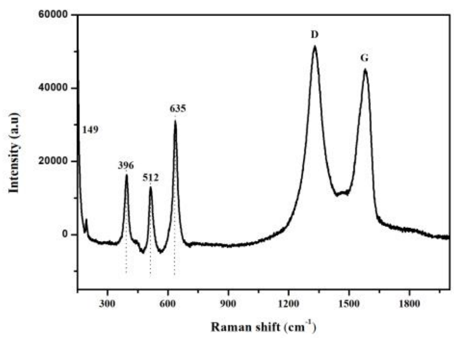

Figure 2. Micro-Raman spectra of the fabricated PVDF/GO_TiO_2 NF membrane

CONCLUSION

PVDF/GO_TiO_2 NF membrane has been successfully fabricated utilising the synthesised GO based on AOT4 surfactant. Based on FESEM images and micro-Raman spectroscopy, the fabricated PVDF/GO_TiO_2 NF membrane possessed large macro-voids structure and I_D/I_G ratio of 1.03. These properties were believed to enhance the membrane performance and can be applied for dye rejection.

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REFERENCES

[1] Wang, Jing, et al. 2015. Improved poly(vinyl butyral) hollow fiber membranes by embedding multi-walled carbon nanotube for the ultrafiltrations of bovine serum albumin and humic acid. *Chemical Engineering Journal*, *260*, 90–98. https://doi.org/10.1016/j.cej.2014.08.082

[2] Liu, Ting, et al. 2017. Trivalent metal cation cross-linked graphene oxide membranes for NOM removal in water treatment. *Journal of Membrane Science*, *542*(July), 31–40. https://doi.org/10.1016/j.memsci.2017.07.061

[3] Park, Hyung Jin, et al. 2018. Nafion/TiO2 nanoparticle decorated thin film composite hollow fiber membrane for efficient removal of SO2 gas. *Separation and Purification Technology*. https://doi.org/10.1016/j.seppur.2018.10.010

[4] Dhand, Vivek, et al. 2019. Fabrication of robust, ultrathin and light weight, hydrophilic, PVDF-CNT membrane composite for salt rejection. *Composites Part B: Engineering*, *160*(December 2018), 632–643.

[5] Liao, Yuan, et al. 2018. Progress in electrospun polymeric nanofibrous membranes for water treatment: Fabrication, modification and applications. *Progress in Polymer Science*, *77*, 69–94. https://doi.org/10.1016/j.progpolymsci.2017.10.003

[6] Li, Renjie, et al. 2018. A novel strategy to develop antifouling and antibacterial conductive Cu/polydopamine/polyvinylidene fluoride membranes for water treatment. *Journal of Colloid and Interface Science*, *531*, 493–501. https://doi.org/10.1016/j.jcis.2018.07.090

[7] Kang, Guo-dong, et al. 2014. Application and modification of poly(vinylidene fluoride) (PVDF) membranes - A review. *Journal of Membrane Science*, *463*, 145–165.

[8] Xia, Shengji, et al. 2014. Preparation of poly(vinylidene fluoride) membranes with graphene oxide addition for natural organic matter removal. *Journal of Membrane Science*, *473*, 54–62.

[9] Ai, Jing, et al. 2018. Applications of graphene oxide blended poly(vinylidene fluoride) membranes for the treatment of organic matters and its membrane fouling investigation. *Applied Surface Science*, 455, 502–512.

[10] Wu, Li-guang, et al. 2018. Enhanced performance of polyvinylidene fluoride ultrafiltration membranes by incorporating TiO_2/ graphene oxide. *Chemical Engineering Research and Design*. https://doi.org/10.1016/j.cherd.2018.11.025

[11] Abu bakar, Suriani, et al. 2018. Reduced graphene oxide/platinum hybrid counter electrode assisted by custom-made triple-tail surfactant and zinc oxide/titanium dioxide bilayer nanocomposite photoanode for enhancement of DSSCs photovoltaic performance. *Optik*, *161*, 70–83. https://doi.org/10.1016/j.ijleo.2018.02.013

[12] Miao, wei, et al. 2017. Improved ultrafiltration performance and chlorine resistance of PVDF hollow fiber membranes via doping with sulfonated graphene oxide. *Chemical Engineering Journal* (Vol. 317). https://doi.org/10.1016/j.cej.2017.02.121

[13] Zhao, Jingjing, et al. 2019. Fabrication of GO modified PVDF membrane for dissolved organic matter removal: Removal mechanism and antifouling property. *Separation and Purification Technology*, *209*, 482–490. https://doi.org/10.1016/j.seppur.2018.07.050

[14] Dadvar, Elahe, et al. 2017. Efficiency of polymeric membrane graphene oxide-TiO_2 for removal of azo dye. *Journal of Chemistry*, vol. 2017.

[15] Zaaba, N.I, et al. 2017. Synthesis of graphene oxide using modified hummers method: solvent influence. *Procedia Engineering*, *184*, 469–477. https://doi.org/10.1016/j.proeng.2017.04.118

[16] Abu bakar, Suriani, et al. 2015. A facile one-step method for graphene oxide/natural rubber latex nanocomposite production for supercapacitor applications. *Materials Letters*, *161*, 665–668. https://doi.org/10.1016/j.matlet.2015.09.050

[17] Md Disa, Nurhafizah, et al. 2015. The synthesis of graphene oxide via electrochemical exfoliation method. *Advanced Materials Research*, *1109*(December), 55–59. https://doi.org/10.4028/www.scientific.net/AMR.1109.55

[18] Abu bakar, Suriani, et al. 2019. Incorporation of electrochemically exfoliated graphene oxide and tio $_2$ into polyvinylidene fluoride-based nanofiltration membrane for dye rejection. *Water Air and Soil pollution* https:// doi.org/ 10.1007/s11270-019-4222-x

[19] Jumini, Sri, et al. 2019. The structural, optical and electrical properties of Graphene oxide-based counter electrode. *International journal of advace multidisciplinary scientific research*. 3, 1-8. https:// doi.org/ 10.31426/ijamsr.2019.2.3.1311

[20] Abu bakar, Suriani, et al. 2019. Improved DSSC photovoltaic performance using reduced graphene oxide–carbon nanotube/platinum assisted with customised triple-tail surfactant as counter electrode and zinc oxide nanowire/titanium dioxide nanoparticle bilayer nanocomposite as photoanode. *Graphene Technology*, 4, 17-31. https://doi.org/10.1007/s41127-019-00024-x

[21] Safarpour, Mahdie, et al. 2015. Development of a novel high flux and fouling-resistant thin film composite nanofiltration membrane by embedding reduced graphene oxide/TiO_2. *Separation and Purification Technology*, *154*, 96–107. https://doi.org/10.1016/j.seppur.2015.09.039

[22] Yang, Mei, et al. 2017. Preparation of graphene oxide modified poly(m-phenylene isophthalamide) nanofiltration membrane with improved water flux and antifouling property. *Applied Surface Science, 394*, 149–159. https://doi.org/10.1016/j.apsusc.2016.10.069