Graphene Oxide Nanocomposite for Sustainable Pure Water by PES Membrane

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Abstract: The urgent concern of water shortage have promoted to the development of different sustainable technologies with the rapid evolution of nanotechnologies. Graphene oxide (GO) is a water-soluble material that can be constructed into a number of structures such as membranes, and has many applications in environmental sectors. Furthermore enhancing membrane characteristics to improve water flux performance and mitigate fouling is crucial issue for membrane separation technology. GO serves as nanofiller to overcome fouling during filtration as well as water flux improvement. This work aimed to develop PES membranes by phase inversion method and the fabricated membranes subjected to series analysis of FESEM, hydrophilicity and water flux. The results showed that the GO improved the contact angle of the pure PES membrane and the water flux increased from 13 to 16 L/m^2.h. Therefore, the PES-GO membrane proved its capability to be used for various applications to reduce membrane fouling.

Keywords: Fouling, Graphene oxide, Membrane, Nanocomposite, PES, Water

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1. Introduction

Water resource sustainability is essential for the long-term evolution of modern society and economy. More strain on water resources deficit has been developed as a result of industrialization and human activities, which has accompanied the rapid expansion of the economy and society (Al Aani, Mustafa, & Hilal, 2020). Due to high growing in industry, the oily wastewater has increased dramatically, thus the surrounding environment such as drinking water as well as ground water and sea water being impacted significantly. As a result, the harm to the ecosystem as a whole may increase. Membranes, electrochemical, biological, UV irradiation, hybrid technologies, and destabilization of emulsions by adding minerals are only a few of the approaches that have been tested so far to remove oil from water. A variety of membranes, including those generated through interfacial polymerization, nanoparticle incorporation, and surface grafting, have been emphasized. (Bolto, Zhang, Wu, & Xie, 2020). Membrane technology, particularly Ultrafiltration (UF) membranes, has advanced significantly since the 1970s as a safe, clean, cost-effective, and effective separation method for a variety of components and contaminants in water and wastewater (Ismail, Khulbe, & Matsuura, 2015). Due to uniformity in the separation process, no chemicals added in or during the filtration process, high separation performance efficiency, ease of operation, and small footprint, polymeric membranes play a large role in separating oil from oil-water emulsions. The membrane acts as a barrier, preventing oil droplets from flowing through but allowing water molecules with smaller hydrodynamic particle sizes to pass through. In general, the membrane's surface parameters (pore size, porosity, surface wettability, and surface roughness) have a big impact on both permeate flux and rejection (Teow & Solihuddin, 2020).

Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), forward osmosis (FO), and membrane distillation (MD) are examples of novel water purification and reclamation techniques (Zhang et al., 2016). These membranes are classified based on pore size measurements (Figure 1), furthermore, each filtration process works in the specific range of operating pressure (Kumar et al., 2013). Among these membranes, MF and UF are the major filtration processes used for oil removal. Furthermore, hydrophilic surfaces will repulse oil droplets from the membrane and facilitate the passage of water (Zhu, Loo, & Bai, 2013). Membrane fouling has been a barrier in membrane water treatment technology since its inception, reducing water permeation flux, degrading product water quality, and increasing energy usage (Shannon et al., 2010). Membrane fouling can result in a temporary or permanent decrease in flux as a result of pore clogging, cake formation, organic and inorganic precipitation, and biological fouling. Because of direct interaction with various types of foulants in the raw water, practically all membrane processes suffer from fouling difficulties when used for long-term water purification (Zhang et al., 2016).

In order to deal with membrane fouling, various routs have been attempted, such as: surface modification by incorporating antifouling materials. In recent years, the Graphene (G) and Graphene Oxide (GO) have become one of the most important commercial nano-sheets in the world. GO is the derivative of the graphene and it has been extensively explored for the adsorption of various pollutants (Baig, Sajid, & Saleh, 2019). Researchers concern attracted to graphene (G) and graphene oxide (GO) due to many reports on their mechanical and chemical properties when incorporated with the membranes (Yeh, Cihlář, Chang, Cheng, & Teng, 2013; Mahmoudi, Ng, Ba-Abbad, & Mohammad, 2015). The GO materials have a great potential for absorbing organic molecules in aqueous solutions. (G. P. Rao, Lu, & Su, 2007), the GO membrane is extremely effective in removing organic pollutants and dyes from solution. Adsorption, flotation, and biological treatment are some of the technologies used in industry to treat produced water for disposal or reuse in other processes. These technologies, on the other hand, are ineffective for de-oiling water with droplet sizes smaller than 10 µm. Hybrid techniques have also been tried to break down the oil droplets, but these are energy-intensive and environmentally damaging. Membrane technology, on the other hand, has been recognized as a viable approach for oil emulsion separation that uses less energy and has a reduced environmental impact. (Alammar, Park, Williams, Derby, & Szekely, 2020). Rao et al. (2014) reported efficient removal of 4-chlorophene, 2,4-dichlorophene, and 2,4,6-trichlorophene by ZrO2 graphene composite compared pure membranes. Chung, Y.T., et al. stated that the PSf composite membrane with Zn-GO exhibits excellent antifouling and antibacterial properties (Chung et al., 2017). Furthermore, the advantages of using GO as a nanoplates with nanoparticles is to offer a good and homogenous distribution of the nanoparticles across the membrane matrix (Mahmoudi et al., 2015). Various kinds

of polymer including Polyethersulfone (PES), Polysulfone (PSf) and Polyamide (PA) are frequently utilized in membrane fabrication as a base polymer (Hamid et al., 2011; Nghiem, 2013; Faneer, Rohani, & Mohammad, 2016; Fujioka). Because of its great thermal stability and mechanical strength, PES is a promising polymer for membrane modification. However, lack of hydrophilicity causes fouling and low permeability considered a main failure of the PES membrane. Thus, recently some researches on incorporation of GO with PES membranes were applied to overcome these obstacles of using a PES as a base polymer (Wu, Tang, & Wu, 2014b; Mahmoudi et al., 2015). Therefore, this work aimed to fabricate PES and PES/GO 1 % membranes to improve membrane performance with enhanced water flux rate and better hydrophilicity which minimize the operation cost.
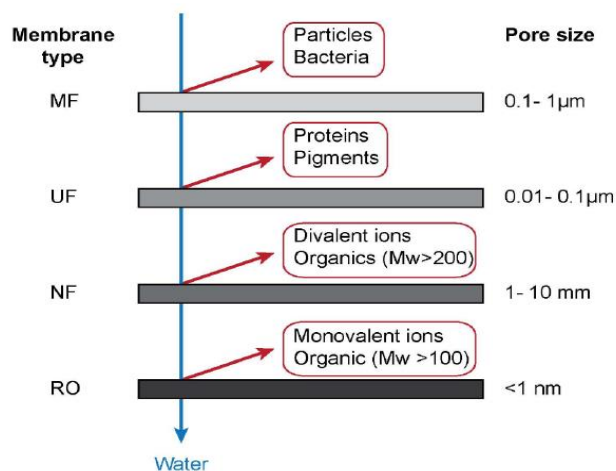

Figure 1: Membrane processes classification according to the pore size

2. Materials and methods

2.1. Materials

Polyethersulfone (PES) granules were used from Goodfellow Cambridge Ltd., England. The solvent 1-methyl-2-pyrrolidinone (NMP, 99.5% purity) was of analytical grade and purchased from Merck Co., Germany. Ultrapure water was used as non-solvent. GO nano-sheets was synthesized in the lab (refer to (Mahmoudi et al., 2015; Ba-Abbad, Mohammad, Mahmoudi, Faneer, & Benamor, 2016). The chemical structure of graphene oxide is illustrated in Figure 2.
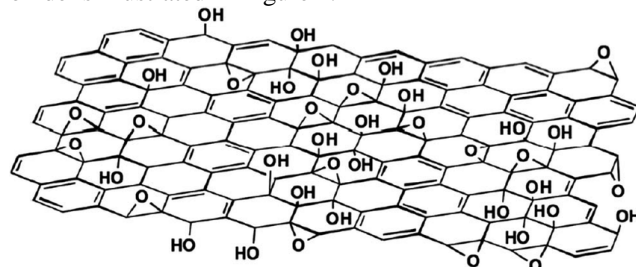

Figure 2: Schematic of graphene oxide structure (Wei, Vo, & Inam, 2015)

2.2. Synthesis of the Membranes

Nanocomposite membranes were created utilizing phase inversion induced by immersion precipitation. For membrane fabrication, a suitable amounts of PES (18 wt. %) were dissolved in solvent NMP (82 wt. %) with mechanical stirring speed at 250 rpm for 6 h. Afterward, the prepared GO nanoparticles with 1 % concentration were added into the mentioned polymeric solutions. Mixing was followed by sonication for 30 min for breaking up aggregates between nanoparticles. To thoroughly remove the air bubbles, the produced solutions were left at room temperature for around 24 hours without stirring. Then, using a film applicator, they were cast onto clean glass plates with a consistent thickness of 200 m. Next, immerse the glass plate horizontally in distilled water having an ambient temperature. The membranes were maintained in fresh distilled water for 24 hours after primary phase separation and solidification to guarantee complete solvent exchange. The membranes could then be used. The polymeric solution compositions are presented in Table 1.

Table 1: Casting solution compositions

Main Polymer	Solvent	GO amount	Remarks
PES 18 %	NMP 82 %	None	
PES 18 %	NMP 82 %	1 %	Sonication for 30 min

2.3. Characteristic techniques

Membrane performance

The fabricated membranes performance was evaluated through a dead-end stirred cell setup. The experiment was performed at fixed pressure (4 bar). The pure water flux was calculated according to the following equation

$$Jw = \frac{V}{A.t}$$

where Jw is the water flux (L/m^2.h), V is the permeate volume (L), A is the membrane effective area (m^2), t is the permeation time (h).

Contact angle measurement

Using a Rame-Hart model 200 standard contact angle goniometer with DROPimage Standard Software, the hydrophilicity of the manufactured membranes was determined with an accuracy of 60.10^0. The membranes were dried for 48 h and the medium used to measure the contact angle was deionized water and air at ambient temperature (25 – 28 ^{0}C).

Field emission scanning electron microscopy (FESEM)

FESEM Merlin Compact (Zeiss, Germany) was used to examine the cross sectional structures of the fabricated membranes. Prior the FESEM analysis, membrane samples were fractured into an appropriate size by using Nitrogen liquid and mounted on the sample holder.

3. Results and discussion

3.1. Membrane performance of water flux and hydrophilicity

Changes in the water flux of the membranes as a result of blending of hydrophilic materials (GO) were investigated via the pure water flux measurements. The GO-PES membrane shows higher water flux at 16 L/m^2.h comparing to PES membrane at 13 L/m2.h as presented in Figure 3.

To evaluate the hydrophilicity of blank and blended PES membranes, the contact angle analysis was used. Figure 3 exhibits the results of contact angle measurements for 1 wt.% of GO. The contact angle of blank PES membrane decreases from 80.0º to 64.7º by blending with 1 wt.% GO. The significant number of oxygenated groups of the GO nanosheets scattered in the polymer matrix can be attributed to the improved hydrophilicity of GO/PES membranes compared to blank PES. (Safarpour, Khataee, & Vatanpour, 2015), this may have a positive impact on the blended membranes' pure water flux.

It clearly observed that the improvement in membrane hydrophilicity has the same trend of water flux increase. Water permeability is increased by enhancing the membrane's hydrophilicity, which attracts water molecules inside the membrane matrix and facilitates their penetration across the membrane. As a result of the high water flux, low contact angle, and good hydrophilic character of the produced GO-PES membrane, the results demonstrated that it performed well. Therefore, adding GO to PES membrane enhancing the membrane antifouling properties.
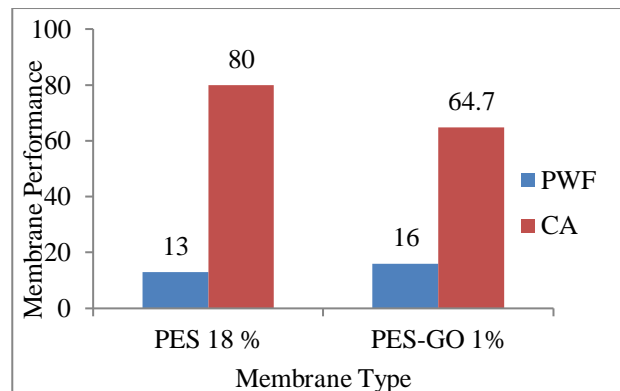

Figure 3: Water flux and contact angle measurements of PES and GO-PES membranes

3.2. FESEM

The cross-sectional FESEM images of the prepared mixed matrix membranes are presented in Figure 4.
The PES and GO-PES membranes have an asymmetric structure with a finger-like porous sub-layer and a skin top-layer. The addition of GO to the polymer matrix modified the membranes' finger-like shape slightly. Also, with the addition of GO to the membrane structure, the pore size of the membrane structure grows greater. The same finding was established by (Wu, Tang, & Wu, 2014a; Shanmugam, 2022) where the membrane pores became bigger as SiO_2/GO added to membrane matrix.
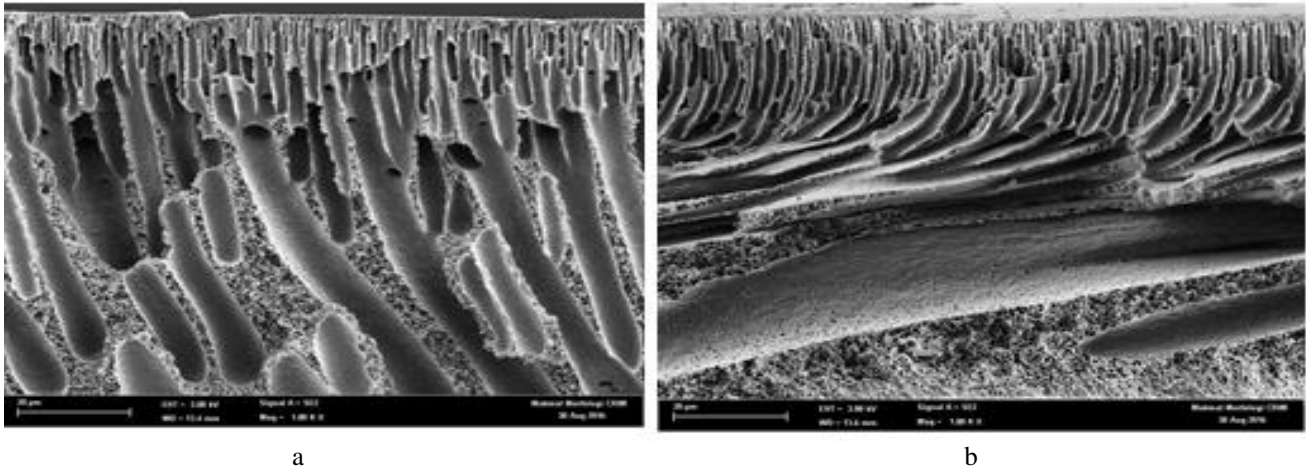

a   b

Figure 4: FESEM cross sectional images of (a) PES and (b) GO-PES membranes

4. Conclusion

GO nanocomposites are promising for high membrane performance to obtain sustainable clean water. In addition, from the aspects of nature of GO hydrophilicity, the membrane morphology and antifouling properties provided efficient outcomes. As a result, it could be concluded that incorporating GO NPs into PES membranes was an advanced method for developing superior membranes with increased hydrophilicity and fouling control that were ideal for a variety of purification and environmental applications.

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