



RR198 and RG19 Dye Removal by using PES membrane modified with graphene oxide nanofillers

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ABSTRACT: The presence of the even very low concentration of dyes makes it undesirable due to its appearance. Most of the organic dyes have aromatic rings in their structure that, make them very toxic and non-biodegradable. So removal of this dyes from industrial effluents before discharging in natural waters is very important. In the present study, Graphene-oxide (GO) nano-sheets were first synthesized using the modified Hammer method and then magnetic graphene oxide (mGO) was prepared using a solvothermal method. The properties of nanofillers were investigated using SEM, XRD and VSM analyzes. The synthesized GO and mGO were embedded in the matrix of polyethersulfone (PES) using phase inversion technique in order to improve its hydrophilicity, permeability, antifouling properties, and rejection performance. Surface and cross-sectional morphology of the resulted bare and nanocomposite membranes were characterized by SEM images. The effect of blended nanoparticles on membrane hydrophilicity and performance were determined using water contact angle, pure water flux, BSA solution filtration, and Reactive Green 19 (RG19) and Reactive Red 198 (RR198) dye solution rejection. Cross-sectional SEM images of the prepared membranes presented an asymmetric structure with a finger like porous sub-layer and dense top-layer. The water contact angle for the bare PES, PES-GO 0.25 wt%, and PES-mGO 0.25wt% were 61.17°, 55.11°, and 51.04°, respectively. The pure water flux of the blended PES membranes was enhanced significantly compared to the bare PES due to the higher hydrophilicity. The results of antifouling properties using BSA filtration demonstrated that the PES-mGO 0.25wt% had the best antifouling properties. Values of flux recovery ratio for the bare PES, PES-GO 0.25 wt%, and PES-mGO 0.25wt% were 45.0, 67.0, and 72.7%, respectively. Dye rejection performance also was increased for the PES-GO 0.25 wt% and PES-mGO 0.25wt% compared to the bare PES. . Compared to the all fabricated membranes, PES-mGO 0.25wt% showed the highest hydrophilicity, permeability, rejection, and antifouling properties.

1. Introduction

Dyes are known as a group of toxic organic pollutants, present in many industrial wastewaters such as plastic, textile, dyestuff, paper, leather, etc., with the annual production of about 700,000 tons [1]. Textile industry is known as an industry producing a huge amount of colored wastewater [2]. Membrane separation is considered as one of the most effective technologies in the field of water and wastewater treatment [3]. membrane separation technology has attracted great attention as a low-cost, easy-operation, environmental-friendly and highly-efficient technology [4].

The goal of the present study is preparation and characterization of polyethersulfone (PES) membrane modified with graphene oxide (GO) and magnetic graphene oxide (mGO) and investigation and comparison their permeability, antifouling properties and rejection performance.

2. Methodology

Graphene oxide was synthesized by using modified Hummer's method [5] and the magnetic graphene oxide was obtained using a solvothermal method [6]. The resulted nanofillers were characterized by using scanning electron microscopy (SEM), X-ray diffraction (XRD), Fourier transform infrared (FTIR), and vibrating sample magnetometer (VSM) techniques.

Nanocomposite PES membranes (including bare PES, PES-GO 0.25wt%, and PES-mGO 0.25wt%) were prepared using the nonsolvent induced phase inversion technique using DMAc as polymer solvent [7]. The surface and cross-sectional morphology of the resulted membranes was characterized using FESEM. The surface hydrophilicity was measured by using contact angle technique. The overall porosity of the prepared membranes was also determined based on gravimetric method.

The performance of the prepared nanofiltration membranes

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was investigated using a dead-end filtration system with a cell capacity of 200 mL and membrane effective area of 19.6 cm² at 3bar operation pressure and room temperature. Antifouling properties was investigated using BSA filtration and rejection performance was determined using reactive green 19 (RG 19) and reactive red 198 dyes (RR 198).

3. Discussion and Results

Cross-sectional SEM images (Fig. 2) present an asymmetric structure with a finger like porous sub-layer and dense top-layer. It is well-known that the dense top-layer limits permeability and enhance separation performance [8]. The results of the water contact angle measurement are showed a decreasing trend by addition of GO and mGO to the PES matrix. Values of water contact angle for the bare PES, GO-PES 0.25wt%, and mGO-0.25 wt% were determined to be 61.17°, 55.11°, and 51.04°, respectively.

The overall porosity of the membrane was also increased by addition of GO and mGO. The GO and mGO modified membranes had a higher porosity (73.9 and 74.6%, respectively) compared to the bare PES (68.5%).

The measured pure water fluxes for the prepared bare and modified membranes are presented in Fig 1. As shown in this Fig, the pure water flux has increased twice for the PES-mGO 0.25wt% (41.0 L/ M² h), compared the bare PES (19.9 L/ M² h). Increasing in hydrophilicity has resulted in an increase in membrane permeability and indicates a close relationship between the hydrophilicity of the membrane surface and PWF. Generally, increasing in hydrophilicity can increase wettability and subsequently permeability of the membrane [9]. The results of Flux recovery ratio (FRR) as the most important fouling index are shown in Fig 2. The FRR results confirmed that addition of GO and mGO has improved antifouling nature of the PES membrane.

The values of FRR for the bare PES and PES- mGO 0.25wt% were calculated to be 45.0 and 72.7%, respectively, showed the excellent fouling resistance of the mGO modified membrane. The trend of FRR changes is harmonized with water contact angle and pure water flux of the prepared membranes.

Dye rejection performance was investigated by filtration of a 100 mg/L solution of two reactive dyes (RG 19 and RR 198) at initial pH of 7. The results of dye rejection performance are presented in Fig 3.

The results showed high performance of the membranes for rejection of both dyes. The rejection performance of RG 19 was higher than RR 198 due to the more molecular weight and bigger size of this dye compared to RR 198.

The rejection performance of PES-GO 0.25wt% and PES-mGO 0.25wt% also was higher than the bare PES due to the more negative charge on the surface of these membranes and repulsion effect.

4. Conclusions

SEM images showed a finger-like cross-sectional structure of the nanocomposite membranes with a smooth surface. Membrane surface hydrophilicity and permeability of the GO and mGO modified membranes was more than the bare PES. Moreover, the fouling resistance of the GO and mGO incorporated membranes was significantly enhanced due to the decreasing water contact angle and increasing surface

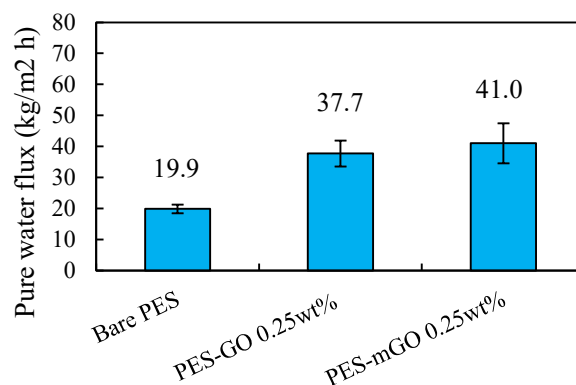


Fig 1. Pure water flux of the prepared membranes

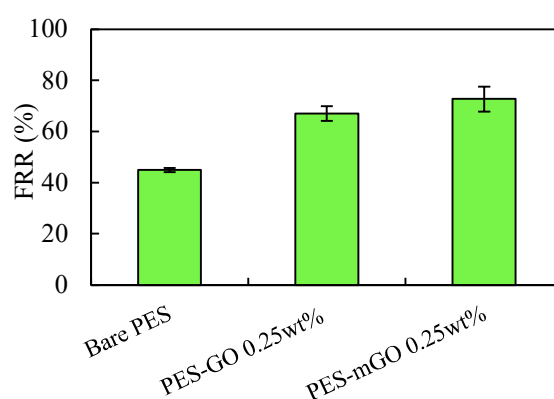


Fig 2. Flux recovery ratio of the prepared membranes

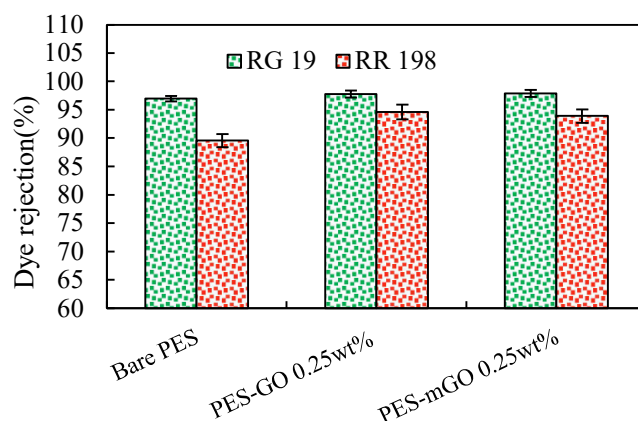


Fig 3. Dye rejection performance of the prepared membranes

hydrophilicity.

The results of dye rejection performance demonstrated high efficiency of the modified membranes for removal of reactive dyes.

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