Chapter 6

Nanocomposite membranes for the removal of dyes

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Abstract

The scarcity of affordable, sustainable, safe, and clean water is one of the major challenges faced by the world. The use of polymeric membranes in wastewater treatment has become a major solution in fighting water scarcity. Some of these membranes (microfiltration, ultrafiltration, and nanofiltration) operate at low pressure when compared to reverse osmosis. However, permeability, selectivity, and fouling limit the application of these polymeric membranes. The incorporation of nanomaterials into the polymeric matrix has resolved such problems in membrane technology. Recent studies show that nanomaterials such as metal organic frameworks (MOFs), graphene oxide (GO), multi-walled carbon nanotubes (MWCNTs), and iron-based nanoparticles are promising nanomaterials for membrane technology with high permeability, selectivity, and antifouling performance. This chapter represents the application of MOF-based nanocomposite membranes for the rejection of anionic and cationic dyes. This chapter reviews various nanocomposite membranes in dye rejection. Conclusions and future perspectives have been drawn and discussed.

Keywords: Membrane technology, Ultrafiltration, Nanomaterials, Cationic dyes, Membrane fouling

1. Introduction

Fast urbanisation and industrialisation cause water contamination and result in a shortage of drinking water (Vila-Traver *et al.*, 2020). Therefore, it is essential to protect the existing water sources by cleaning wastewater contaminated with different pollutants. The removal of dyes and heavy metals has become vital due to their toxicity and irreversibility at wide concentration ranges (Varghese, Paul and Latha, 2019) (Nqombolo *et al.*, 2019). Textile and paper industries generate large amounts of effluent. The discharged effluents contain water pollutants such as salt, suspended organics, toxic ions, coloured organics, and pH (Ahmed *et al.*, 2020). Dyes can be categorised as basic, reactive, acidic, direct or as metal complexes (Minitha *et al.*, 2017) (Chaari *et al.*, 2019). Acidic dyes are negatively charged while basic dyes are positively charged. Synthetic dyes are toxic due to their accumulation in organisms, mutagenic properties, biodegradable and carcinogenic (Almeida and Corso, 2019) (Suzuki *et al.*, 2020). Hence, it is essential to remove dyes before discharging them to the environment (Deng *et al.*, 2019). Membrane technology has attracted more researchers due to its efficiency in water purification (Mansor *et al.*, 2020).

Methylene blue (MB) is considered as one of the hazardous dyes as it is used in industrial applications such as chemicals and textile (Mouni *et al.*, 2018). However, the accumulation of MB is toxic and dangerous to humans. MB is water soluble and is normally used in dyeing leather, colouring paper and as an anticeptic (Fadillah *et al.*, 2019). Researchers have developed various technologies for dye rejection, such as precipitation (Pandey, Singh and Hitkari, 2018), chemical oxidation (Zhu *et al.*, 2020), ultrafiltration (Bouazizi *et al.*, 2017), adsorption (Bhatti *et al.*, 2020), photo-catalysis (Nguyen and Juang, 2019), and coagulation (Beluci *et al.*, 2019). However, post-treatment is required for these methods, hence membranes have been widely used and they have shown great performance in dye rejection.

Membranes can selectively remove targeted pollutants as they have different sizes (Judd, 2017). Membrane filtration processes require pressure and these pressure-driven membranes include microfiltration (Cheng and Hong, 2017), ultrafiltration (UF) (Xu *et al.*, 2018), nanofiltration (NF) (Oatley-Radcliffe *et al.*, 2017), reverse osmosis (RO) (Jiang, Li and Ladewig, 2017), and forward osmosis (M. Zhang *et al.*, 2020). Microfiltration is normally used in removal of yeasts, prokaryotes, fungi, and suspended solids. UF is used in the removal of macromolecules, colloids, and viruses, while NF removes organic matters and heavy metals. RO is used in the production of ultrapure water, water reuse, and salt rejection (desalination). NF and UF have been explored in nanocomposite membranes for dye rejection. These filtration methods are considered to be environmentally friendly, energy-efficient, and low-cost when compared to the reverse osmosis membranes that require high pressure.

Apart from the widespread application of membranes and their efficiency, they are prone to fouling, which becomes a huge setback in their application (Goh *et al.*, 2018). Fouling occurs when hydrophobic polymers interact with the pollutant to form a cake layer. These hydrophobic polymers include polyethersulfone (PES), polysulfone (PSF), and polyvinylidene difluoride (PVDF). These polymers have high mechanical and thermal resistance, firmness and are simple to process. The membrane roughness and hydrophilicity can affect the fouling, which results in decreased water permeation due to pore blocking or formation of cake layer (Yin *et al.*, 2020) (Zheng *et al.*, 2018). Membrane fouling results in a short lifespan of the membrane and low selectivity. Most pollutants are hydrophobic bovine serum albumin (BSA), therefore, increasing the hydrophilicity of the membrane is crucial (Wang *et al.*, 2019). Various methods such as blending of fillers with polymer, blending (Kausar, 2017), and grafting (S. Zhang, Manasa, *et al.*, 2020) have shown great improvement in membrane hydrophilicity.

Blending of different polymers has shown enhanced performance of the polymeric membranes. Also, grafting has demonstrated high protein resistance during rejection. Moreover, composite membrane is one of the trends in improving the performance of the blended polymers with nanofillers. Such nanofillers include metal organic frameworks (S. Zhang, Liu, *et al.*, 2020), TiO_2 (Li *et al.*, 2017), graphene oxide (GO)(Nawaz *et al.*, 2020) and Al_2O_3 (Uzal *et al.*, 2017). This chapter highlights MOF-based nanocomposite membranes for rejection of both anionic dyes and cationic dyes.

2. Application of nanocomposite membranes for dye rejection

Nano-enabled PSF/PVA membranes have been used for the rejection of Congo red. The 0.5 wt% showed high rejection when SiO_2 was used instead of ZnO (Khumalo *et al.*, 2019). The increase in the content loadings of the nanoparticles increased the rejection of Congo red (CR) This is attributed to the electrostatic interaction between the dye (CR) and the membrane surface. Moreover, the size exclusion from the membrane pores also played a role in the rejection process (Khumalo *et al.*, 2019). PVA/PEI nanocomposite membranes used in rejection of three dyes (Congo red, bromomethylmol blue, and direct yellow) exhibited high rejection of 99.7% (Soyekwo *et al.*, 2020). Another study that was carried out by Mehdi and co-workers showed high rejection of reactive red 120; they used PVDF nanofiltration membranes. The increase in contact angle and water permeation was due to the modification of the bare PVDF membranes with HDTMA clinoptilolite nanoparticles (Hosseinifard, Aroon and Dahrazma, 2020).

The study reported by Hebbar showed the rejection of methylene blue and rhodamine B with rejection percentages of 97% and 94%, respectively. This was done at pH 7, using halloysite nanotubes nanocomposite membranes (Hebbar *et al.*, 2018). Clay-hyperbranched epoxy/PPSU composite membranes exhibited high rejection of both dyes when compared to the pristine polymer membrane. Methyl orange (MO) showed higher rejection than methylene blue (MB), which is attributed to electrostatic interaction between the negatively charged MO and clay-hyperbranched epoxy (Mahmoudian and Balkanloo, 2017). The study showed the rejection of CR and MB using PDA/RGO/HKUST-1 yielding a 89.2 % and 99.8 % rejection for CR and MB, respectively (Liu *et al.*, 2019). Also, tannic acid coated boehmite (TA-BM)/PES membranes for the rejection of Direct Red 16 (96%) compared to the PES membrane. The 0.5 wt.% of TA-BM/PES membrane showed high antifouling performance (96% Flux recovery ratio (FRR)) and 3.604 irreversible fouling resistance (R_{ir}) (Oulad *et al.*, 2020). A study reported by Alam and colleagues showed the use of PVDF/ Biopolymer k-carrageenan (kCg) membrane for methyl orange (MO) rejection. The optimum condition (1 wt %) of kCg gave a percentage rejection of 71 % (Alam *et al.*, 2019). Another anionic dye RR198 has been rejected using NCD-blended membranes. The high rejection observed (99,2 %) was due to the electrostatic repulsion between the negatively charged membrane and the anionic dye (Koulivand *et al.*, 2020). SiO_2/Fe_3O_4/PES membranes (Sc-MNP-COOH)/PES for Reactive Green 19, Direct Black 38 and Rhodamine B rejection of more than 90 % due to the electrostatic interaction between the surface of the membrane and the dyes. The bare PES membrane showed low percentage rejection due to the absence of these electrostatic interactions (Vatanpour *et al.*, 2019). Studies showing rejection of anionic and cationic dyes using nanocomposite membranes are shown in Table 1 & 2, the incorporation of nanocomposites in the membranes enhanced the rejection performance of the bare polymer membranes.

Table 1: Nanocomposite membranes for anionic dye rejection

Material	Analytes	Performance (%)	References
DNDs/PES	Reactive Orange 29 and Reactive Green 19	86.9 and 89.4	(Vatanpour et al., 2018)
Fe3O4-MDA	Reactive green 19	Above 98	(Koulivand, Shahbazi and Vatanpour, 2019)
Silver loaded chitosan nanoparticles	Reactive Orange 16 Reactive Black 5	86.13 and 81.21	(Kolangare et al., 2019)
Layered GO (Borate GO)	Methyl orange	74.02	(Yan et al., 2020)
PANI nanofibers	Reactive red 120	99.25	(Kajekar et al., 2015)
(TA-BM)/PES membrane	Licorice and Direct Red	96	(Oulad et al., 2020)
Graphene quantum dots (GQDs)/PVC	Reactive blue	96	(Vatanpour et al., 2020)
PANI-GO/PVDF	Methyl orange	95	(Nawaz et al., 2020)
PES-Fe3O4-APTES	Reactive green 19	96 & 98	(Koulivand, Shahbazi and Vatanpour, 2019)
TOC/PVDF	Eriochrome black T	83.5	(Van Tran, Kumar and Lue, 2019)
H-PAN-ETA	Acid fuchsin	94	(Yun et al., 2020)

Table 2: Nanocomposite membranes for rejection of cationic dyes

Membrane material	**Analytes**	**Performance (%)**	**References**
Layered GO (Borate GO)	Methylene blue	88.56	(Yan et al., 2020)
Ag+-PEI@ HPAN	Crystal violet	99.2	(Liu et al., 2018)
TETA-MWCNT/ PES	Crystal violet and Rhodamine B	98.43 and 99.23	(Peydayesh, Mohammadi and Bakhtiari, 2018)
MIL-53/PVDF UF membrane	Rhodamine B	99.7	Zhao Siyu 2020
PEBAX/CWCTs	Malachite green	98.7	(Mousavi, Asghari and Mahmoodi, 2020)
Nanoparticle/ PES	Rhodamine B	91.96–96.92	(Otitoju et al., 2020)
Pd nanoparticles/ PSF	Crystal violet	99	(Goswami et al., 2018)
GO/PES	Methylene blue	71	(Marjani et al., 2020)
UiO-66/PGPTFC	Methylene blue & Rhodamine B	94	Fang Si-Yuan 2020
Zr-MOF-PUF	Methylene blue & Rhodamine B	97.57 and 98.80	(Li et al., 2018)

3. Conclusion

Ever since membrane technology emerged in water treatment application, much research has been done to modify these membranes to improve their fouling resistance, mechanical or thermal properties. Various nanomaterials have been used to develop hydrophilic polymeric membranes with enhanced properties and performance. MOFs and GO are widely used as fillers in nanocomposite membranes for the removal of both organic and inorganic pollutants from water. In rejection of anionic and cationic dyes, UF and NF membranes have been used with different fillers. Water molecules are transported through these hydrophilic fillers, while rejection occurs due to the electrostatic interactions between the dyes and the filer in the composite membranes. Nanocomposite membranes are potential candidates in water treatment application due to the exceptional properties from the incorporated nanomaterials. The use of nanocomposite membranes in dye rejection has stirred tremendous interest as promising future membranes to treat wastewater treatment.

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