Polymeric Wastewater Purification Membranes

Subjects: Polymer Science

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Over the past few years, polymeric membranes are widely used for applications in separation technologies. They show better flexibility, pore formation mechanism, thermal and chemical stability, and demand less area for installation. Wastewater purification is among the most desirable application of these membranes. In comparison to conventional separation materials, membranes offer economical and efficient treatment. The polymers employed in the synthesis of each membrane are selectively chosen to enhance the optimal performance in wastewater purification.

Keywords: carbon nanotubes ; dendrimers ; waste water purification ; zeolites ; zwitterion

1. Introduction

In India, around 75% of households are short of clean drinking water, and by 2030, 40% of the total population will completely run out of drinking water. Considering water quality, an adequate fresh supply of clean water has become a pivotal challenge. Particularly in India, around 200,000 people die every year because of the unavailability of clean drinking water [1]. Present-day infrastructure for wastewater treatment and safe water production struggles to keep pace with demand within both developed and developing nations.

The exchange of materials between stomachic tissues of the body and the liver is quite similar to the microfiltration and ultrafiltration processes. Membrane technologies are becoming an important aspect of industrial effluent treatment, including those in the pharmaceutical, food, biotechnology, petrochemical, and chemical industries. In comparison to conventional separation materials, membranes offer economical and efficient treatment. Among the investigated materials, polymeric membranes such as polyvinylidene fluoride (PVDF), polyacrylonitrile (PAN), polyvinyl chloride (PVC), polysulfone (PSf), polyethylene (PE), polyamide, polyethersulfone (PES), polyvinyl chloride (PVC), polypropylene (PP), and polyvinyl alcohol (PVA) are applied the most.

Dendrimers are considered to be novel synthetic polymeric nanostructures with balanced structure and distinctive 3-D configuration. Its functional groups are capable of showing intermolecular interactions with various functional moieties. They are non-toxic, cost-effective, and can be prepared from easily available materials. Their adsorption mechanisms, fillers, and the influence of doping materials is intensively reviewed.

Advanced nanomaterials are also well-investigated compounds for water and wastewater purification. Carbon nanotubes and graphene/graphene oxide composites have shown positive results. The cost of fabrication of carbon nanotubes (CNTs) and graphene nano-composites poses a few challenges for their commercialization; however, these materials are the most potential candidates for the future of water purification. The paper also discusses a few chemical modifications for improving membrane performance and neutralizing water-borne bacteria.

Incorporating nanoparticles (NPs) into polymeric membranes is another way of improving membrane properties. Nano-sized metals and their oxides have shown interesting properties when incorporated into polymeric membranes. They have shown enhancements in the optical, electronic, catalytic, and magnetic properties of the membrane [1]. Metal NPs of alumina, zirconium, silver, magnesium, and copper can be employed in almost every polymeric membrane to get targeted characteristics.

Zwitterionic polymers are potential antifouling materials as they are capable of forming a hydration shell through electrostatic interactions. These interactions are much stronger than hydrogen bonds and thus result in closely packed adsorbed water [2]. In the past few years, a wide range of research has been carried out to incorporate zwitterionic materials on the membrane surface. Hence, a brief perspective of the most famous approaches is focused on in the present article.

Thin-film nanocomposite (TFN) membranes were first investigated in 2007, with an aim of enhancing membrane separation capacity, selectivity, and permeability properties. By introducing a minute amount of zeolite into polyamide (PA), water permeability can be highly improved. Zeolites also offer preferential flow paths to water molecules between their super hydrophilic passages and mesoporous structure [3].

2. Carbon-Based Polymeric Membranes

Carbonaceous nanomaterials have high sorption capacity, selectivity, and surface area, due to which they act as potential sorbents to organic solutes in an aqueous medium. Along with this, they also exhibit good thermal conductivity, electrical conductivity, steady reactivity, and exceptional antioxidants. Highly discussed carbon nanomaterials include nanowires, activated carbon, carbon nanotubes (CNTs), diamonds, and fullerenes (C60) [4].

2.1. Carbon Nanotubes-Based Polymeric Membranes

Process simulations are an important part of examining water transportation through CNTs. Thomas and McGaughey [5] conducted the examination of dynamics of water directed by pressure in CNTs by varying the dimensions (i.e., diameter and length) of the tube. They also proposed a method to configure the flow of water in CNTs and its dependence on tube diameter. It was noted that a single string chain of water molecules, having high flow velocity with fixed pressure, is accomplished in tube diameters of 0.83 nm.

CNTs are also very well known for rejecting dissolved ions during water desalination. Instead, there are few effects observed on water flux and design elements to gain the optimal ion rejection at 0.6 nm to 0.8 nm diameter. Therefore, researchers are finding different ways in which CNTs with large diameters can be used for effective ion rejection applications [6]. This addition blocks the entry of charged ions and improves the ion rejection capacity. Various important innovations in wastewater pollutants degradation using CNTs are given in Table 1.

Table 1: Important developments in the field of wastewater purification using different CNTs .

CNTs	Modification technique	Target contaminants	Adsorption capacity	Reference
MWCNTs	-	Eriochrome cyanine R	73.18 mg/g	[7]
Granular CNTs	Al_2O_3 was bonded with g-CNTs	Diclofenac sodium and carbamazepine	157.4 and 106.5 μmol/g	[8]
MWCNTs	Alkali-activated	Magnetic carbon (MO) & Methylene blue (MB)	MO: 2-10 MB: less than 4 and more than 8	[9]
SWCNTs	-	Bisphenol A and 17α-ethinyl estradiol	75-98%	[10]
SWCNTs & MWCNTs	Carboxylated	Oseltamivir carboxylate (OC) & Oseltamivir (OE)	OC: 6-8 OE: alkaline	[11]

MWCNTs	Grafted with PAAM	Humic acid	80-85%	[12]
Polypyrrole/CNTs-$CoFe_2O_4$	one-pot solvothermal method	Methyl blue	500 mg/L	[13]
Ho_2O_3/CNT	MOF assisted route	Tetracycline	98%	[14]

2.2. Graphene/Graphene Oxide-Based Polymeric Membranes

Several methods, such as ball milling, electrochemical exfoliation, and high shear, have been suggested to synthesize graphene at affordable prices. During the early research into graphene, Brodie continuously treated graphite with potassium chlorite ($KClO_3$) and nitric acid (HNO_3) to obtain GO. This method has been regularly refined by adjusting oxidant quantity and time of oxidation, enabling the production of lamellar structures with different sizes. Alternative techniques such as liquid auxiliary electronic stripping [15], potassium ferrate [16], sealed oxidation [17], etc. A few other methods used to prepare GO-based separation membranes include electric field-induced assembly, coating, layer-on-layer self-assembly, filtration, and evaporation. This further undergoes filtration, drying, and other processes, resulting in the formation of a porous membrane layer. The graphene and CNT grains, which were dispersed and reduced, underwent filtration in a vacuum atmosphere within a PVDF UF membrane of 100 mm effective diameter to produce G-CNTs. This method may also be used for preparing GO films, ranging from several nanometers to microns.

Sun *et al.* [18] prepared a new type of nano-filtration (NF)-membrane composite with PAN-UF membrane and coated it with a thin ethanol gel layer. Further, he prepared the GO film through the evaporation method. It was performed at an interface linking liquid and gas. The thicker the interface is, the easier it becomes to prepare a membrane of large transverse size.

GO with more carboxyl, hydroxyl, and epoxy clusters follow the layer-by-layer self-assembly route. In one case, the negative charge and carboxyl and the positive charge of the organic matter group was used to generate self-assembly. In another case, the reactivity of appropriate functional groups was employed to achieve self-assembly. For a carboxyl-rich GO surface, the more probability there is of the creation of a negative charge for groups dispersed in water [19].

Researchers are currently focused on combining the properties of various nanomaterials to produce a single nanocomposite membrane with enhanced performance. Kou and Gao [20] successfully prepared a GO-SiO2nano-hybrid for employment in the applications of super hydrophilic coatings. Gao *et al.* [21] prepared a TiO2(P 25)–GO nanocomposite and found that the photocatalytic activity under the near violet-blue spectrum represents layer-by-layer self-assembly of GO on the top of TiO_2. The adsorption capacities of several other GO-based adsorbents can be further seen in Table 2.

Table 2: Adsorption capacities of various GO-based adsorbents.

GO adsorbent	**Targeted contaminant**	**Adsorption capacity**	**Reference**
PEI-GO	Pb(II)	1000 mg/g	[22]
GO-$MnFe_2O_4$	Pb(II), As(V), As(III)	673, 207, 146 mg/g	[23]
GO-Fe_3O_4	Methyl blue and neutral red	167.2, 171.3 mg/g	[24]

PVP-RGO	Cu(II)	539.53 mg/g	[25]
rGO/ZnO	Methyl blue	80%	[26]
GO/BNC	Congo red and Basic blue	99% and 96%	[27]

3. Zwitter-Ion Based Polymeric Membranes

Antifouling properties of zwitterionic materials follow two main paths when the surface is below water. The first one includes hydration shell formation through electrostatic interactions. According to thermodynamics, foulants require high energy to break the hydration shell. In the case of macromolecular biological foulants, the hydrophilic materials shell creates a small water atmosphere and liberates water to maintain the conformation, hence giving materials lesser adsorption tendencies and high biocompatibility [28].

In zwitterionic polymers, the steric effect works quite similarly to hydrophilic polymer chains, having higher embargo volume by virtue of motility and hydrophobicity. Subsequent experiments were performed for protein absorption using zwitterionic polymers with an unequal charge. The outcome showed that zwitterionic polymers with unequal charges show reduced antifouling properties. Finally, they proposed that good anti-fouling characteristics are assisted by strong hydration, moderate self-associations, and lower protein interactions [29].

The surrounding environmental conditions in which zwitterionic materials operate also play a major role in their anti-fouling activity. In conditions where positive and negative charges have strong interactivity, the zwitterionic polymers tend to aggregate and hence give more surface to external hydrophobic and hydrocarbon groups. This eliminated the steric hindrance and hydration shell. This expands the polymer chain and creates a strong steric hindrance with a dense hydration shell [30].

A new technique is proposed for preparing high permissive polyamide thin-film nanocomposite membranes (TFNMs) by embedding soft zwitterionic copolymers onto sodium carboxy-methyl cellulose. This technique shows improved water permeability and fouling resistance. The synthesized zwitterionic nano-gel (ZNG) shows good interactions with organic particles and polymer grids. Organic materials (i.e., antibiotics) and salts (i.e., NaCl, MgCl, Na2SO4, etc.) can be ideally separated with high water permeation, meaning that ZNG thin-film nanocomposite membranes have the potential of separating organic pollutants and salts [31].

4. Zeolite-Based Polymeric Membranes

Various studies have shown zeolites as a potential material for permeability and selectivity applications. This experiment studied the influence of zeta potential and its content and the micro-channel dimensions and particle size of UF membranes. Introducing Zeolite 4A accurately into a PSf membrane creates swift nanoscale water routed for its flow. This also creates a negatively charged surface, with higher density and roughness.

Han *et al.* [32] experimented with the effect of NaA-zeolite particles in UF membrane composite with poly-(phthalazinone ether sulfone ketone) (PPESK). On examination, membranes displayed high hydrophobicity, water permeability, and antifouling capacity with 3 wt.% NaA. Additionally, the PEG 6000 rejection was 77.9% and 96.8% in the absence and presence of NaA-zeolite particles, respectively. It was further concluded that NaA/PPESK UF-membranes have better performance than commercially prepared UF membranes.

He et al. [33] further experimented with UF-PVDF membranes by adding an MCM-41 zeolite particles into it. These membranes have high permeability and mechanical strength. In comparison to this, pristine UF-PVDF membranes show only 22.5 MPa tensile strength. Additionally, the permeability of zeolite NPs enhanced UF membranes increased from 91,200 M/m2h bar to 118,900 L/m2h bar.

Leo *et al.* [34] used SAPO-44 as a filler against the fouling generated from the humic acid and organic matter present in PSf-UF membranes. The results demonstrated that a membrane containing 15 wt.% SAPO-44 zeolite shows the highest water flux, with a 164% increase as compared to pure membrane. At 15 wt.% SAPO-44, 80% permeate flux was able to be maintained throughout the operation. On the other hand, there was a slight drop in permeability and water flux with 20 wt.% SAPO-44.

Zeolite membranes have also been widely employed in applications such as acid separation, alcohol dehydration, and organic/inorganic separations. It is used where separations are comparatively difficult with conventional techniques. Alumina was used as a support to modify the zeolite membrane. The experiment produced a stable membrane with high separation efficiency.

5. Conclusion

Surface and functionalization are two critical parameters in determining the operational efficiency of new adsorbents. Both specific surface area and intrinsic cavities can bond with pollutants through different interactions. Carbon-based nanomaterials are presently dominating as adsorbents for water treatment. Graphene provides the potential for preparing size-selective membranes due to its excellent mechanical properties and atomic thickness. In CNT and graphene-based membranes, a relatively lower number of components are needed for its operation. Zwitterionic materials have shown potential applications in antifouling membranes through steric hindrance effects. Moreover, Guidelines should be strictly followed about the harmful effects of nanomaterials on human and aquatic habitats.

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