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A Review on the Use of Nanocomposite Membranes as an Emerging Technology for Water/Wastewater Treatment

S. Krishnaveni¹, V. Meera²

P. G. Student, Department of Civil Engineering, Govt. Engineering College Thrissur, India¹

Associate Professor, Department of Civil Engineering, Govt. Engineering College Thrissur, India²

ABSTRACT: Developing cost-effective technologies to solve water pollution problems are critical to the sustainable development of human society. Membrane technology has emerged as a powerful and attractive tool for the removal of hazardous materials such as dyes and heavy metal ions, mainly in trace amounts, from water resources due to its simple operation, no involvement of phase change or chemical additives, and modular design for easy scale up. Among membrane classifications, the nanocomposite membranes play very important role in water as well as wastewater treatment for removing heavy metals and anions. The aim of this review is to summarize the recent scientific and technological advances in the development of nanocomposite membranes for water as well as wastewater treatment.

KEYWORDS: Chitosan, Heavy metals, Nanocomposite membrane, Nanosilica, Polysulfone, Nano titanium dioxide, Water treatment.

I. INTRODUCTION

Water is the source of life for plants, animals, and the human population. However, due to the rapid growth of population, abuse of water resources, and water pollution, water shortage problem becomes more and more serious. Worldwide, around 780 million people still lack access to the improved drinking water sources (according to WHO, 2012) [1]. Hence, cost-effective technologies must be developed to extend water resources and solve water pollution problems. Membrane technology is one of the most promising technologies that may provide a solution to the challenging water problem. It has already been widely used in many areas including drinking water treatment, brackish and seawater desalination, and wastewater treatment. Membranes are favored over many other technologies for water treatment because, in principle, they require no chemical additives or thermal inputs and they do not require regeneration of spent media [2].

Membranes are considered as the key part of membrane separation technology. Membranes are defined as a thin and selective barrier which enables the transport or the retention of compounds between the two media [3]. Membranes can be prepared by inorganic materials (such as ceramics) or organic materials (such as polymers). Inorganic materials including ceramics and metallics usually have better chemical/solvent resistance and could tolerate a wide range of temperature, pH, and pressure. However, they are also restricted by several disadvantages such as limited pore size availability and high operating and capital costs. Accordingly, current research on membranes mainly focuses on the polymeric membranes due to the better control of pore forming mechanism, higher flexibility, smaller footprints required for installation, and lower costs compared to inorganic ceramic membranes [4,5].

Polymeric materials that are commonly used to prepare polymeric membranes include cellulose acetate (CA),



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polysulfone (PSU), polyethylene (PE), polypropylene (PP), poly(vinylidene fluoride) (PVDF), polyethersulfone (PES), polycarbonates (PC) and polyacrylonitrile (PAN). Among them, PSU, PVDF and PES are the most commonly used materials because of their relatively low cost, decent thermal and chemical stabilities [6].

Though polymeric membranes have unique properties which make them better materials for many applications, they also have met some challenges which need to be overcome. The first challenge is the relationship between the permeability and selectivity which was first realized in gas separation membranes. Similarly in water treatment area, membranes with higher water permeability usually possess relatively lower solute rejection. It is the intrinsic property of polymer materials, so it seems not possible to solve the problem just based on the development of polymer material. The second most important challenge towards the membrane usage is obviously membrane fouling, which is the accumulation of retained substances (such as colloidal and suspended particles, organic matter, inorganic matter, and biological foulants) on the membrane surface and/or within the membrane pores leading to the deterioration of membrane performance. Membrane fouling causes a number of problems including the increase in the operational pressure and the decline in the permeate quantity and quality of the membrane systems. The low mechanical strength of polymer materials is also a challenge, especially for a membrane which has a very thin structure [7].

Nanocomposite (combine polymeric materials with nanomaterials) was first developed to improve the mechanical properties of polymeric materials. Recently, nanocomposite membrane has been carried out as a barrier for water and wastewater treatment. The incorporation of nanomaterials with conventional membrane polymers could not only tune structure and its physicochemical properties (hydrophilicity, porosity, charge density, thermal, and mechanical stability) of membranes, but also introduce some unique functionalities such as antibacterial property and photocatalytic capability into the membranes [8].

According to membrane structure and location of nanomaterials, nanocomposite membranes can be classified into four categories: (a) conventional nanocomposite; (b) thin-film nanocomposite (TFN); (c) thin-film composite (TFC) with nanocomposite substrate; and (d) surface located nanocomposite. Nanofillers used in conventional nanocomposite membranes can be inorganic, organic, biomaterial or hybrid one. Thin film composite (TFC) membrane consists of an ultra-thin barrier layer at the top and a more porous supporting layer. The performance of the membrane can be improved by surface modification. TFC with nanocomposite substrate are another class, which was first developed by Pendergast et al. The membranes were prepared by embedding silica nanoparticles in polysulfone substrate using interfacial polymerization process [9]. In surface located nanocomposite membranes also, the surface are modified. The methods for fabricating these membranes include self-assembly, coating, electrostatic attraction, chemical grafting or colloidal precipitation. Addition of nanoparticles to interfacial polymerization processes or surface attachment via self-assembly has introduced the concept of nanocomposite (TFN) membranes, which offer potential benefits of enhanced separation performance, reduced fouling, antimicrobial activity, and other novel functionality. Fabrication of nanocomposite membranes is mostly based on phase inversion (PI) method in which nanofillers are dispersed in polymer solution prior to the PI process, and can be prepared in either flat sheet or hollow fiber configurations. The typical structures of the membranes classified are illustrated in Fig. 1.1.



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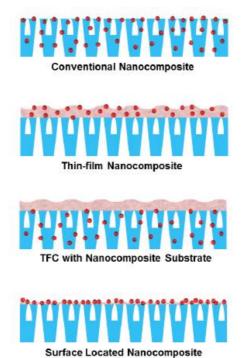


Fig. 1.1 Typical types of nanocomposite membranes related to water treatment applications.

II. EFFECT OF MEMBRANE STRUCTURE AND ITS PROPERTIES

To function as a barrier, the membranes should have the right right structure (such as pore size and porosity) and physicochemical properties (surface hydrophilicity and roughness, chemical, thermal and mechanical stability) suitable for the proposed applications. Surface pore size and membrane porosity will determine the permeability and rejection. Surface hydrophilicity and membrane roughness largely control the fouling characteristics, while chemical, thermal and mechanical stability determine the membrane durability under various environments. Incorporation of various nanofillers into polymer matrices affects the membrane structure and properties. Most studies reported the enhancement of membrane surface hydrophilicity by incorporating hydrophilic nanomaterials that are partially exposed to the surface.

When preparing polyaniline/polysulfone (PANI/PSU) nanocomposite membranes, Fan et al. [10] found that hydrophilic PANI nanofibers migrated from polymer matrix to the surface during the membrane formation, leading to the formation of membranes with higher hydrophilicity, porosity and pore connectivity.

When preparing polysulfone nanocomposite membrane with nanosilica, Saeidi and Zafarmehrabian [11], also observed the migration of hydrophilic nanofibers from polymer matrix to the surface during the membrane formation, which improved the membranes hydrophilicity, porosity and pore connectivity.

Zhao et al. [12] found that hydrophilic PANI nanofibers would migrate toward water bath or the polymer-poor phase of the casting film during the PI process to reduce interfacial energy. The existence of PANI nanofibers near the membrane surface will improve membrane hydrophilicity.

When preparing hyperbranched poly(amine-ester) functionalized multiwalled nanotubes/ poly(vinylidene fluoride) or (MWNTs/PVDF) nanocomposite membranes, Zhao et al. [13] observed a significantly improved surface hydrophilicity, which could be attributed to the increased surface coverage of hydrophilic -OH groups from the nanofillers. In general, increasing the surface hydrophilicity of a membrane decreases membrane fouling because the



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hydrogen bonding formation between the surface hydrophilic groups and water molecules creates a water layer on membrane surface that could prevent foulant adsorption.

Nayak et al. [14] observed an improvement in the membrane surface hydrophilicity while preparing polymeric mixed matrix membranes which were imparted by the addition of TiO_2 nanoparticles. The hydrophilic nature of the prepared membranes was studied by measuring the contact angle for each membrane. Universally, the contact angle continues to decrease as the hydrophilic nature of the membrane increases.

Incorporation of nanomaterials can also change the porosity and pore size of membranes, and subsequently, change their water permeability and solute rejection. Yin et al. [15] studied the membrane properties by incorporating oxidized MWNTs into polysulfone (PSU) polymer matrix and found that its incorporation led to an increase in the surface pore size initially, but afterwards, it shows a decrease with increasing the MWNTs concentration. This trend was consistent with the result of water flux result that showed a convex profile with increase in the nanofiller concentration. Generally saying, the addition of hydrophilic substance in the dope solution leads to an accelerated solvent and non-solvent exchange and thus encourages formation of a more porous structure.

Liao et al. [16] found that zeolite/PVDF nanocomposite membranes possessed enhanced mechanical stability in terms of tensile strength and break elongation. In this case, zeolite NPs acts as a crosslinking agent for the polymeric chains and thereby increases the membrane rigidity.

The changes of membrane structure and property due to the presence of nanomaterials rely on the good dispersion of nanomaterials in casting solution and the polymer matrix also. Many effects were devoted to improve the dispersion of nanofillers either through modifying nanomaterial surface or applying surfactant, especially for those materials that are inherent hydrophobic.

Lee et al. prepared composite polyamide (PA) thin film NF membranes with titania (~60 nm) nanoparticles in the skin layer through interfacial polymerization [167]. As the titania concentration increases towards 5 wt.%, water flux increases and decreases salt rejection, suggesting significant defects formed in the nanocomposite coating film.

III. APPLICATIONS IN WATER AND WASTEWATER TREATMENT

Nanocomposite membranes are not only capable of filtering solute and particles from water, but also suitable for adsorbing, degrading, and/or deactivating them. These membranes are normally designed and optimized with specific applications in mind.

A. ADSORPTION

Usually, nanoparticles are incorporated inside the polymer matrix to make the membranes functionally efficient with specific capability to adsorb heavy metals from wastewater. Daraei et al. [18] incorporated polyaniline (PANI) /Fe₃O₄ NPs inside polyethersulfone (PES) matrix through PI method to remove Cu (II) from aqueous solution. They observed the efficient dispersion of NPs at relatively low loading (0.1%), providing more accessible active sites for sorption of copper ions. Regeneration of this membrane by EDTA is possible for reuse.

Tetala and Stamatialis [19] developed a copper removal membrane by incorporating chitosan beads (20-40 μ m) inside ethylene vinyl alcohol (EVAL) matrix. The membrane showed fast adsorption kinetics and high adsorption capacity (225.7 mg/g) of Cu²⁺ ions was obtained.

Suriyaraj et al. [20] developed a hybrid $Al_2O_3/bio-TiO_2$ nanocomposite (ABN) impregnated electrospun thermoplastic polyurethane nanofibrous membrane and studied its adsorption capacity towards the nitrate. The maximum adsorption capacity of the adsorbent was found to be 14.9 mg/g and developed hybrid adsorbent is biocompatible and safe for drinking water purification. The membrane was used towards the natural ground water and obtained nitrate concentration was found to be within the maximum permissible limit in drinking water.

Gohari et al. [21] developed a polyether sulfone/hydrous manganese dioxide (PES/HMO) ultrafiltration mixed matrix membrane by embedding HMO into PES and studied its adsorption capacity towards Pb (II) from aqueous solution. In another study conducted by Gholami et al. [22] iron oxide (Fe_3O_4) NPs (60 nm) were embedded into the polyvinyl chloride (PVC) matrices to remove lead from aqueous solution.



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Gohari et al. [23] conducted another study by preparing PES nanocomposite membrane for As (III) removal. Fe-Mn binary oxide (FMBO) was used to prepare PES nanocomposite membrane. The maximum adsorption capacity towards As (III) removal was found to be 73.5 mg/g. Water flux of the membrane was as high as 94.6 L/m² h at pressure of 1 bar. The experiment showed that the PES/FMBO mixed matrix membrane can be potentially utilized for effective removal of As(III) from contaminated groundwater by producing permeate containing <10 μ g/L As(III). The membrane adsorption capacity could be regenerated using NaOH and NaOCl solution.

B. PHOTOCATALYTIC ACTION OF NANOPARTICLES

 TiO_2 has been widely used for water splitting, water treatment, air purification and self-cleaning of surfaces because of its unique photocatalytic properties (such as photodegradation and photoinduced super hydrophilicity), stability, commercial availability, and ease of preparation [24,25]. In the nanocomposite membrane research area, TiO_2 has also been incorporated into various membrane matrices to make the membrane photocatalytically active.

Rahimpour et al. [26] studied the effects of UV irradiation on the performance of TiO_2/PES nanocomposite membranes, and found that UV-irradiated TiO_2/PES membranes had higher flux and thereby it enhanced fouling resistance. They attributed this enhancement to the photocatalysis and super hydrophilicity of TiO_2 under the UV irradiation.

Damodar et al. [27] developed a superior membrane by incorporating TiO_2 into PVDF membrane matrix and providing UV irradiation. Incorporation enhanced permeability and fouling resistance, as well as high antibacterial capability of the membrane.

In another study, UV-cleaning process was carried out by Ngang et al. [28] on their TiO₂/PVDF nanocomposite membranes, allowing the recovery of water flux after filtration process. The hydrophilicity of the mixed-matrix membrane was enhanced and resulted in the improved pure water permeability ($392.81 \pm 10.93 \text{ l/m}^2 \text{ h}$ bar). The mixed-matrix membranes were further investigated in terms of filtration, adsorption and UV-cleaning properties based on methylene blue (MB) solution and the membranes showed excellent removal efficiency (~99%) when sodium dodecyl sulfate (SDS) was introduced into the MB feed solution.

C. ANTIMICROBIAL ACTIVITY

The most challenging problem of membrane separation process is obviously membrane biofouling which is due to the microbial growth and biofilm in water and wastewater treatment. It decreases membrane permeability, reduces permeate quality, and increases energy costs of the separation process. Developing antimicrobial membranes will increase membrane efficiency and application duration significantly. In addition, the use of antimicrobial membrane helps to provide pathogen-free clean water.

The most widely used and acceptable antimicrobial agent in water treatment is silver (Ag) due to its excellent biocidal properties. It has been successfully applied in many areas such as antimicrobial plastics, coatings, and wound and burn dressing. Chou et al. [29] incorporated AgNPs into cellulose acetate hollow fiber matrix for anti-bacterial applications. The residual silver content of hollow fiber was about 60% of the original silver added in the polymer solution. Zodrow et al. [30] impregnated AgNPs into polysulfone (PSU) matrix to improve biofouling resistance and virus removal. Silver nanoparticles (nAg) incorporated into polysulfone ultrafiltration membranes (nAg–PSf) exhibited antimicrobial properties towards a variety of bacteria, including *Escherichia coli* and *Pseudomonas mendocina*, and the bacteriophage. Nanosilver incorporation also increased membrane hydrophilicity, reducing the potential for other types of membrane fouling.

Flower et al. [31] fabricated and evaluated the performance Ag-PSf nanocomposite membrane towards microbial filtration of contaminated water. The results show that the membrane has good silver retention indicating that very little of the silver, if any gets into the treated water.

Silver NPs could also be attached onto membrane surfaces through an adsorption-reduction mechanism, where silver ions were first adsorbed by the membrane surface, and then reduced by chemical agents such as formaldehyde, vitamin C and under light irradiation. Using this approach, Zhu et al. [32] prepared chitosan based membranes immobilized with ionic or metallic silver and then compared their anti-biofouling performances by using two typical



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bacteria (*E. coli* and *Pseudomonas sp.*). Although both membranes showed effective anti-bacterial and anti-biofouling activities, the membrane with metallic silver possessed more stable and better overall anti-biofouling performance.

IV. CONCLUSION

In conclusion, nanocomposite membranes are highly efficient in removing various pollutants both organic as well as heavy metals. The incorporation of nanomaterials with conventional membrane polymers could not only tune structure and physicochemical properties such as hydrophilicity, porosity, charge density, thermal, and mechanical stability of membranes, but also introduce unique functionalities which include antibacterial property and photocatalytic capability into the membranes. These membrane technologies are advantageous due to its simple operation, no involvement of phase change or chemical additives, and modular design for easy scale up. In addition, functionalization of these membranes improves the hydrophilic properties and efficiency. Overall, nanocomposite provides polymeric membrane design a new dimension, which could lead to the next generation of high performance membranes.

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