Chapter

Preparation and Applications of Nanocomposite Membranes for Water/Wastewater Treatment

Muharrem Ince and Olcay Kaplan Ince

Abstract

Because of scarcity of clean water all over the globe, it is leads to serious challenges to the survival of all living species. Advanced treatment of water/wastewater techniques such as filtration separation and ion exchange separation are necessary for degradable or non-biodegradable detrimental and hazardous wastes removal from water. Membrane technology is of critical importance to solve this vital problem. In membrane technology, nanocomposite membranes (NCMs) are the most preferred in terms of their convenience. These membranes and their constituent materials are eco-friendly, low-cost, and energy-efficient materials. Also they have operational flexibility and feasibility. The current study presents an overview of the progress in NCMs to treat water/wastewater. To prepare NCMs, various used methods are discussed. Also, to improve the mechanical, antibacterial, and adsorption, properties of NCMs have been investigated. The objective of this work was to summarize the removal of toxic wastes from water/wastewater using various NCMs and to emphasize the shortfalls, and future prospective of NCMs technology are highlighted.

Keywords: nanocomposite membranes, water/wastewater, toxic wastes, membrane technology, adsorption properties

1. Introduction

The rapid growth of the world population and industrial activities has caused a significant increase in water consumption. These developments lead to a serious water shortage all around world especially in arid regions. Recently, the main problem affecting humane societies around the globe is the scarcity of water and increasing demand to it [1, 2]. As stated by United Nations’ reports, worldwide, about 1.2 billion people live in the region of physical scarcity. While another half a billion people are approaching this condition, about two billion people are facing economic water shortage. In addition to the treatment and reuse of wastewater, desalination is one of the technologies widely applied in the world. During recent decades, more than 100 countries have been using these processes [3, 4]. It is not possible to survive without clean water, unfortunately, based on international standards and various organizations, less than 1% of total water is clean. Rest of the water quantity is contaminated by various human-source pollutants such as agricultural activities, municipal wastewater, and industrial wastes [5, 6]. The major
water pollutants can be specified as toxic heavy metals, pesticides, dyes, organic acids, halogenated compounds, fertilizers, and microorganisms [7–10]. Because of non-biodegradability and toxicity, among these pollutants, heavy metals are the most hazardous materials for ecosystem and organism, because these toxic and dangerous metals tend to accumulate in ecosystem especially the food chain and the living organism. In addition, the polluted water intake leads to various health problems, such as organ damage, skin irritation, cancer, rupture of nasal septum, diarrhea, appetite loss, abdominal pain, and headache [11–14]. For the reasons stated above, and especially in order to provide clean water to all living creatures to survive in a healthy life, these pollutants must be removed. Various membrane technologies have recently been used for removing these contaminants from water/wastewater. Among these technologies and applications, those of the greatest interest to researchers are listed below:

• Microfiltration (MF; range from 0.05 μm to 1.0 μm),
• Ultrafiltration (UF; range from 0.005 μm to 0.5 μm),
• Nanofiltration (NF; range from 0.0005 μm to 0.01 μm),
• Reverse osmosis (RO; range from 0.0001 μm to 0.001 μm),
• Forward osmosis (FO),
• Membrane distillation,
• Pressure retarded osmosis (PRO),
• Membrane bioreactor (MBR),
• Pervaporation (PP), and
• Separation using liquid membranes [10].

In the last decade, various water/wastewater purification technologies such as NF, FO, and RO have been developed and effectively used [15]. It is inevitable that membrane-based processes will play an increasingly important role in water/wastewater treatment. These processes are expected to take a key role in solving many problems by developing further in a short period of time due to some advantages such as requiring less energy, ease of use, and making them easily modular [16]. Among the membrane technologies performed in the wastewater treatment, especially application of NF, FO, and NF processes will be increased in the near future [1]. Polymers are widely preferred materials in water/wastewater treatment, despite some disadvantages such as relatively high-energy consumption, permeability, short lifetime, relatively consumption of high energy, and low resistance to fouling. It is vital to develop low-energy, cost-effective, and functional membranes for contaminants removal from water/wastewater. In particular, the inclusion of nano-sized materials in the polymer matrix has made a significant progress in overcoming the challenges of water treatment of polymeric membranes developed and synthesized. Studies conducted in the last few years, especially nano-sized structures such as carbon nanotubes (CNTs), graphene, zeolites, silica, zinc, iron oxide, and other metal oxides, have been added to the polymer matrix and tested [17]. Supported by various nanostructures, NCMS have been used effectively in
many applications including liquid-solid, liquid-liquid, and gas-gas separations. The PCMs have attracted great attention for water/wastewater cleaning because of overcoming trade-off between permeability and solute rejection along with fouling reduction property. Also, for water/wastewater treatment process, they are known as high-performance membrane [18]. As a result, it can be clearly stated that although there are some difficulties in industrial applications, nanomaterials offer outstanding benefits. For example, modification of the NCMs’ surface provides a great advantage in water treatment applications as it significantly changes its efficiency, such as pore size and hydrophilicity [18]. A brief schematic summary of NCM processes to treat water/wastewater is presented in Figure 1.

In order for designing membranes for water/wastewater treatment, various natural and synthetic polymer types, including chitosan, cellulose acetate, polystyrene, polyamide, have been preferred [10, 19, 20]. Barriers including low contaminant removal, low chemical stability under pH change, biological fouling, loss of mechanical strength, and hydrophobicity prevent the widespread application of the polymeric membrane. The advantages and disadvantages of polymers used in NCMs are given in Table 1 [10].

For water/wastewater treatment, nanotechnology has brought a great revolution. During the formation of polymeric nanocomposite membranes (PNCMs) process, when nanoscale entities such as nanoparticles and nanofibers add to PNCMs; it gives them unique properties. In the water/wastewater treatment processes, because of some outstanding properties such as permeability, mechanical and chemical stability, superior flexibility, less installation space requirement along with selectivity to chemical species, and high removal capability, PNCMs have become an ideal choice. The significance of PNCs for water/wastewater treatment can be tracked by the continuous rise in publications, also. Using PNCMs for treatment of water/wastewater is an energy-efficient eco-friendly and technology besides low-cost. Moreover, PNCMs technology can be feasibly combined with various processes [10, 21]. For example, the inclusion of metallic and metallic oxide nanoparticles in the polymer matrix has added antifouling properties to the membranes, as well as increased thermal and mechanical stability. Because of their low cost, they are often added in a small quantity of nanofibers into the polymer matrix. When the nanofibers are added to polymer matrix, in addition to the mechanical strength and thermal stability of polymer-based nanocomposites, its flexibility also increases [22–25]. Nanoparticles
composed of metal or metal oxides, which contain Ag, Cu, TiO$_2$, and Fe$_2$O$_3$, are main examples of nanoparticles [26]. This nanoscale entity class of spherical shape demonstrates some superior properties such as increased lipophilicity and good dispersibility in organic solvent along with chemical stability. Another important class of nanoscales are carbon nanotubes (CNTs), nano-diamonds, and graphene oxides (GO). The GO is a 2D carbon-based nanomaterial, and they contain many oxygenated functionalities such as carbonyl or hydroxyl groups in their interconnected carbon layers. The CNTs with 1D carbon-based tubular layers have often been used in wastewater treatment process. Because of some vital properties including hydrophobic surfaces and low surface energy besides spherical nanoscale such as Cu, TiO$_2$, ZnO entities are often used as disinfectants or antimicrobial agents [26]. Very small spherical nanoscale entities such as nanofibers, nanoplatelets, or polymers can be grown at the surface of the substrate to obtain functional nanoscale structures for applications such as catalysis. Various nanostructures obtained in this way are used for water purification as environmentally friendly, cost-effective, and quality products [10, 27]. A schematic illustration of pressure-driven NCMs for water/wastewater treatment is presented in Figure 2.

This study provides an overview of the applications of these current technologies in water/wastewater treatment for heavy metal removal, focusing on the latest technological developments in this field, as well as techniques for preparing NCMs. The properties and performance of PNCMs will be discussed considering different
polymers and nanoscale entities. It is expected that this study would arouse curiosity and interesting for the development and application of functional NCMs to treatment water/wastewater. Moreover, preparation and application of the NCMs and future prospectives will be discussed.

2. Preparation techniques of PNCMs

Two main factors such as the nature of the polymer and the final required membrane structure play a key role in determining the type of membrane preparation method. Several different techniques may be used for fabricating of polymeric membranes. However, in general, some techniques are often preferred among them, and these are summarized below:

- Interfacial polymerization technique
- Phase inversion technique
- Blending technique
- Electrospinning technique [28].

Unfortunately, current membrane preparation techniques are not suitable for industrial-scale use. Therefore, efforts to develop and promote preparation techniques to overcome the limitations of existing strategies for membrane preparation should be strongly supported (Figure 3).

2.1 Interfacial polymerization technique

The technique called interfacial polymerization (IP) is often preferred for PNCMs production, and it is the most considerable technique for commercially fabricating various essential and important membranes including nanofiltration (NF) and thin-film composite (TFC) besides reverse osmosis (RO). For RO applications, the development and use of interfacial polymerized TFC
membranes were an important milestone in the performance of the membranes [29]. Various types of TFC membrane have been fabricated using IP technique because of noticeably superior properties such as microporous substrate layer and independent optimization of the skin layer. In addition to TFC membranes, RO and NF membranes, which have many uses, were also produced using this useful technique. Barrier membrane layer composition and structural morphology are affected with many factors including monomer concentration, solvent type, reaction time, and subsequent treatment [30, 31]. In the highlighted technique, the interaction of two different monomers is as schematized in Figure 4. Before the polymerization process begins, nano-sized structures are incorporated and polymerization occurs at the interface between the two phases. The distinctive layer produced on the substrate in IP has very less thickness and due to these properties, possesses superior membrane flux. Also, for polymerization, suitable monomers selection can produce selective polymer layers, resistant to chemicals, good thermal stability, and better durability [10, 28].
2.2 Phase inversion technique

For membrane preparation, the most usable method is phase inversion (PI) technique. For different applications, various kinds of morphologies can be obtained when using IP technique [32]. To design membranes, various polymers can be used effectively because the PI method is cost-effective, time-efficient, facile, flexible in use, and feasible to scale-up; therefore, PI technique is used for the manufacture of lab membranes and commercial [33]. All PI membranes are based on polymer precipitation in homogeneous casting solution. The polymer solution precipitation is governed by PI process kinetics and thermodynamics. Therefore, this process affects the prepared membranes final morphology. The precipitation takes place through a demixing process. In this mixing process, the polymer solution is converted from a liquid to a porous solid due to the exchange between solvent and non-solvent [34]. In other words, the PI process is a uniform polymer and a mixing process in which the solution of nanoscale structures is converted into a solid phase in a controlled manner. The PI technique is based on the change in solution stability of the dissolved polymer and nanoscale entity caused by temperature instability, mass change between coagulant bath/non-solvents, and solvent evaporation. The PI technique is frequently selected for preparation of asymmetric PNCMs with thin and dense layers. For membrane preparation (Figure 5), some parameters including solvent, non-solvent, choice of composition of polymer solutions, coagulation bath composition, and film casting conditions are the key parameters. As result, PI technique provides the advantage of large morphological differences by altering process parameters that are mentioned before [35, 36].

2.3 Blending technique

Blending technique (BT) is based on the direct mixing of nanoscale entities such as metal oxide or metal nanoparticles and polymer. Also, to form PNCMs, the BT is the easiest technique. Such mixing/mixing can be accomplished in two ways, solution blending technique and melt blending technique.

Figure 5.
A schematic illustration of phase inversion technique.
2.3.1 Solution blending technique (SBT)

In this technique, whole components such as polymer and nano-dimensional structures are disseminated in an appropriate common solvent. It is supported in the polymer matrix by adding nano-sized structures to the mentioned solvent. In this step, first the surface-modified nanoscale entities are dispersed in the solvent by means of ultrasonic waves. The mixture is then added to the polymer solution to obtain a homogeneous dispersion. The polymer chains remained intact and formed NCM upon solvent evaporation. Also, to select a convenient solvent for proper nanoscale entities and polymer mixing, it should not be forgotten that it is essential. For water-soluble polymers, the SBT is more useful. However, using organic solvents that are toxic and expensive is a main problem in application of SBT \[37\]. If NCM is to be obtained from polymers insoluble in low-boiling-point solvents, SBT is not preferred \[38\].

2.3.2 Melt blending technique (MBT)

NCMs are formed from the polymer melt, after the nanoscale structures and polymers are injected into the extruder and melted at high temperature during intensive mixing. For providing polymer chains mobility, thermal energy is used. Generally, the MBT is preferred because of its environment-friendly nature along with higher effectiveness. At a large scale, some parameters including use of high temperature and setup of processing may lead to limitations to their use \[39\]. During blending, the extruder configuration and screw affect the nanoscale entities dispersion quality, also \[38, 40\]. For preparation of NCMs, both SBT and MBT are simple and frequently used techniques. Generally, BTs are feasible to operate and appropriate for all nanoscale entities types.

2.4 Electrospinning technique

Electrospinning technique (ET) that is simple and effective method is usable for producing fibrous membranes. Because of a number of uses including filtration and desalination, the ET is relatively new. It is a preferred technique for fabricating particularly porous membranes. The ET is often preferred for membrane preparation due to some superior features including excellent interconnectivity besides relatively homogeneous pores distribution. Also, because of nanofibers’ large surface area, these membranes functionalization are easier. The ET is a durable technique with very good control over the membrane structure. On the other hand, dense membranes are not produced using ET, because these membranes are required for a diffusion processes, for example, NF and RO \[41\]. In ET, an application of high potential using a voltage source is made between polymer solution droplet and grounded collector. When electrostatic potential is raised to an adequate level, it overcomes the droplet’s surface tension and forms a charged liquid jet. The solution of polymer contains the nanoscale entities and dissolved polymer. The fiber-containing membranes are perfect in that the fiber and morphology of the aspect ratios of the nano/microfibers can be controlled by variable parameters such as the applied electrical potential level, the flow rate of the membrane solution, and the membrane solution viscosity \[10, 42, 43\].

Finally, because of the forces present between them, jet leaves the tip. During thinning of the polymer jet, solvent phase evaporates and nanofibers are formed. Then, nanofibers are collected on the collector. Owing to rheological properties, jet instabilities arising of polymer melt are important in the shaping of fibrous membrane. By controlling the parameters and operating conditions, the fibrous
membranes properties including morphology, porosity, aspect ratio, pore size distribution, and hydrophobicity can be regulated. Electrospun fibrous membranes are a preferable choice in applications filtration, because morphology, fiber shape, and size can be precisely controlled. It has been reported in many studies that nanostructured morphology and fiber diameter can be significantly affected by applied potential strength, solution feed rates, ionic salt addition, and polymer solution viscosity. Depending on the chosen polymer and its molecular weight, the minimum viscosity is decided [28, 42, 44].

3. Antibacterial, mechanical, and adsorption properties of NCMs

In the processing and application of NCMs for the water/wastewater treatment, biofouling is one of the main drawbacks of membrane technology. Biologically sourced membrane contamination leads to clogging of the pores and thus to a serious decrease in performance. Moreover, biofouling increases the maintenance and operational cost of membranes. It decreases the membrane average lifetime, also. Microbial increase and biofilm formation are the main problems that increase the flow in the membranes and consequently require more energy. To overcome these problems, it is vital to prepare NCMS with antimicrobial activity. Preparation of NCMS with antimicrobial activity both increases the efficiency of the membranes and saves time by shortening the application time. Recently, many researchers have focused on using polymers with biocidal materials in designing NCMS with antibacterial properties. Metal oxides such as Ni$_2$O$_3$ [45], TiO$_2$ [46], and ZnO Al$_2$O$_3$ [47] are frequently preferred because of their biocidal properties, that is, they directly target bacteria. For this reason, to reduce biofouling, metal oxides are commonly used to design the antibacterial NCMS. For example, owing to the outstanding biocidal properties of Ag, it is one of the most studied nanomaterials to create antimicrobial activity. Other nanostructures such as titanium, chromium, and copper are also metals that are highly preferred in antimicrobial NCMS production [18]. The optimal concentrations of most metal oxide nanoparticles used to destroy bacterial cells have no toxic effects on human health, which has also fueled interest in the use of these materials. The PNCM antimicrobial effectiveness is based on the electrostatic interaction between the membrane and bacteria. Commonly, the nanoscale structure found in PNCM contains a positive charge that attracts the negatively charged bacterial cell on their surface. This electrostatic interaction breaks the structural integrity of the bacteria and leads to the bacteria death [48].

The NCMS, during water/wastewater treatment, must possess good mechanical features including toughness, to endure the pressure. Because to define processability and stability besides end use of NCMs, enough mechanical strength is essential. The interaction between nanoscale entities and polymer components is vital in NCMS that impart mechanical properties. Nanoscale entity uniformity, size, and volume fraction affect mechanical properties. The good and homogeneous nanoparticle distribution in the polymer matrix restricts the chain movements and thus increases the mechanical strength. As a result, it can be clearly stated that anisotropy is an important property that is also responsible for its mechanical properties [11, 49, 50].

Several technologies are available for removing pollution including organic and inorganic from water/wastewater. The removal techniques such as chemical precipitation, coagulation/flocculation, membrane processes, reverse osmosis, ion-exchange/solvent extraction, biological operations, ultrafiltration, and adsorption have been used. Other techniques including precipitation and ion-exchange other than adsorption are not preferred because of the production of various secondary
pollutants and their high operating efficiency. Apart from these techniques, the adsorption technique has come to the fore and has been accepted due to some advantages such as simple, efficient, and cost-effective. Adsorption that is most effective techniques is often preferred to remove heavy metals due to flexibility in design and operation. This technique contains a surface phenomenon where pollutants are deposited over the adsorbent surface. Ekstra energy, excess water, or additional chemicals are not used in adsorption process [26]. For the aforementioned reasons, the adsorption technique has become a unique phenomenon in removing contaminants from water/wastewater. Adsorbate and adsorbent surface interaction called physisorption or chemisorption occurs in adsorption process. The pollutants especially heavy metals may interact with the adsorbent surface with various forces including electrostatic interactions, van der Waals, or hydrogen bonding [26].

Functional groups included in PNCMs take part in pollutants and heavy metal ions removal by adsorption and can be regenerated by desorption process [51]. In recent years, various materials including nanoparticles [51] and beads [52] as adsorbent forms have been developed and used. In the last few years, many composite materials such as graphene oxide have widely used to remove heavy metal contaminants as novel adsorbents for the adsorption.

4. New trends for removing hazardous metals from water/wastewater using advanced membrane technologies

Detrimental heavy metals such as As and Ni are the biggest and most important pollutants for ecosystem. These toxic and carcinogenic pollutants can be discharged into the water sources in almost all walks of numerous industrial activities. They have damaged the environment and human health in many aspects. Since these metals, which are harmful and destructive, can enter the human body at more than the allowed concentration and accumulate in our tissues, they cause various harmful health problems. Since metals that are toxic effect are used in many fields of industry, without discharge of their release to the environment is also increasing. Toxic materials especially heavy metals, which spread to the environment and do not degrade, reach people especially through the food chain and water [53–55].

As practical and environmental approach for treating wastewater, separation technologies using membranes have been known as worldwide one of the best technology [56]. Membranes used for this purpose can be divided into two basic classes: inorganic membrane and polymeric membrane (Figure 6). There are four types of membranes, based on pore sizes, which are MF, UF, NF, and RO. It should not be forgotten that, during the water/wastewater treatment, heavy metals ions such as Ni$^{2+}$, Cd$^{2+}$, and Hg$^{2+}$ were tiny, and sometimes they are soluble in which

![Figure 6](image6.png)

*Figure 6.* A schematic illustration of membrane type.
it is necessary to reverse the osmosis membrane’s size [57]. Therefore, recently, materials known as hybrid or specifically adsorptive membranes produced by the combination of adsorption and membrane separation processes have been the focus of many researchers [58, 59].

Because of sieving and surface charge effects, both NF and RO can effectively be used for heavy metal ions removal [58]. At the same time, for modification conventional UF and MF membranes to improve the membranes selectivity toward heavy metal ions, various studies have been made. Studies conducted in recent years for heavy metal ions removal from water/wastewater are summarized below.

For selective ion removal from water, Ag-doped multiwalled carbon nanotube (MWCNT)/polyphenylsulfone (PPSU) was prepared as NCM by Shukla et al. Silver-doped MWCNTs prepared and characterized based on specific surface area and distribution of particle size. For characterization, various properties such as porosity, topography, morphology, surface charge, and contact angles were investigated. To examine mentioned properties, several spectroscopic techniques besides transmission electron microscopy were used. It was stated that Ag-MWCNT/PPSU NCM achieves optimal performance and exhibits unique properties. When PPSU membrane is compared with NCMs, it was mentioned that NCMs exhibit significantly improved selective removal of several ions such as Na⁺, As⁵⁺, and Mg²⁺ ions from aqueous medium. Also, antibacterial activity of Ag-MWCNTs was evaluated using some bacteria such as *Escherichia coli* and is reported that the Ag-MWCNTs exhibited excellent antibacterial activity. Finally, it was emphasized that the applications of developed nanocomposite Ag-MWCN/PPSU membranes, which have antibacterial activity, in removing several metal ions in drinking water applications can be performed successfully [59].

Delavar et al. reported the removal of Cd²⁺ and Cu²⁺ ions using mixed matrix membranes (MMMs) alumina nanoparticles fabricated as UF membranes and incorporated with alumina nanoparticles [60]. The characterization of structural morphology and hydrophilicity of synthesized MMMs was made by using field emission scanning electron microscope (FESEM), water contact angle, and Fourier transform infrared spectroscopy (FTIR) techniques. The alumina and hydrous manganese oxide (HMO) loading affected some properties such as pure water flux, mean pore size, porosity, and water contact angle of the membranes. In the light of this information, the performance of UF membranes for removal of Cd²⁺ and Cu²⁺ ions was also investigated. Based on obtained data from UF experiments, when prepared MMM with a high HMO nanoparticles loaded, it was stated that they have had very fast kinetics and demonstrated the highest Cu²⁺ ions and Cd²⁺ ions removal efficiency (97% and 98%, respectively). This study results indicated that HMO nanoparticles can be a good candidate for preparation of MMMs. Also, to remove Cu²⁺ ions and Cd²⁺ ions from polluted water resources, it can be conveniently used [60].

In another study, to remove Cr(VI) ions using UF membranes that contain cellulose acetate, this is incorporated with TiO₂ nanoparticles [61]. In addition, TiO₂ nanoparticles were preferred to increase the affinity of heavy metal ions to the membrane and increase the removal efficiency. Moreover, TiO₂ nanoparticles’ presence improved the membranes’ antifouling properties because of easily cleaned and regenerated. At pH 3.5, in the presence of aminated TiO₂ nanoparticles, Cr(VI) removal efficiency was achieved as 99.8%. Present in the anions form such as Cr(VI) ions, the protonated amine group on the TiO₂ nanoparticles established electrostatic interaction with the Cr(VI) species. Gebru and Das also reported, after four cycles of washing and regeneration processes, efficiency of removal was only slightly reduced to 96.6% [61].

In another study, for the treatment of water contaminants such as organic fouling agents and toxic heavy metal ions, a thin film composite (TFC) NF membrane
that contains poly(piperazineamide) [poly(PIP)] was developed by Bera et al. It has been reported that the synthesized NCM has high performance in anti-organic fouling, anti-biofouling, and removal of multivalent cations. Also, they reported the thin film nanocomposite (TFNC) NF membranes preparation with improved rejection of heavy metals efficacy, anti-biofouling property, and anti-organic fouling properties compared with that of poly(PIP) TFC NF membrane. Using IP technique, FNC NF membranes were prepared and PEI-polyethylene glycol conjugate and then immobilization of Ag-NP. The IP was performed on a polyethersulfone/poly(methyl methacrylate)-co-poly(vinyl pyrrollidone)/Ag-NP mixture UF membrane support. The synthesized TFNC membranes exhibited a good performance for several heavy metals as >99% for Pb$^{2+}$, 91–97% for Cd$^{2+}$, 90–96% for Co$^{2+}$, and 95–99% for Cu$^{2+}$ at applied 0.5 MPa pressure. It was mentioned that heavy metal ions rejection effect of the modified NF membranes is attributed to the positive surface charge development [62].

In the study carried out by Deng et al., a novel NCM containing improved physical properties and enhanced metal ions removal efficiency was prepared using ET technique. By reacting MWCNT-COOH with polyethylenimine (PEI), modified MWCNTs were fabricated, which was further embedded within polyacrylonitrile (PAN) nanofibers using ET technique. The MWCNT-PEI and NCM (MWCNT-PEI/PAN) properties such as physical properties, morphology, and structure were characterized using various techniques including TEM, SEM, FTIR besides mechanical test and contact angle measurements. When NCMs compare to plain PAN membrane because of hydrophilicity, higher mechanical strength, high permeation, and filtration efficiency, it is undisputed that the NCMs are clearly superior. Experiments studies revealed that synthesized NCMs such as MWCNT-PEI/PAN exhibited higher adsorption capacity for several heavy metals such as Cu$^{2+}$ and Pb$^{2+}$ ions compared with other NCMs. It was reported that Langmuir isotherm and dynamic adsorption results proved that the synthesized and designed NCMs exhibited improved rejection ability for heavy metal ions with a water flux at 145.8 L m$^{-2}$ h$^{-1}$ under 0.2 bar pressure. It is stated that these new and functional membranes synthesized have promising potential for contaminated water treatment due to their heavy metal removal properties [63]. In another study carried out, to functionalize graphene oxide (GO) nanoparticles using polyaniline (PANI), a polymerization technique was performed by Ghaemi et al. After NCMs were prepared by embedding PANI@GO nanoparticles into matrix of PES membrane, it was characterized by SEM and AFM for measuring various properties including porosity and permeability besides mean pore size. A response surface methodology compatible with central composite experimental design was carried out for membrane Pb$^{2+}$ removal performance from water besides to optimize experimental conditions. Although the NPs addition to membrane matrix reduces the porosity, permeability, and hydrophilic properties, it has been reported that Pb$^{2+}$ removal activity is significantly increased. It has been stated that increasing the pH and increasing the proportion of GO particles up to 25% by weight cause an increase in Pb$^{2+}$ removal from the water and almost all Pb$^{2+}$ ions are successfully removed by the NCMs. Ghaemi et al. examined adsorption mechanism, isotherm model, and the kinetic properties along with reusability performance of NCMs. They reported that Langmuir isotherm and pseudo-first order kinetic offered the most appropriate models for Pb$^{2+}$ removal from water using synthesized NCMs [64].

Gohari et al. developed an UF mixed matrix membranes (MMMs) using polyethersulfone (PES)/hydrous manganese dioxide (HMO) for Pb$^{2+}$ adsorption and removal by varying the weight ratio of PES:HMO in the membrane. The HMO loading effect on the membrane in terms of porosity, hydrophilicity, and pure water flux and adsorption capacity MMMs for Pb$^{2+}$ studied, also. Moreover, prepared
membranes properties such as structural morphology using and surface roughness were characterized by using SEM, AFM, and FTIR techniques. It was reported that in spite of pore size of membrane tended to decrease with increasing PES:HMO weight ratio, it has been stated that the water flux of the membrane is not affected. It was found that the Pb$^{2+}$ removal capacity of the MMM prepared with the highest PES:HMO ratio was 204.1 mg g$^{-1}$ and this adsorption capacity was quite promising, when compared with literature. It was observed that Pb$^{2+}$ adsorbed on the membrane can be easily desorbed by using HCl solution. Studies conducted by repeating the adsorption-desorption process proved that this MMM can be used repeatedly [65].

In another study, the synthesis of polymeric membranes based PES and modified by the activated carbon addition and the removal potential of this developed membrane in removing Cu$^{2+}$ ions from the aqueous medium were investigated. It has been reported that after modification of the PES membrane with the activated carbon addition, the retention capacity of Cu$^{2+}$ ions are significantly increased [66]. Moreover, the synthesis of various membranes and their application to various samples for heavy metal removal are summarized in Table 2.

### Table 2.

| Used membrane                                | Removed metal | Reference |
|----------------------------------------------|---------------|-----------|
| PAH and PSF/PAH blend membranes              | Pb$^{2+}$, Cd$^{2+}$ | [67]      |
| Cellulose NCMs                               | Ag$^+$, Cu$^{2+}$, Fe$^{3+}$, Fe$^{2+}$ | [68]      |
| FO membranes                                | Co$^{2+}$     | [69]      |
| CTA-ES membrane                             | Cs$^+$        | [70]      |
| NCMs (PDA/GNPs/PEI)                         | Zn$^{2+}$, Ba$^{2+}$, Ni$^{2+}$, Cd$^{2+}$ | [71]      |
| Fe-Ag/F-MWCNT/PES                           | Cr(VI)        | [72]      |
| Thin-film nanocomposite FO membrane          | Pb$^{2+}$, Cd$^{2+}$, Cr$^{6+}$ | [73]      |
| Ultra-thin NCMs (chitosan/GO NCM)           | Mn$^{2+}$     | [74]      |
| MWCNT/polysulfone composite membranes       | Cr$^{6+}$, Cd$^{2+}$ | [75]      |
| Ag-doped MWCNT NCMs                         | As$^{3+}$, Cr$^{6+}$, Mg$^{2+}$ | [59]      |
| NF membranes (quaternized polyelectrolyte complex membranes) | Na$^+$, Mg$^{2+}$, Ca$^{2+}$, Cu$^+$, Mg$^{2+}$, Zn$^{2+}$ | [76]      |
| Thin-film inorganic forward osmosis membrane| Cd$^{2+}$, Pb$^{2+}$, Cu$^{2+}$ and Zn$^{2+}$ | [77]      |
| Functionalized MWCNTs/PVA nanocomposite films | Zn$^{2+}$, Ni$^{2+}$, Mn$^{2+}$, Cr$^{3+}$, Cd$^{2+}$, Pb$^{2+}$ | [78]      |

5. Conclusions: suggestions and future perspectives

Recently, for water/wastewater treatment, PNCMs that have perfect antibacterial, mechanical, and adsorption properties and have become a globally known and usable method. Because of these outstanding performances, they managed to attract the attention of academia and industry. It is a variety of functional nanoscale materials and different architectures that allow PNCMs to have some outstanding properties. It has enabled an impressive improvement in the treatment of water/wastewater using PNCMs technology, which is open to this and similar developments. Over the past decade, to treat water/wastewater especially for removing toxic heavy metals, immense progress has been made in developing PNCMs. Membranes exhibit a unique useful behavior due to some of their physicochemical aspects.
properties including charge density, porosity, hydrophilicity, mechanical and thermal stability, and nanoscale entities addition. Although two important parameters such as the polymer nature and the final required membrane structure determine the type of the method used in the preparation of the membrane, different methods such as IP and PI are the most frequently used methods for PNCMs preparation. The addition of nanomaterials such as nanofibers is of vital importance in improving the mechanical and, in particular, adsorption capability of membranes.

It can be clearly stated that spherical nanomaterials, for example, metal oxide nanoparticles, protect the membrane against biological contamination and provide antibacterial activity. However, it is clear that the methods for the synthesis, development, and distribution of nanoscale materials in the polymer matrix need more research. These research studies should focus especially on the following subjects:

- The aggregates formation is a major problem for nanoscale entities dispersion into the polymer matrix as homogeneous.
- The compatibility of polymer and nanoscale materials plays a key role in the formation of a successful membrane.
- Focus should be placed on possible solutions for the stability of nanoscale entities in the polymer matrix that affect membrane performance.
- Further studies are needed on the functionalization of the surface of nanosized materials or optimization of the manufacturing process, the ability to increase the dispersion of nanoscale entities into polymer matrices. In the near future, it will be possible to optimize the distribution and hence the interaction between nanoscale entities and the polymer matrix.

While studies have shown that nanomaterials have unique properties that can contribute to the development of high-tech and new NCMs with advanced capabilities to treat water/wastewater, optimizing the durability of nanoscale assets and the loading concentration in NCMs is key to achieving the best performance. On the other hand, recently, for removing or reducing the heavy metals amount in water/wastewater bodies, extensive efforts have been made. Various methods have been applied, each of which has advantages and disadvantages. For removing of toxic metallic ions, membrane technology including UF, NF, RO, and FO membranes exhibits huge potential as it offers different separation mechanisms and a wide range of membrane properties. One of the best examples of this is that the adsorptive UF membrane shows a significant improvement in membrane morphology where the increase in water permeability is achieved. Even at low membrane pressure, mentioned membrane is convenient to treat low metal concentrations by enabling the complete filtration-adsorption metal ions removal. The NF, RO, and FO membranes have proven effective to remove metal ions from water/wastewater. In addition, the NF process has been reported to be efficient and effective even at an industrial level. Because it requires high energy consumption and is therefore costly, the RO method is preferred only to treat water resources that must meet drinking water standards. For these reasons, to reduce costs and expand usage, more research is still needed for RO. If the respective limitations can be overcome, it is highly likely that membrane technology will become a serious alternative method to remove heavy metal ions in the future. In summary, adsorptive membranes have a wide range of applications including wastewater treatment. Advances in the development and manufacture of adsorptive membranes are increasing day by day. Besides adsorptive removal of contaminants from the water/wastewater has
also technologically advanced, it has played an important role with the development of adsorptive membrane technologies. Thus, multi-stage pollutant removal processes, which were previously applied, can now be carried out in a single-stage pollutant removal process. As a result, the contribution of properties such as pore size and surface hydrophilicity of the membrane has not yet been fully explained by researchers. Despite current developments in membrane technology, the practical wastewater applications of PNCMs do not yet have the potential to fully meet expectations. Most of the current investigations on applications of PNCMs are at laboratory scale and unfortunately there are limited studies for industrial production and application. For practical and safe applications, further studies are required to produce economic and industrial-scale membranes.

Conflict of interest

The authors declare no competing interest.

Author details

Muharrem Ince* and Olcay Kaplan Ince

1 Department of Food Processes, Tunceli Vocational School, Munzur University, Tunceli, Turkey

2 Faculty of Fine Arts, Design and Architecture, Department of Gastronomy and Culinary Arts, Munzur University, Tunceli, Turkey

*Address all correspondence to: muharremince@munzur.edu.tr

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