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A review of nanomaterials based membranes for removal of contaminants from polluted waters

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Abstract. Safe water has becoming a competitive resource in many parts of the world due to increasing population, prolonged droughts, climate change etc. The development of economical and stable materials and methods for providing the fresh water in adequate amounts is the need of the water industry. Nanomaterials have unique characteristics e.g., large surface areas, size, shape, and dimensions etc. that make them particularly attractive for removing various contaminants from polluted waters. Nanotechnology based multifunctional and highly efficient membrane processes are providing affordable solutions in the new era that do not rely on large infrastructures or centralizes systems. The objective of the current study is to review the possible applications of the membrane based nanomaterials/composites for the removal of various contaminations from polluted waters. The article will briefly overview the availability and practice of different nanomaterials based membranes for removal of bacteria and viruses, organic compounds and inorganic solutes etc. present in surface water, ground water, seawater and/or industrial water. Finally, recommendations are made based on the current practices of nanofiltration membranes in water industry for a stand-alone membrane filtration system in removing various types of contaminants from polluted waters.

Keywords: contaminants; economical; nanofiltration; membranes; stand-alone

1. Introduction

The world's single largest water problem is its scarcity (Jury and Vaux 2005). There are one billion people who do not have access to a reliable source of safe drinking water and according to this situation, around 6000 children die daily due to dehydration caused by various pathogens in polluted water (Black 1998, Meierhofer 2006). In addition to the environmental, economic, and social impacts of poor water supply and sanitation (Moore *et al.* 2003, Mara 2003, Montgomery and Elimelech 2007, Johnson *et al.* 2008), the supply of fresh water is essential for the safety of children and poor (Theron and Cloete 2002, Eshelby 2007). Despite its critical roles as a resource, projections by as many as 23 UN agencies indicate that due to deteriorating global water supply, an estimated two billion people will lack access to safe drinking water by the middle of this century (Parmar 2003).

There is limited possibility for the growth of fresh water due to competing demands of increasing populations throughout the world and problems of water are expected to increase

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further due to climate change and population growth over next two decades (Vörösmarty *et al.* 2000). Shortage of fresh water supply is also resulted due to exploitation of water resources for domestic, industry and irrigation purposes in many parts of the world (Shannon *et al.* 2008). Rising population and urbanization coupled with climate change may reduce water supply globally during the 21st century (Murad *et al.* 2007, Wheida and Verhoeven 2007) in addition to the increased pressure on freshwater resources due to the increasing world's demand of food, energy etc. (Mulder *et al.* 2010, Godfray *et al.* 2010). Polluting surface/ground water sources in another reason of reduced fresh water supplies (Kemper 2004, Foley *et al.* 2005, Coetser *et al.* 2007).

The occurrence new/emerging micro-contaminants (for example, endocrine disrupting compounds (EDCs)) in polluted waters have rendered existing conventional water/wastewater treatment plants ineffective to meet the environmental standards. The traditional materials and treatment technologies like biological treatment systems such as activated sludge and biological trickling filters are unable to remove a wide range of emerging contaminants (Ozaki 2004, Servos *et al.* 2005, Urase and Kikuta 2005). Similarly, physicochemical treatment such as coagulation, flocculation or lime softening remained ineffective for removing different EDCs and pharmaceutical compounds in various researches (Petrovic *et al.* 2003, Westerhoff *et al.* 2005, Vieno *et al.* 2006). Other advanced methods like chlorination, ozonation, ultraviolet (UV) photolysis and ion exchange are not effective to treat complex and complicated polluted water comprising a wide variety of contaminants (Suffet *et al.* 1995, Becher 1999, Hozalski *et al.* 2001, Szewzyk *et al.* 2000, Adams *et al.* 2002, Zhang and DiGiano 2002, Sadiq and Rodriguez 2004, Gopal *et al.* 2007). This is the right time to address water problems by adopting better purification technologies which can reduce problems of water shortages, health, energy and climate change.

Membrane processes like microfiltration, ultrafiltration, nanofiltration (NF) and reverse osmosis (RO) are considered highly effective processes in removing huge amounts of organic micropollutants (Mulder 1994, Kiso *et al.* 2001, Strathmann 2001, Adams *et al.* 2002, Van der Bruggen and Vandecasteele 2002, Ahmad *et al.* 2004, Bodzek *et al.* 2004, Walha *et al.* 2007, Qin *et al.* 2007). NF and RO have proved to be quite effective filtration technologies in removing micro pollutants (Yoon *et al.* 2004, 2006). RO is relatively more effective than NF but higher energy consumption in RO make it less attractive than NF where removal of pollutants is caused by different mechanisms including convection, diffusion (sieving) and charge effects (Braeken *et al.* 2006). Although NF based membrane processes are quite effective in removing huge loads of micro pollutants (Bolong *et al.* 2009), advanced materials and treatment methods are required to treat newly emerging micro pollutants.

Most research pertaining to fabrication and modification of NF membranes is still focused on synthesis of polymeric NF membranes; however, use of nanoparticles has also been emphasized in many studies. A review on this topic can provide researchers with information pertaining to potential benefits of using such materials for fabrication of NF membranes for treating polluted waters. Among other novel technologies in this category, such as electrospun nanofibrous scaffolds, biomimetic aquaporin and EMT-zeolite NF membranes, NF forward osmosis membranes are the most promising. Fig. 1 depicts the major applications driving trends in studies of fabricating and modifying NF membranes in recent six years.

Nanotechnology has been considered effective in solving water related problems of quality and quantity (Bottero *et al.* 2006). Nanomaterials (e.g., carbon nanotubes (CNTs) and dendrimers) are contributing to the development of more efficient treatment processes among the advanced water systems (Obare and Meyer 2004). This study provides a unique perspective on basic research of nanotechnology for membrane based water/wastewater treatment and reuse. Authors have focused

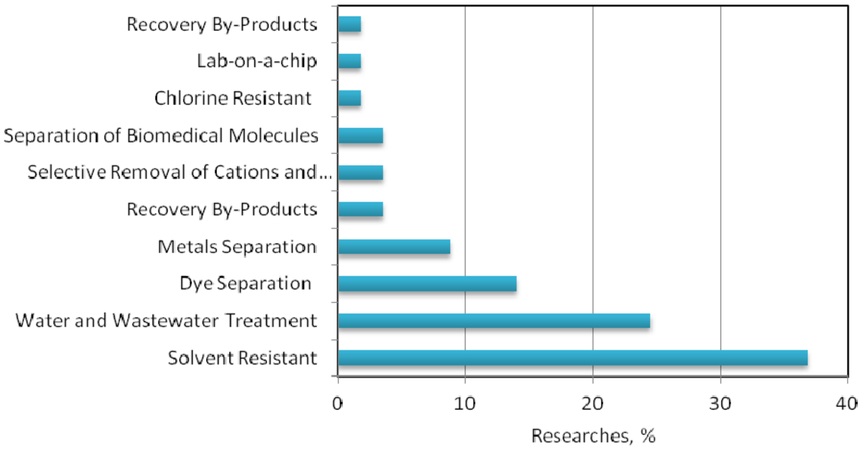


Fig. 1 Major applications driving trends in studies of fabricating and modifying NF membranes, 2007-2012

Table 1 Examples of potential applications of nanotechnology in water/wastewater treatment

Applications	Examples of nanomaterials	Some of novel properties
Adsorption	CNTs/nanoscale metal oxide and nanofibers	High specific surface area and assessable adsorption sites, Selective and more adsorption sites, short intraparticle diffusion distance, tunable surface chemistry, easy reuse etc.
Disinfection	Nano-silver/titanium dioxide (Ag/TiO_2) and CNTs	Strong antimicrobial activity, low toxicity and cost, high chemical stability ease of use etc.
Photocatalysis	Nano-TiO_2 and fullerene derivatives	Photocatalytic activity in solar spectrum, low human toxicity, high stability and selectivity, low cost etc.
Membranes	Nano-Ag / TiO_2 / Zeolites / Magnetite and CNTs	Strong antimicrobial activity, hydrophilicity low toxicity to humans, high mechanical and chemical stability, high permeability and selectivity, photocatalytic activity etc.

on different types of nanomaterials used in fabrication of membranes for treating a variety of contaminants in polluted waters.

2. Nanotechnology for nanofiltration membrane processes

2.1 Process description

Developments in nanoscale research have made it possible to invent economically feasible and environmentally stable treatment technologies to effectively treat the polluted waters meeting the ever increasing water quality standards. Advances in nanotechnology have provided the opportunities to meet the fresh water demands of the future generations. It is suggested that nanotechnology can adequately address many of the water quality issues by using different types of nanoparticles and/or nanofibers (Savage and Diallo 2005). Nanomaterials possess novel and significantly changed physical, chemical and biological properties mainly due to their structure,

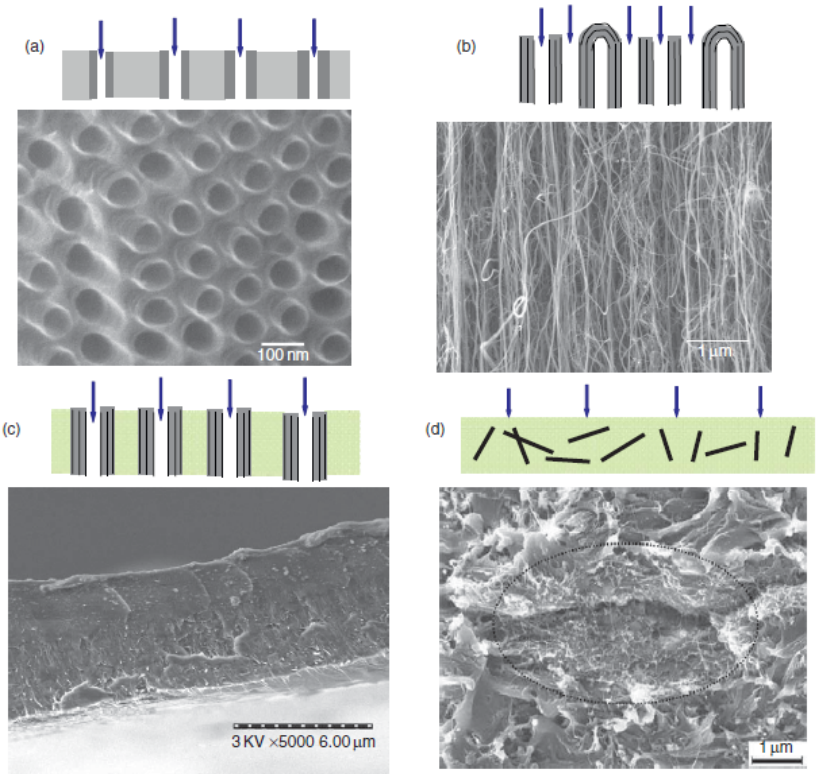


Fig. 2 Examples of different approaches of synthesizing CNT membranes: (a) Majumder and Ajayan (2010); (b) Srivastava *et al.* (2004); (c) Hinds *et al.* (2004), Majumder *et al.* (2007); (d) Geng *et al.* (2007)

higher surface area-to-volume ratio offering treatment and remediation, sensing and detection, and pollution prevention (Rickerby and Morrison 2007, Vaseashta *et al.* 2007).

The unique properties of nanomaterials, for example, high reactivity, strong sorption etc. are explored for application in water/wastewater treatment based on their functions in unit operations as highlighted in Table 1 (Qu *et al.* 2013)**.** Nanoparticles can penetrate deeper and thus can treat water/wastewater which generally is not possible by conventional technologies (Prachi *et al.* 2013). The higher surface area-to-volume ratio of nanomaterials enhances the reactivity with environmental contaminants.

In the context of treatment and remediation, nanotechnology has the potential to promise both water quality and quantity in the long run through the use of, for example, membranes enabling water reuse, desalination. Fig. 2 shows some of approaches for synthesizing CNT membranes with (below) scanning electron micrograph (SEM) images in each of four types.

Nanomaterials have effectively contributed in the development of more efficient and cost-effective water filtration processes since membrane technology is considered one of the advanced water/wastewater treatment process (Bhattacharyya *et al.* 1998, Ritchie *et al.* 1999, 2001, DeFriend *et al.* 2003, Stanton *et al.* 2003, Hollman and Bhattacharyya 2004, Chatterjee *et al.* 2005, Xu *et al.* 2005, Arkas *et al.* 2006, Dotzauer *et al.* 2006, Allabashi *et al.* 2007). Nanoparticles have been used frequently in the manufacturing of membranes allowing permeability control and fouling-resistance in various structure and relevant functionalities (Cortalezzi *et al.* 2003, Li *et al.* 2009a). Both polymeric and inorganic membranes are manufactured by either assembling nanoparticles into porous membranes or by blending process (Bottino *et al.* 2002, Kim *et al.* 2003, Li *et al.* 2009b).

The examples of nanomaterials used in the membrane formation include, for example, metal oxide nanoparticles like TiO_2, CNTs have resulted in desired outputs of improved permeability, inactivation of bacteria etc. (Kim *et al.* 2008, Chae *et al.* 2009). Nanofibrous media have also been used to improve the filtration systems due to their high permeability and small pore size properties (Barhate and Ramakrishna 2007). They are synthesized by a new and efficient fabrication process, namely electrospinning and may exhibit different properties depending on the selected polymers (Frenot and Chronakis 2003). In short, the development of different nanomaterials like nanosorbents, nanocatalysts, zeolites, dendrimers and nanostructured catalytic membranes etc. have made it possible to disinfect disease causing microbes, removing toxic metals, organic and inorganic solutes from polluted waters.

3. Different treatment processes & nanomaterials based membranes

3.1 Disinfection

Nano-sized inorganic material blended composite membranes have proven effective in treating polluted waters because of their enhanced properties, such as high permselectivity, higher hydrophilicity, and enhanced fouling resistance (Yang *et al.* 2007). The efficacy of metal ions in water disinfection haven been displayed by many researchers (Jain and Pradeep 2005). Owing to their charge capacity, they possess antibacterial properties. There are many different types of nanomaterials such as silver, titanium, zinc etc. capable of disinfecting waterborne disease-causing microbes.

3.1.1 Silver nanoparticles

Silver is the most widely used material due to its low toxicity and microbial inactivation in water (Spadaro *et al.* 1974, Zhao and Stevens 1998, Inoue *et al.* 2002, Kumar *et al.* 2004) with well reported antibacterial mechanism (Feng *et al.* 2000, Yamanaka *et al.* 2005). Silver nanoparticles are derived from its salts like silver nitrate, silver chloride, etc. and their effectiveness as biocides are documented in the literature (Sondi and Salopek-Sondi 2004, Baker *et al.* 2005, Panáček *et al.* 2006, Kim *et al.* 2007, Shrivastava *et al.* 2007). Though the antibacterial effect is size dependent (Morones *et al.* 2005), its shape dependency (Pal *et al.* 2007) is also well known. The mechanisms involved during the bactericidal effects of Ag nanoparticles include, for example, the formation of free radicals damaging the bacterial membranes (Xiu *et al.* 2011, 2012), interactions with DNA, adhesion to cell surface altering the membrane properties, enzyme damage etc. (Liau *et al.* 1997, Danilczuk *et al.* 2006, Kim *et al.* 2007).

Immobilized nanoparticles have gained importance due to high antimicrobial activity (Esteban-Cubillo *et al.* 2006). Embedded Ag nanoparticles have been reported very effective against both Gram-positive and Gram-negative bacteria (Savage and Diallo 2005). In a study, the cellulose acetate fibers embedded with Ag nanoparticles by direct electrospinning method (Son *et al.* 2004) have been shown effective against both types of bacteria. Ag nanoparticles are also incorporated into different types of polymers for the production of antimicrobial nanofibers and nanocomposites (Balogh *et al.* 2001, Chen *et al.* 2003, Botes and Cloete 2010). Poly (ε-caprolactone)-based polyurethane nanofiber mats containing Ag nanoparticles was prepared as antimicrobial nanofilter in a study (Jeon *et al.* 2008). Different types of nanofibers containing Ag nanoparticles are prepared for antimicrobial application and exhibited very good antimicrobial properties (Lala *et al.* 2007, Chen and Chiang 2008, Vimala *et al.* 2009). Water filters prepared by polyurethane's foam coated with Ag nanofibers have shown good antibacterial properties against *Escherichia coli* (*E. coli*) (Jain and Pradeep 2005). There are other examples of low-cost potable microfilters prepared by incorporating Ag nanoparticles that can be used in remote areas in developing countries (Peter-Varbanets *et al.* 2009). Ag nanoparticles also find their applications in water filtration membranes, for example in polysulfone membranes (Zodrow *et al.* 2009), for biofouling reduction and have proved effective against variety of bacteria and viruses (Lee *et al.* 2003, Lv *et al.* 2009, Ma *et al.* 2009a, b, 2011, De Gusseme *et al.* 2011, Mauter *et al.* 2011). These Ag nanoparticles laden membranes had good antimicrobial activities against *E. coli*, and *Pseudomonas* etc. (Chou *et al.* 2005, Lee *et al.* 2007). Finally, Ag nanocatalyst alone and incorporated with carbon coverage in alumina have been demonstrated as efficient for degradation of microbial contaminants in water (Chaturvedi *et al.* 2012).

Although Ag nanoparticles have been used efficiently for inactivating bacteria and viruses as well as reducing membrane biofouling, its long-term efficacy against membrane biofouling has not been reported mainly due to loss of silver ions with time (Yu *et al.* 2003, Taurozzi *et al.* 2008). So, further work to reduce this loss of silver ions is required for long-term control of membrane biofouling. Alternatively, doping of Ag nanoparticles with other metallic nanoparticles or its composites with metal oxides nanoparticles can solve the issue and this could also lead to the parallel removal of inorganic/organic compounds from polluted water.

3.1.2 TiO_2 nanoparticles

TiO_2 nanoparticles are among the emerging and promising photocatalysts for water purification (Adesina 2004, Li *et al.* 2008a). The basic mechanism of a semiconductor-based photocatalysts like low-cost TiO_2 having good photoactivity and nontoxicity (Hashimoto *et al.* 2005) involves the production of highly reactive oxidants, such as OH radicals, for disinfection of microorganisms, bacteria, fungi, algae and viruses etc. (Einaga *et al.* 1999, Fujishima *et al.* 2000, Shephard *et al.* 2002, Ibáñez *et al.* 2003, Liu and Yang 2003, Cho *et al.* 2004, 2005, Hajkova *et al.* 2007, Zan *et al.* 2007). The limited photocatalytic capability of TiO_2 i.e., only under UV light, has improved drastically by extending its optical absorbance to the visible-light region (Ni *et al.* 2007, Fujishima *et al.* 2008). This was achieved by doping transition metals (Subramanian *et al.* 2001), and anionic nonmetal such as nitrogen (Asahi *et al.* 2001, Burda *et al.* 2003, Di Valentin *et al.* 2004, Diwald *et al.* 2004, Irie *et al.* 2003, Liu *et al.* 2005, Livraghi *et al.* 2005, 2006, Wong *et al.* 2006, Yang and Gao 2004), carbon (Khan *et al.* 2002, Sakthivel and Kisch 2003, Noworyta and Augustynski 2004, Wang and Lewis 2005, Lin *et al.* 2005, Rincón *et al.* 2005, Mitoraj *et al.* 2007), sulfur (Umebayashi *et al.* 2002, Ohno *et al.* 2003, Yamamoto *et al.* 2004), or fluorine (Yu *et al.* 2002) into TiO_2. Recently, Ag doping of TiO_2 has resulted in improved bacterial inactivation either by complete removal or decreased time of *E. coli* inactivation thereby enhancing disinfection under UV wavelengths and solar radiations (Sökmen *et al.* 2001, Vamathevan *et al.* 2004, Sung-Suh *et al.* 2004, Zhang *et al.* 2005, Page *et al.* 2007, Liga *et al.* 2011).

Nanostructured TiO_2 films and membranes are capable of disinfecting microorganisms in addition to the decomposition of organic pollutants under UV and visible-light irradiation (Choi *et al.* 2009). Due to its stability in water, TiO_2 can be incorporated in thin films or membrane filters for water filtration (Belháčová *et al.* 1999, Kwak *et al.* 2001). TiO_2 nanorods and nanofilms exhibited a higher photocatalytic activity than commercial TiO_2 nanoparticles and TiO_2 thin films, respectively, for the photocatalytic inactivation of *E. coli* (Yu *et al.* 2002, Joo *et al.* 2005, Shieh *et al.* 2006). The inactivation mechanism of *E. coli* when using TiO_2 thin films was also investigated by many researchers (Sunada *et al.* 2003, Kiwi and Nadtochenko 2005). In a study, TiO_2 nanocomposites with multi-walled CNTs showed the complete inactivation of bacterial endospores (Bacillus cereus) as compared with commercial TiO_2 nanoparticles (Lee *et al.* 2005). Immobilized TiO_2 nanoparticle films successfully inactivated *E. coli K12* in surface and distilled water (Alrousan *et al.* 2009). TiO_2 nanoparticles incorporated into an isotactic polypropylene polymeric matrix showed highest biocidal activity against *Enterococcus faecalis* and *Pseudomonas aeruginosa* (Kubacka *et al.* 2009).

The detailed review has demonstrated the antibacterial efficiency of TiO_2 nanoparticles, however, the exact underlying mechanisms are not well defined especially under visible light. In addition, composites of TiO_2 nanoparticles by doping with other metallic nanoparticles have also shown their effectiveness but the applications of using TiO_2 nanofibers and thin film membranes need to be investigated for the effective removal of both inorganic/organic compounds in addition to the disinfection.

3.1.3 CNTs and others

CNTs have proven are very effective in removing bacterial pathogens. CNTs (one of nanosorbents) which have been used for removal of biological impurities have received special attention for its excellent capabilities to remove biological contaminants from water (Savage and Diallo 2005). CNTs possesses antimicrobial characteristics against a wide range of microorganisms including bacteria such as *E. coli*, *Salmonella* etc. (Zhu *et al.* 2006, Deng *et al.* 2008, Upadhyayula *et al.* 2008a, b, Nepal *et al.* 2008, Akasaka and Watari 2009) and viruses (Brady-Estévez *et al.* 2008, Mostafavi *et al.* 2009). Researchers have attributed the antimicrobial effects of CNTs to their unique physical, cytotoxic and surface functionalizing properties (Upadhyayula *et al.* 2009), their fibrous shape (Kang *et al.* 2007, 2008a), the size and length of the tubes, and number of layers (single- or multi-walled) (Wick *et al.* 2007, Li *et al.* 2008a). The mechanisms of killing bacteria by CNTs are also due to the production of oxidative stress, disturbances to cell membrane, etc. (Vecitis *et al.* 2010). Although single-walled CNTs are more detrimental against microorganisms than multi-walled CNTs (Kang *et al.* 2008b) but dispersivity of CNTs is more important parameter than length (Arias and Yang 2009).

Filtration membranes containing radially aligned CNTs are very effective in removing both bacteria and viruses in very short time due to size exclusion and depth filtration (Brady-Estévez *et al.* 2010, Vecitis *et al.* 2011, Rahaman *et al.* 2012) and thus enabling such filters to be used as cost-effective and point-of-use water disinfection devices. CNTs can also reduce membrane biofouling and a nanocomposite membrane of single-walled CNTs and polyvinyl-N-carbazole showed high inactivation of bacteria upon direct contact in a study (Ahmed *et al.* 2012). Another example of controlling the biofouling in thin film nanocomposite membranes is the covalently bonded single-walled CNTs to a thin film composite membrane surface which have exhibited moderate anti-bacterial properties (Tiraferri *et al.* 2011).

3.2 Desalination

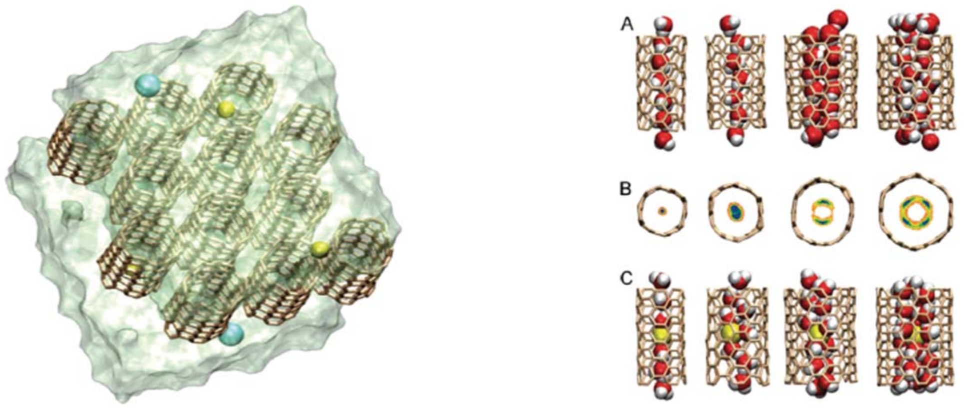
NF has also been evaluated for desalinating seawater in several studies (Mohsen *et al.* 2003).

Nanomaterials are very useful in developing more efficient and economical nanostructured and reactive membranes for water/wastewater treatment and desalination such as CNT filters (Srivastava *et al*. 2004). Nanomaterials offer opportunities to control the cost of desalination and increase its energy efficiency and among these are CNTs (Holt *et al*. 2006, Fornasiero *et al*. 2008), zeolites (Li *et al*. 2004, 2008b) and graphene (Sint *et al*. 2008, Jiang *et al*. 2009, Bai *et al*. 2010). The controlled synthesis of both the length and diameters of CNTs have enabled them to be used in RO membranes to achieve high water fluxes (Pint *et al*. 2008, Arjmandi *et al*. 2009, Song and Corry 2009).

By grafting functional groups, such as carboxyl, at opening of CNTs, membranes have better selective rejection of some components but it has resulted in reduced permeability rendering CNTs incapable for desalination (Nednoor *et al*. 2005, Mauter and Elimelech 2008, Hoek and Ghosh 2009). Hinds concluded a uniform CNT diameter of less than 0.8 nm for high salt rejection (Hinds 2012). Fig. 3(a) shows a simulation system where a CNT membrane was formed by hexagonally packing 12 nanotubes in a periodic cell while Fig. 3(b) shows the water and ions in the nanotubes (Corry 2008).

Thin film nanocomposite membranes containing Ag and TiO_2 nanoparticles exhibited good salt rejection (Lee *et al*. 2007, 2008). Membrane permeability and salt rejection are shown to be affected by the numbers of coating in TiO_2/Al_2O_3 (Aluminum oxide) composite ceramic membranes coated by iron oxide nanoparticles (Fe_2O_3) (Cortalezzi *et al*. 2002, Karnik *et al*. 2005a). A high sodium chloride rejection was obtained by using a alumina ceramic membranes fabricated with silica nanoparticles (Duke *et al*. 2007). Zeolite-based membranes for RO have exhibited high flux with excellent ion rejection characteristics (Jeong *et al*. 2007, Li and Lee 2009). Studies also have indicated the potential of graphene membranes for water desalination with higher fluxes than polymeric RO membranes (Suk and Aluru 2010).

Other nanostructures such as lyotropic liquid crystals, aquaporins etc. also have exhibited high flux and selective water transport (Striemer *et al*. 2007, Gong *et al*. 2007, Zuo *et al*. 2010).



(a) (b)

Fig. 3 (a) A simulation system showing the formation of a CNT membrane; and (b) water and ions in the nanotubes: A – Plane of the CNT membranes; B – Top views of different sizes of the tubes; C – Location of sodium ions

Zeolite–polyamide thin film nanocomposite membranes offered new ways of designing NF and RO membranes with increased water permeability and high salt rejection (Jeong *et al*. 2007, Hoek and Ghosh 2009). The use of nano-zeolites in thin film nanocomposite membranes have resulted in enhanced permeability and salt rejection (Lind *et al*. 2009, 2010). Nanocomposite membranes may serve as ideal membranes for desalination but a basic understanding of transport mechanism along with proper pore size selection by keeping the uniformity is required for economically feasible and commercially acceptable desalination membranes. The effects of real seawater feed on the efficiency of different nanomaterials need to be investigated in terms of long-term operation and maintenance of membrane performance.

3.3 Removal of heavy metals and ions

Different types of nanomaterials have been introduced for heavy metals removal from polluted waters such as nanosorbents including CNTs, zeolites, dendrimers etc. and they have exceptional adsorption properties (Savage and Diallo 2005). Metal based nanomaterials have been shown better in removing heavy metals than activated carbon (Sharma *et al*. 2009), for example, arsenic adsorption by using TiO_2 nanoparticles and nanosized magnetite (Deliyanni *et al*. 2003, Mayo *et al*. 2007).

NF is reviewed for the removal of cations and arsenic from ground/surface waters (Van der Bruggen and Vandecasteele 2003) and it has been shown very effective to remove uranium (VI) from seawater (Favre-Reguillon *et al*. 2003). Novel nanofilter membranes prepared by assembling positive poly (allylamine hydrochloride) and negative poly (styrene sulfonate) onto porous alumina exhibited a high retention of Ca^{2+} and Mg^{2+} (Stanton *et al*. 2003). The incorporation of iron (hydr)oxide nanoparticles into porous carbon materials have made possible to remove both inorganics and organics thus enabling such filters to be used as point-of-use applications (Hristovski *et al*. 2009a, b). A dendrimer-UF system was used for the removal of Cu^{2+} and the complete removal from water was obtained (Diallo *et al*. 2005).

3.4 Removal of organic contaminants

Different types of nanomaterial like nanosorbents such as CNTs, polymeric materials (e.g., dendrimers), zeolites have exceptional adsorption properties and are applied for removal of organics from polluted waters (Savage and Diallo 2005). Photocatalysts like TiO_2 nanoparticles are used effectively for the treatment of water contaminated with organic pollutants like polychlorinated biphenyls (PCBs), benzenes, chlorinated alkanes, etc. (Kabra *et al*. 2004). Decomposition of organic compounds can be enhanced by noble metal doping into TiO_2 due to enhanced hydroxyl radical production etc. (Murakami *et al*. 2009, Han *et al*. 2009, Liu *et al*. 2011).

CNTs are very helpful to improve the material properties of polymers due to high aspect ratio in combination with low density, and high strength and stiffness (Gojny *et al*. 2004). In a study, multi-walled CNTs blended polysulfone microfiltration membranes have shown slightly higher flux and rejection rates than the polysulfone membranes (Choi *et al*. 2006a). Similar results are reported in another study but with lower rejection rates of multi-walled CNT blended polysulfone ultrafiltration membranes as compared with polysulfone membranes (Qiu *et al*. 2009). Recently, multi-walled CNT/polyethersulfone blend membranes showed better hydrophilicity, higher flux and slower fouling rates than the polyethersulfone membranes (Celik *et al*. 2011).

The immobilization of metallic nanoparticles in membrane has also been shown for effective degradation and dechlorination of toxic contaminants (Xu *et al.* 2009). Inorganic membranes containing nano-TiO_2 or modified nano-TiO_2 have been used effectively for reductive degradation of contaminants, particularly chlorinated compounds (Choi *et al.* 2006b, Wu and Ritchie 2008). The use of TiO_2 immobilized on a polyethylene support and a TiO_2 slurry in combination with polymeric membranes have proved very effective in degrading 1,2 dichlorobenzene and pharmaceuticals, respectively (Lin *et al.* 2002, Molinari *et al.* 2002). Polyethersulfone composite membranes with nano-TiO_2 as additive showed higher fluxes enhanced antifouling properties (Yang *et al.* 2007, Rahimpour *et al.* 2008, Wu *et al.* 2008). Ceramic composite membrane made of TiO_2 nanoparticles inside a tubular Al_2O_3 substrate showed improved water quality and flux as compared with Al_2O_3 membranes (Yang and Li 2006). By doping TiO_2 nanoparticles to the Al_2O_3 membrane, it was possible to control the membrane fouling by decreased adsorption of oil droplets to membrane surface in the treatment of oily wastewater (Zhang *et al.* 2009). Nanostructured composite of TiO_2 and Fe_2O_3 incorporated into ultrafiltration membranes successfully reduced the fouling burden and improve the permeate flux (Sun *et al.* 2004).

Cellulose acetate membrane laden with zero-valent iron (nZVI) was found effective in dechlorination of TCE (Wu *et al.* 2005). A reactive membrane of bimetallic nZVI and Pt^0 nanoparticles was found to be very effective at reducing TCE (Meyer *et al.* 2004). Bi-metallic Fe/Ni and Fe/Pd nanoparticles incorporated in nZVI as polymer–inorganic porous composite membranes have been successfully used for the reductive degradation of halogenated organic solvents (Wu and Ritchie 2006, Xu and Bhattacharyya 2007, Tarabara 2009). Polyvinylidene fluoride film containing Pd and Pd/Fe was used effectively for dechlorination of PCB's in one study (Xu and Bhattacharyya 2005) leading to the development of a membrane reactor for dechlorination of a wider range of compounds (Xu and Bhattacharyya 2008, Venkatachalam *et al.* 2008). Alumin–zirconia–titania ceramic membrane coated with Fe_2O_3 nanoparticles was observed to reduce the dissolved organic carbon better than the uncoated membrane enhancing the degradation of NOM (Karnik *et al.* 2005a, b). Finally, ceramic composite membranes of TiO_2 and CNTs have resulted in enhanced membrane permeability and photocatalytic activity (Verweij *et al.* 2007, Yao *et al.* 2008, Yang and Tsai 2008).

NF is reviewed for the removal of NOM and nitrates from ground/surface waters (Van der Bruggen and Vandecasteele 2003) and it has been reported to improve the water quality with a substantial reduction in organic contaminants (Peltier *et al.* 2003). Cost-effective nanostructured and reactive membranes are fabricated using different nanomaterials to develop more efficient water purification methods and in a study, ceramic membrane of Al_2O_3 nanoparticles alone and doped with Fe, Mn and La showed selectivity toward three different synthetic dyes (DeFriend *et al.* 2003). An improved anti-fouling performance and flux increase was also observed in silica incorporated membranes (Jadav and Singh 2009).

In the context of improving the UF processes for water treatment containing organic and inorganic solutes, dendritic polymers are used as water-soluble ligands for, radionuclides and inorganic anions (Ottaviani *et al.* 2000, Birnbaum *et al.* 2003). Nearly complete reduction of 4-nitrophenol was seen when using a composite membrane composed of alumina and polymers through layer-by-layer adsorption of polyelectrolytes and citrate-stabilized Au nanoparticles (Dotzauer *et al.* 2006). Finally, the addition of metal oxide nanoparticles including silica, TiO_2, alumina and zeolites to polymeric ultrafiltration membranes has helped reducing fouling (Bottino *et al.* 2001, Ebert *et al.* 2004, Bae and Tak 2005, Maximous *et al.* 2010, Pendergast *et al.* 2010).

4. Conclusions

Ever increasing population, depleting water resources and climate change resulting prolonging droughts and floods has rendered drinking water a competitive resource in many parts of the world. Traditional water/wastewater treatment technologies remain ineffective for providing adequate safe water due to increasing demand of water coupled with stringent health guidelines and emerging contaminants. The review of literature has shown that water/wastewater treatment using nanomaterials is a promising field for current and future research. Surface modifications of different nanomaterials like nanoscale TiO_2, nZVI etc. by coupling with a second catalytic metal can result enhanced water/wastewater quality when applied for this purpose by increasing the selectivity and reactivity of the selected materials. Bimetallic nanoparticles have also proved effective for remediation of water contaminants. However, further studies are required for comprehending the mechanism of degradation on bimetallic nanoparticles responsible for the improved efficiency. For real field applications, an improved understanding of the process mechanism is very important for the successful applications of innovative nanocomposites for treating polluted waters.

Electrospinning offers the way to modify the surface properties of nanomaterials and different nanofibrous filters have been used successfully as antifouling water filtration membranes. They have extremely high surface-to-volume ratio and porosity, very active against waterborne pathogens, less toxic with minimum health risks, and provide solutions to ensure safe water. It is very easy to dope functional nanomaterials to form multifunctional media/membrane filters with increased reactivity and selectivity for different contaminants. Although these electrospun nanofibers are prepared by simple and cost effective method, there manufacturing at industrial scale is still a challenge and it is vital to consider the subject from an engineering aspect. The use of nanofibrous composites membranes for treating polluted water is very limited and a single stand-alone composite membrane unit of As, TiO_2 and nZVI can be a vital solution for removing various types of pollutants including bacteria/viruses, complex organic compounds, heavy metals and ions.

The use of nanofibers and composite nanostructures membranes can help degrading a wide range of organic and inorganic contaminants in real field applications. The better understanding of nanocomposites membranes formation will certainly be a step towards improving the performance of multifunctional nanocomposites membranes. The pattern of nanoparticles within the host matrices of membranes and change in the structures and properties of both nanomaterials and host matrices could be among the priority concerns in the real field applications of the nanofibrous membranes for removing a wide variety of contaminants from polluted waters.

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