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Review Article

A Review of Removal of Pollutants from Water/Wastewater Using Different Types of Nanomaterials

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The rapidly increasing population, depleting water resources, and climate change resulting in prolonged droughts and floods have rendered drinking water a competitive resource in many parts of the world. The development of cost-effective and stable materials and methods for providing the fresh water in adequate amounts is the need of the water industry. 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. Nanotechnology-based multifunctional and highly efficient processes are providing affordable solutions to water/wastewater treatments that do not rely on large infrastructures or centralized systems. The aim of the present study is to review the possible applications of the nanoparticles/fibers for the removal of pollutants from water/wastewater. The paper will briefly overview the availability and practice of different nanomaterials (particles or fibers) for removal of viruses, inorganic solutes, heavy metals, metal ions, complex organic compounds, natural organic matter, nitrate, and other pollutants present in surface water, ground water, and/or industrial water. Finally, recommendations are made based on the current practices of nanotechnology applications in water industry for a stand-alone water purification unit for removing all types of contaminants from wastewater.

1. Introduction

Water has a broad impact on all aspects of human life including but not limited to health, food, energy, and economy. In addition to the environmental, economic, and social impacts of poor water supply and sanitation [1–4], the supply of fresh water is essential for the safety of children and the poor [5, 6]. It is estimated that 10–20 million people die every year due to waterborne and nonfatal infection causes death of more than 200 million people every year [7]. Every day, about 5,000–6,000 children die due to the water-related problem of diarrhea [8, 9]. There are currently more than 0.78 billion people around the world who do not have access to safe water resources [10] resulting in major health problems. It is estimated that more than one billion people in the world lack access to safe water and within couple of decades the current water supply will decrease by one-third.

The portion of total run-off which constitutes stable run-off flow is considered as the freshwater resource upon which humans depend. This stable fresh water flow has been estimated at 12,500–15,000 km^3 per year [11, 12], from which 4000 km^3 per year is considered to be the total freshwater for irrigation, industry, and domestic purposes [13], and which is estimated to increase to a range of 4300–5000 km^3 per year in 2025 [14–16]. Alternatively, only accessible fresh water is 0.5% of the world's 1.4 billion Km^3 of water which is furthermore poorly distributed across the globe [17].

There is limited possibility of an increase in the supply of fresh water due to competing demands of increasing populations throughout the world; also, water-related problems are expected to increase further due to climate changes and due to population growth over the next two decades [18]. It is estimated that worldwide population will increase by about 2.9 billion people between now and 2050 (according to UN's average projections) [15]. Shortage of fresh water supply is also a result of the exploitation of water resources for domestic, industry, and irrigation purposes in many parts of the world [19]. The pressure on freshwater resources due to the increasing world's demand of food, energy, and so forth [20, 21] is increasing more and more due to population growth and threats of climate change. Polluting surface/ground water sources is another cause of reduced fresh water supplies [22–24]. Aquifers around the world are depleting and being polluted due to multiple problems of saltwater intrusion, soil erosion, inadequate sanitation, contamination of ground/surface waters by algal blooms, detergents, fertilizers, pesticides, chemicals, heavy metals, and so forth [25–28].

The occurrence of new/emerging microcontaminants (e.g., endocrine disrupting compounds (EDCs)) in polluted water/wastewater has rendered existing conventional water/wastewater treatment plants ineffective to meet the environmental standards. The discharge of these compounds into the aquatic environment has affected all living organisms. The traditional materials and treatment technologies like activated carbon, oxidation, activated sludge, nanofiltration (NF), and reverse osmosis (RO) membranes are not effective to treat complex and complicated polluted waters comprising pharmaceuticals, personal care products, surfactants, various industrial additives, and numerous chemicals purported. The conventional water treatment processes are not able to address adequately the removal of a wide spectrum of toxic chemicals and pathogenic microorganisms in raw water.

This is the right time to address water problems since aquifers around the world are depleting due to multiple factors such as saltwater intrusion and contamination from surface waters. Using better purification technologies can reduce problems of water shortages, health, energy, and climate change. A considerable saving of potable water can be achieved through reuse of wastewater which, in turn, requires the development materials and methods which are efficient, cost-effective, and reliable. Although dilution of complex wastewater effluents can help decreasing the load of micropollutants downstream [29, 30], however, much of them pass through conventional water treatment due to occurrence of these substances in micro- or even in nanograms per liter.

Biological treatment systems such as activated sludge and biological trickling filters are unable to remove a wide range of emerging contaminants and most of these compounds remain soluble in the effluent [31–33]. Physicochemical treatments such as coagulation, flocculation, or lime softening proved to be ineffective for removing different EDCs and pharmaceutical compounds in various studies [34–36]. Chlorination, though providing residual protection against regrowth of bacteria and pathogens [37, 38], results in undesirable tastes and odors [39] in addition to the forming of different disinfection by-products (DBPs) in portable drinking water [40–43]. Ozonation has been considered to be a less attractive alternative due to expensive costs and short lifetime. Both ultraviolet (UV) photolysis and ion exchange, though being advanced type of treatments, are not feasible alternatives for micropollutants removal [44].

Membrane processes like microfiltration, ultrafiltration, NF, and RO, which are pressure-driven filtration processes, are considered as some new highly effective processes [44–49]. These are considered as alternative methods of removing huge amounts of organic micropollutants [50–52]. Water/wastewater treatment by membrane techniques is cost-effective and technically feasible and can be better alternatives for the traditional treatment systems since their high efficiency in removal of pollutants meets the high environmental standards [53]. NF and RO have proved to be quite effective filtration technologies for removal of micropollutants [54, 55]. RO is relatively more effective than NF but higher energy consumption in RO makes it less attractive than NF where removal of pollutants is caused by different mechanisms including convection, diffusion (sieving), and charge effects [56]. Although NF based membrane processes are quite effective in removing huge loads of micropollutants [57], advanced materials and treatment methods are required to treat newly emerging micropollutants.

Since the water industry is required to produce drinking water of high quality, there is a clear need for the development of cost-effective and stable materials and methods to address the challenges of providing the fresh water in adequate amounts. There are inventions of new treatment methods; however, they need to be stable, economical, and more effective as compared with the already existing techniques. For this, traditional treatment technologies have to be modernized, that is, updated or modified or replaced by developing materials and methods which are efficient, cost-effective, and reliable. This is particularly important to achieve a considerable potable water savings through reuse of wastewater in addition to tackling the day-by-day worsening quality of drinking water.

Nanotechnology has been considered effective in solving water problems related to quality and quantity [58]. Nanomaterials (e.g., carbon nanotubes (CNTs) and dendrimers) are contributing to the development of more efficient treatment processes among the advanced water systems [59]. There are many aspects of nanotechnology to address the multiple problems of water quality in order to ensure the environmental stability. This study provides a unique perspective on basic research of nanotechnology for water/wastewater treatment and reuse by focusing on challenges of future research.

The paper has three main sections following the introduction which briefly discusses the traditional and current practices in water/wastewater treatment. Section 2 describes mainly the properties and types of nanomaterials and their importance in water/wastewater treatment. Section 3 discusses different types of nanomaterials focusing on membranes for treating a variety of pollutants in water/wastewater. The application of nanomaterials is reviewed based on their functions in unit operation processes. Section 4 provides a summary and outlook in the form of conclusions and recommendations for their full-scale application.

2. Nanotechnology for Water/Wastewater Purification

There are rising demands of clean water throughout the world as freshwater sources/resources are depleting due to prolonged droughts, increasing population, climate changes threats, and strict water quality standards [60, 61]. Masses in developing countries are using unconventional water sources (e.g., stormwater, contaminated fresh water, brackish water) due to limited and depleting fresh water supplies. The existing water treatment systems, distribution systems, and disposable habits coupled with huge centralized schemes are no more sustainable. The current researches do not adequately address the practices that guarantee the availability of water for all users in accordance with the stringent water quality standards [62].

Several commercial and noncommercial technological developments are employed on daily basis but nanotechnology has proved to be one of the advanced ways for water/waste water treatment. Developments in nanoscale research have made it possible to invent economically feasible and environmentally stable treatment technologies for effectively treating water/wastewater 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 [63]. Nanotechnology uses materials of sizes smaller than 100 nm in at least one dimension (Figure 1) meaning at the level of atoms and molecules as compared with other disciplines such as chemistry, engineering, and materials science [64, 65].

At this scale, materials possess novel and significantly changed physical, chemical, and biological properties mainly due to their structure, higher surface area-to-volume ratio offering treatment and remediation, sensing and detection, and pollution prevention [66, 67]. These unique properties of nanomaterials, for example, high reactivity and strong sorption, are explored for application in water/wastewater treatment based on their functions in unit operations as highlighted in Table 1 [68].

Nanoparticles can penetrate deeper and thus can treat water/wastewater which is generally not possible by conventional technologies [69]. 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 provide both water quality and quantity in the long run through the use of, for example, membranes enabling water reuse, desalination. In addition, it yields low-cost and real-time measurements through the development of continuous monitoring devices [70, 71]. Nanoparticles, having high absorption, interaction, and reaction capabilities, can behave as colloid by mixing mixed with aqueous suspensions and they can also display quantum size effects [72–76]. Energy conservation leading to cost savings is possible due to their small sizes; however, overall usage cost of the technology should be compared with other techniques in the market [77]. Figure 2 depicts some of the different types of nanomaterials that can be used in water/wastewater treatment [78–80].

Nanomaterials have effectively contributed to the development of more efficient and cost-effective water filtration processes since membrane technology is considered as one of the advanced water/wastewater treatment processes [81–91]. Nanoparticles have been frequently used in the manufacturing of membranes, allowing permeability control and fouling-resistance in various structures and relevant functionalities [92, 93]. Both polymeric and inorganic membranes are manufactured by either assembling nanoparticles into porous membranes or blending process [94–96]. The examples of nanomaterials used in this formation include, for example, metal oxide nanoparticles like TiO_2. CNTs have resulted in desired outputs of improved permeability, inactivation of bacteria, and so forth [97, 98].

Finally, nanofibrous media have also been used to improve the filtration systems due to their high permeability and small pore size properties [99]. They are synthesized by a new and efficient fabrication process, namely, electrospinning and may exhibit different properties depending on the selected polymers [100]. In short, the development of different nanomaterials like nanosorbents, nanocatalysts, zeolites, dendrimers, and nanostructured catalytic membranes has made it possible to disinfect disease causing microbes, removing toxic metals, and organic and inorganic solutes from water/wastewater. An attempt is made to highlight the factors that may influence the efficiency of the removal processes based on the available literature in the following section.

3. Pollutants Removal Using Different Nanomaterials

3.1. Disinfection. Biological contaminants can be classified into three categories, namely, microorganisms, natural organic matter (NOM), and biological toxins. Microbial contaminants include human pathogens and free living microbes [101–105]. The removal of cyanobacterial toxins is an issue in conventional water treatment systems [106, 107]. Many adsorbents including activated carbon have reasonably good removal efficiencies and again a number of factors influence the removal process [108–111].

Contamination from bacteria, protozoans, and viruses is possible in both ground and surface water. The toxicity of the standard chlorine chemical disinfection in addition to the carcinogenic and very harmful by-products formation is already mentioned. Chlorine dioxide is expensive and results in the production of hazardous substances like chlorite and chlorate in manufacturing process. Ozone, on the other hand, has no residual effects but produces unknown organic reaction products. For UV disinfection, longer exposure time is required for effectiveness and also there is no residual effect. Despite advances in disinfection technology, outbreaks from waterborne infections are still occurring. So, advanced disinfection technologies must, at least, eliminate the emerging pathogens, in addition to their suitability for large-scale adoption. There are many different types of nanomaterials such as Ag, titanium, and zinc capable of disinfecting waterborne disease-causing microbes. Due to their charge capacity, they possess antibacterial properties. TiO_2 photocatalysts and metallic and metal-oxide nanoparticles are among the most promising nanomaterials with antimicrobial properties.

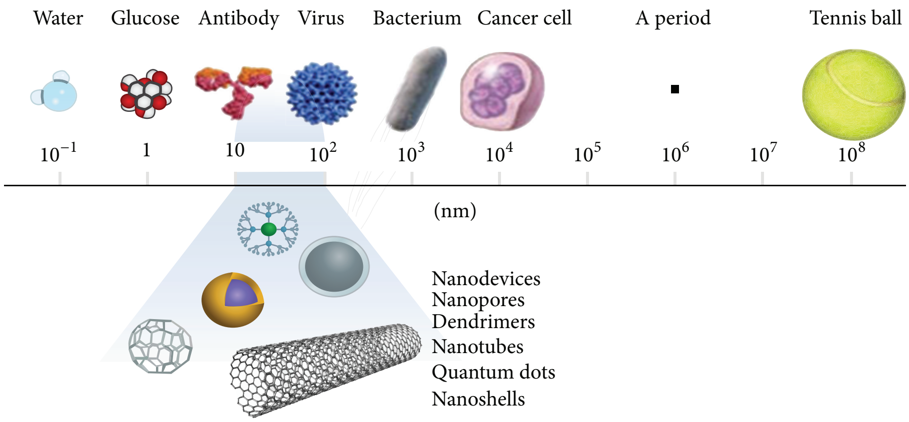


Figure 1: A size comparison of nanoparticle with other larger-sized materials.

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, and so forth.
Disinfection	Nanosilver/titanium dioxide (Ag/TiO_2) and CNTs	Strong antimicrobial activity, low toxicity and cost, high chemical stability ease of use, and so forth.
Photocatalysis	Nano-TiO_2 and Fullerene derivatives	Photocatalytic activity in solar spectrum, low human toxicity, high stability and selectivity, low cost, and so forth.
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, and so forth.

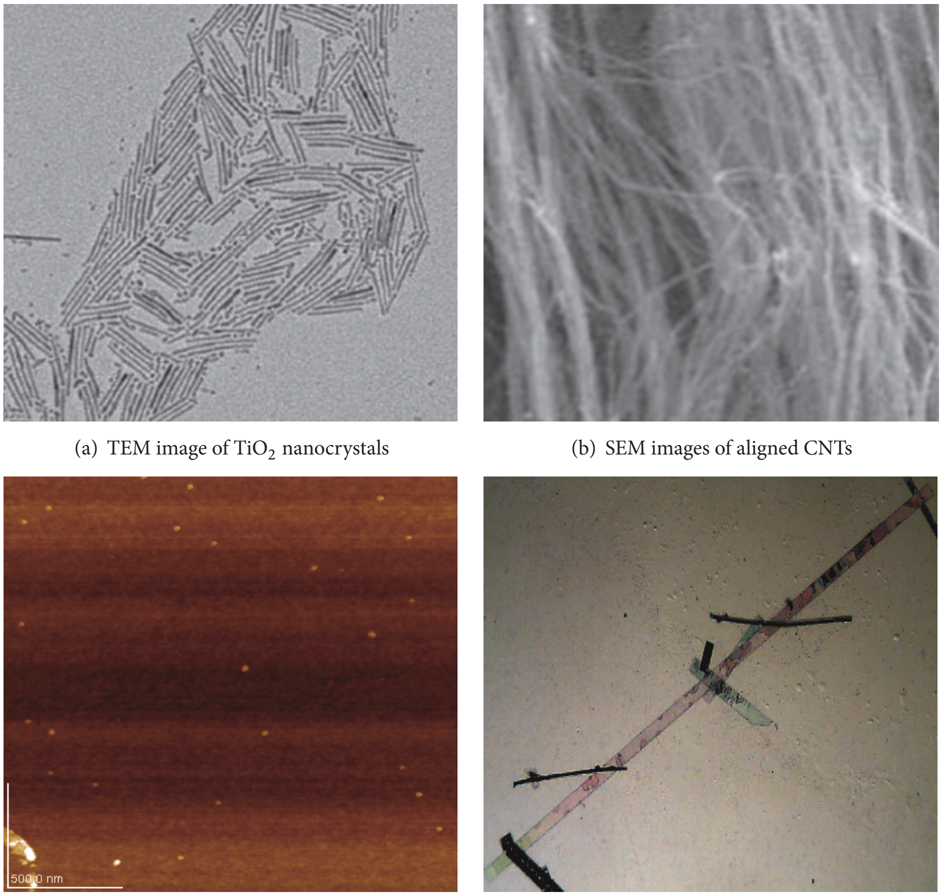
The efficacy of metal ions in water disinfection has been highlighted by many researchers [112]. This part of the paper covers the application of these antimicrobial nanomaterials for water disinfection.

3.1.1. Silver Nanoparticles. Silver is the most widely used material due to its low toxicity and microbial inactivation in water [113–116] with well-reported antibacterial mechanism [117, 118]. Silver nanoparticles are derived from its salts like silver nitrate and silver chloride, and their effectiveness as biocides is documented in the literature [119–123]. Though the antibacterial effect is size dependent [124], smaller Ag nanoparticles (8 nm) were most effective, while larger particle size (11–23 nm) results in lower bactericidal activity [125]. Also, truncated triangular silver nanoplates exhibited better antibacterial effects than the spherical and rod-shaped nanoparticles indicating their shape dependency [126]. The mechanisms involved during the bactericidal effects of Ag nanoparticles include, for example, the formation of free radicals damaging the bacterial membranes [127, 128], interactions with DNA, adhesion to cell surface altering the membrane properties, and enzyme damage [122, 129, 130].

Immobilized nanoparticles have gained importance due to high antimicrobial activity [131]. Embedded Ag nanoparticles have been reported as very effective against both Gram-positive and Gram-negative bacteria [63]. In a study, the cellulose acetate fibers embedded with Ag nanoparticles by direct electrospinning method [132] were 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 [133–135]. Poly (ε-caprolactone-) based polyurethane nanofiber mats containing Ag nanoparticles were prepared as antimicrobial nanofilters in a study [136]. Different types of nanofibers containing Ag nanoparticles are prepared for antimicrobial application and exhibited very good antimicrobial properties [137–139]. Water filters prepared by polyurethane's foam coated with Ag nanofibers have shown good antibacterial properties against *Escherichia coli* (*E. coli*) [112].

There are other examples of low-cost potable microfilters prepared by incorporating Ag nanoparticles that can be used in remote areas in developing countries [140]. Ag nanoparticles also find their applications in water filtration membranes, for example, in polysulfone membranes [141], for biofouling reduction and have proved effective against variety of bacteria and viruses [142–148]. These Ag nanoparticles laden membranes had good antimicrobial activities against *E. coli*, *Pseudomonas*, and so forth [149, 150].

Finally, Ag nanocatalyst alone and incorporate with carbon covered in alumina has been demonstrated as efficient for degradation of microbial contaminants in water [151].



(a) TEM image of TiO_2 nanocrystals

(b) SEM images of aligned CNTs

(c) AFM image of Zinc oxide (ZnO) nanoparticles

(d) Optical microscope image of Sn-doped ZnO nanobelts

Figure 2: Examples of different types of nanomaterials including particles, crystals, tube, and belts.

Although Ag nanoparticles have been used efficiently for inactivating bacteria and viruses as well as reducing membrane biofouling, their long-term efficacy against membrane biofouling has not been reported mainly due to loss of silver ions with time [152, 153]. 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-oxide nanoparticles can solve the issue and this could also lead to the parallel removal of inorganic/organic compounds from water/wastewater.

3.1.2. TiO_2 Nanoparticles. TiO_2 nanoparticles are among the emerging and most promising photocatalysts for water purification [154, 155]. The basic mechanism of a semiconductor-based photocatalysts like low-cost TiO_2 having good photoactivity and nontoxicity [156] involves the production of highly reactive oxidants, such as OH radicals, for disinfection of microorganisms, bacteria, fungi, algae, viruses, and so forth [157–165]. TiO_2, after 8 hours of simulated solar exposure, has been reported to reduce the viability of several waterborne pathogens such as protozoa, fungi, *E. coli*, and *Pseudomonas aeruginosa* [166]. A complete inactivation of fecal coliforms under sunlight is reported in a study expressing the photocatalytic disinfection efficiency of TiO_2 [167].

The limited photocatalytic capability of TiO_2, that is, only under UV light, has improved drastically by extending its optical absorbance to the visible-light region [168, 169]. This was achieved by doping transition metals [170] and anionic nonmetals such as nitrogen [171–180], carbon [181–187], sulfur [188–190], or fluorine [191] 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 [192–197].

As highlighted, the synthesis of visible-light-activated TiO_2 nanoparticles has attracted considerable interest [198], and TiO_2 nanoparticles and nanocrystallines irradiated with UV-visible light exhibited strong bactericidal activity against *E. coli* [199–202]. Metal-doped TiO_2 nanoparticles, sulfur, and iron in couple of studies have shown strong antibacterial effects against *E. coli* [203, 204]. TiO_2 was further modified by nanoparticles of transition metal oxides and nanostructured

TiO_2 photocatalysts showed great potential for water disinfection. For example, metal-ion-modified nitrogen-doped TiO_2 nanoparticle photocatalysts (palladium (Pd) in a study) have shown enhanced disinfection efficiency against *E. coli* (Gram-negative bacterium), *Pseudomonas aeruginosa*, and *Bacillus subtilis* spores due to photocatalytic oxidation under visible-light illumination [205, 206].

Nitrogen-doped TiO_2 nanoparticles catalysts have proved their efficiency for degradation of microbial contaminants in water [151]. 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 [207]. Due to its stability in water, TiO_2 can be incorporated in thin films or membrane filters for water filtration [208, 209]. 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* [191, 210, 211]. The inactivation mechanism of *E. coli* when using TiO_2 thin films was also investigated by many researchers [212, 213]. In a study, TiO_2 nanocomposites with multiwalled CNTs showed the complete inactivation of bacterial endospores (*Bacillus cereus*), compared to commercial TiO_2 nanoparticles [214]. Immobilized TiO_2 nanoparticle films successfully inactivated *E. coli K12* in surface and distilled water [215]. TiO_2 nanoparticles incorporated into an isotactic polypropylene polymeric matrix showed higher biocidal activity against *Enterococcus faecalis* and *Pseudomonas aeruginosa* [216].

The detailed review has demonstrated the antibacterial efficiency of TiO_2 nanoparticles, but the actual 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 films 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 proved to be very effective in removing bacterial pathogens. CNTs (one of nanosorbents) which have been used for removal of biological impurities have received special attention for their excellent capabilities of removing biological contaminants from water [63]. CNTs possess antimicrobial characteristics against a wide range of microorganisms including bacteria such as *E. coli* and *Salmonella* [217–222] and viruses [223, 224]. The adsorption of cyanobacterial toxins on CNTs is also higher when compared with carbon-based adsorbents mainly due to large specific surface area, external diameter of CNTs, large composition of mesoporous volume, and so forth [225–228].

Researchers have attributed the antimicrobial effects of CNTs to their unique physical, cytotoxic, and surface functionalizing properties [229], their fibrous shape [230, 231], the size and length of the tubes, and number of layers (single- or multiwalled) [155, 232]. The mechanisms of killing bacteria by CNTs are also due to the production of oxidative stress, disturbances to cell membrane, and so forth [233]. Although single-walled CNTs are more detrimental against microorganisms than multiwalled CNTs [234], dispersivity of CNTs is a more important parameter than length [235]. Many researchers observed an extremely high adsorption rate of bacteria by single-walled CNTs, in addition to their high sorption capacities by many researchers [236–244].

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 [245–247] and thus enable 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 [248]. Another example of controlling the biofouling in thin film nanocomposite membranes is the single-walled CNTs covalently bonded to thin film composite membrane surface which have exhibited moderate antibacterial properties [249].

Among the other nanomaterials for microbial disinfection, bifunctional ferrous oxide (Fe_3O_4) @Ag and Fe_3O_4 @TiO_2 nanoparticles were employed successfully against pathogenic bacteria including *E. coli*, *Staphylococcus epidermidis*, and *Bacillus subtilis* thus covering both Gram-positive and Gram-negative bacteria as well as spores [250, 251]. Magnesium oxide nanoparticles were also used effectively as biocides against both Gram-positive and Gram-negative bacteria and bacterial spores [252, 253].Nanotungsten oxide and palladium-incorporated ZnO nanoparticles have shown good antibacterial properties at removal of *E. coli* from water [254, 255]. Nitrogen-doped ZnO and Zirconium oxide nanoparticles also have proved good antibacterial nanostructured materials [151, 256].

3.2. Desalination. Desalination is considered an important alternative for obtaining fresh water source. Though expensive, membrane based desalination processes cover most of the desalination capability out of which only RO accounts for 41% [257]. Parameters that control the desalination cost include maximizing the flux of water through membrane to minimize the fouling. Recent developments in membrane technology have resulted in energy efficiency in RO plants [258]. NF has also been evaluated for desalinating seawater [259].

Nanomaterials are very useful in developing more efficient and cheaper nanostructured and reactive membranes for water/wastewater treatment and desalination such as CNT filters [260]. Nanomaterials offer opportunities to control the cost of desalination and increase its energy efficiency and among these are CNTs [261, 262], zeolites [263, 264], and graphene [265–267]. The controlled synthesis of both the length and diameters of CNTs has enabled them to be used in RO membranes to achieve high water fluxes [268–270].

Thin film nanocomposite membranes containing Ag and TiO_2 nanoparticles exhibited good salt rejection [150, 271]. Membrane permeability and salt rejection are shown to be effected by the number of coatings in TiO_2/Al_2O_3 (aluminium oxide) composite ceramic membranes coated by iron oxide nanoparticles (Fe_2O_3) [272, 273]. A high sodium chloride rejection was obtained by using alumina ceramic membranes fabricated with silica nanoparticles [274]. Zeolite-based membranes for RO have exhibited high flux with excellent ion rejection characteristics [275, 276]. Studies also have indicated the potential of graphene membranes for water desalination with higher fluxes than polymeric RO membranes [277].

Other nanostructures such as lyotropic liquid crystals and aquaporins also have exhibited high flux and selective water transportation [278–280]. Zeolite-polyamide thin film nanocomposite membranes offered new ways of designing NF and RO membranes with increased water permeability and high salt rejection [275, 281]. The use of nanozeolites in thin film nanocomposite membranes has resulted in enhanced permeability and salt rejection [282, 283].

By grafting functional groups, such as carboxyl, at opening of CNTs, membranes have better selective rejection of some components but this has resulted in reduced permeability rendering CNTs incapable for desalination [281, 284, 285]. Hinds concluded a uniform CNT diameter of less than 0.8 nm for high salt rejection [286]. 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 removal of heavy metals from water/wastewater such as nanosorbents including CNTs, zeolites, and dendrimers and they have exceptional adsorption properties [63]. The ability of CNTs to adsorb heavy metals is reviewed by many researchers [287] such as Cd^{2+} [288], Cr^{3+} [289], Pb^{2+} [290], and Zn^{2+} [291] and metalloids such as arsenic (As) compounds [292]. Composites of CNTs with Fe and cerium oxide (CeO_2) have also been reported to remove heavy metal ions in few studies [293–295]. Cerium oxide nanoparticles supported on CNTs are used effectively to adsorb arsenic [289]. Fast adsorption kinetics of CNTs is mainly due to the highly accessible adsorption sites and the short intraparticle diffusion distance [287].

Metal based nanomaterials proved to be better in removing heavy metals than activated carbon [296], for example, adsorption of arsenic by using TiO_2 nanoparticles and nanosized magnetite [297, 298]. The utilization of photocatalysts such as TiO_2 nanoparticles has been investigated in detail to reduce toxic metal ions in water [299]. In a study, the effectiveness of nanocrystalline TiO_2 in removing different forms of arsenic is elaborated and it has shown to be more effective photocatalyst than commercially available TiO_2 nanoparticles with a maximum removal efficiency of arsenic at about neutral pH value [300]. A nanocomposite of TiO_2 nanoparticles anchored on graphene sheet was also used to reduce Cr(VI) to Cr(III) in sunlight [301]. Similar Cr treatment was carried out by using palladium nanoparticles in another study [302].

The capability of removing heavy metals like As is also investigated by using iron oxide nanomaterials (Fe_2O_3 and Fe_3O_4) as cost-effective adsorbents by many researchers [303–305]. Arsenic removal was also investigated by using high specific surface area of Fe_3O_4 nanocrystals [306]. Polymer-grafted Fe_2O_3 nanocomposite was effectively used to remove divalent heavy metal ions for copper, nickel, and cobalt over a pH range of 3 to 7 [307].

Bisphosphonate-modified magnetite nanoparticles were also used to remove the radioactive metal toxins, uranium dioxide (UO_2^{2+}) with high efficiency from water [308]. Studies have shown that zero-valent iron or iron nanoparticles (nZVI or Fe^0) are very effective for the transformation of heavy metal ions such as As(III), As(V), Pb(II), Cu(II), Ni(II), and Cr(VI) [309–313]. Reduction of Cr(VI) to Cr(III) was also done by using nZVI and bimetallic nZVI nanoparticles in a study [314].

Novel self-assembled 3D flower-like iron oxide nanostructures were also used to successfully adsorb both As(V) and Cr(VI) [315]. The 3D nanostructures of CeO_2 are used as good adsorbents for both As and Cr [316]. The efficiency of NaP1 zeolites was evaluated for removal of heavy metals (Cr(III), Ni(II), Zn(II), Cu(II), and Cd(II)) from wastewater [317, 318]. Dendritic polymers were also used for treatment of toxic metal ions [319]. The applicability of self-assembled monolayers on mesoporous supports for removing toxic metal ions novel was also evaluated by many researchers [320–322]. Biopolymers have been used for heavy metal remediation from aqueous wastes [323, 324]. Chitosan nanoparticles for the sorption of Pb(II) were also used in one study [325].

NF is reviewed for the removal of cations and arsenic from ground/surface waters [326] and it has been shown very effective to remove uranium (VI) from sea water [327]. 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+} [85]. The incorporation of iron (hydr)oxide nanoparticles into porous carbon materials has made it possible to remove both inorganics and organics thus enabling such filters to be used as point-of-use applications [328, 329]. A dendrimer-UF system was used for the removal of Cu^{2+} and the complete removal from water was obtained [330]. Finally, there are commercial products for efficient removal of arsenic and these include iron oxide nanoparticles and polymers and nanocrystalline titanium dioxide medium in the form of beads [331, 332].

3.4. Removal of Organic Contaminants. NOM constitutes a diverse group of hydrophobic (humic and fulvic acids) and hydrophilic organic compounds and it contributes significantly towards water contamination [333–339]. A variety of carbon-based adsorbents have been used for the removal of NOM from raw water and several factors affect this sorption of NOM [340–343].

3.4.1. CNTs. Different types of nanomaterial like nanosorbents such as CNTs, polymeric materials (e.g., dendrimers), and zeolites have exceptional adsorption properties and are applied for removal of organics from water/wastewater [63]. CNTs have received special attention due to their exceptional water treatment capabilities and adsorption of organics on CNTs is widely studied and extensively reviewed [287]. The removal of NOM by CNTs is higher when compared with carbon-based adsorbents mainly due to large surface areas and other factors [344, 345]. CNTs are also effective to remove polycyclic aromatic organic compounds [346–348] and atrazine [345].

Nanoporous activated carbon fibers, prepared by electrospinning of CNTs, showed much higher organic sorption equilibrium constants for benzene, toluene, xylene, and ethylbenzene than granular activated carbon [349]. The sorption of 1,2-dichlorobenzene by CNTs has been shown as effective in one study [350]. Compared with activated carbon, both single- and multiwalled CNTs displayed higher adsorption capacities for trihalomethanes [293, 351]. Multiwalled CNTs have been used as sorbents for chlorophenols, herbicides, DDTs, and so forth [352–355]. Finally, novel polymers with functionalized CNTs showed effective removal of organic pollutants from water [295].

3.4.2. TiO_2 Nanoparticles. Metal oxide nanomaterials such as TiO_2 in addition to CeO_2 have also been used as catalysts for fast and comparatively complete degradation of organic pollutants in ozonation processes [356, 357]. Photocatalysts like TiO_2 nanoparticles are used effectively for the treatment of water contaminated with organic pollutants like polychlorinated biphenyls (PCBs), benzenes, and chlorinated alkanes [299]. Removal of total organic carbon from wastewater was enhanced by the addition of TiO_2 nanoparticles in a study [358]. TiO_2 nanoparticles were also used in a "falling film" reactor for the degradation of microcystins in water [159]. Multiwalled CNTs functionalized with Fe nanoparticles have been proved effective sorbents for aromatic compounds like benzene and toluene [359].

Decomposition of organic compounds can be enhanced by noble metal doping into TiO_2 due to enhanced hydroxyl radical production and so forth [360–362]. For example, the doping Si into TiO_2 nanoparticles was effective to improve its efficiency due to the increase in surface area and crystallinity [363, 364]. TiO_2 nanocrystals modified with noble metal deposits and so forth were used for the degradation of methylene blue in the visible-light range [198, 365, 366]. Nitrogen- and Fe(III-) doped TiO_2 nanoparticles were better in degrading azo dyes and phenol, respectively, than commercially available TiO_2 nanoparticles [367, 368]. TiO_2 nanoparticles deposited onto porous Al_2O_3 were used effectively for the removal of TOC in a study [369]. TiO_2 nanocomposites with mesoporous silica were also used for the treatment of aromatic pollutants [370–373]. A composite of nanosized SO_4^{2-}/TiO_2 was used for the degradation of 4-nitrophenol in one study [374]. TiO_2 nanotubes have been used effectively to degrade toluene better than bulk structural materials [375] and were found to be more efficient than TiO_2 nanoparticles for degrading organic compounds [376]. A commercial product (Purifics Photo-Cat system) has been shown highly efficient at removing organics [377–379].

3.4.3. Zero-Valent Iron. Nanocatalysts including semiconductor materials, zero-valence metal, and bimetallic nanoparticles have been used for degradation of environmental contaminants such as PCBs, pesticides, and azo dyes due to their higher surface area and shape dependent properties [380]. Magnetic nanosorbents also have proved effective in organic contaminants removal [381]. Iron oxide nanomaterials have shown better removal capabilities of organic pollutants than bulk materials [303, 382, 383]. Fe_2O_3 nanoparticles have also been used for the removal of colored humic acids from wastewater [384]. Chlorinated organic compounds and PCBs have been transformed successfully using nZVI [385–387] as well as inorganic ions such as nitrate and perchlorate [388, 389].

Particles of nZVI have been proved effective in degradation of toxic chlorinated organic compounds, for example, 2,2′-dichlorobiphenyl in a few remediation studies [390, 391]. The stabilized nZVI particles could also be an effective way for in situ remediation of groundwater or industrial effluents [392]. The nZVI and bimetallic nZVI have emerged as effective redox media for reducing a variety of organic pollutants such as PCBs, pesticides, organic dyes, chlorinated alkanes, and alkenes and inorganic anions (e.g., nitrates) in water/wastewater due to larger surface areas and reactivity [393, 394]. Bimetallic Ni^0/Fe^0 and Pd^0/Fe^0 nanoparticles were more effective than commercial microscale Fe for reductive dehalogenation of chlorinated organics and brominated methanes, hydrodechlorination of chlorinated aliphatics, chlorinated aromatics, and PCBs as reported by many researchers [395–402].

3.4.4. Other Nanomaterials. In a study, nanostructured ZnO semiconductor films were used for degradation of organic contaminants (4-chlorocatechol) [403]. The nanocatalyst of Ag and amidoxime fibers was used efficiently for degradation of organic dyes [404]. A bimetallic nanocomposite of Pd-Cu/γ-alumina was used for the reduction of nitrate in one study [405]. Traces of halogenated organic compounds were biodegraded using hydrogen and the Pd based nanoparticles [301, 406]. Pd nanoparticles and bimetallic Pd/Au (gold) nanoparticles were used effectively for hydrodechlorination of trichloroethylene (TCE) [407, 408]. Mineralization of organic dyes was accelerated in one study by using films of manganese oxide (MnO_2) nanoparticles and hydrogen peroxide [409]. Similarly, MnO_2 hierarchical hollow nanostructures were used for the removal of organic pollutant in waste water [410]. The immobilized nanoparticles of metallo-porphyrinogens have also been successfully used for the reductive dehalogenation of chlorinated organic compounds (TCE and carbon tetrachloride) [411]. The applicability of self-assembled monolayers on mesoporous supports for removing anions and radionuclides was also evaluated by many researchers [412, 413]. Molecularly imprinted nanospheres were used for the removal of micropollutants from hospital waste water [414]. Finally, single-enzyme nanoparticles could be used for decontaminating a broad range of organic contaminants as highlighted in one study [415].

3.4.5. Membranes. The immobilization of metallic nanoparticles in membrane has also been proved effective for degradation and dechlorination of toxic contaminants [416]. Inorganic membranes containing nano-TiO_2 or modified nano-TiO_2 have been used effectively for reductive degradation of contaminants, particularly chlorinated compounds [417, 418]. The use of TiO_2 immobilized on a polyethylene support and a TiO_2 slurry in combination with polymeric membranes has proved very effective in degrading 1,2-dichlorobenzene and pharmaceuticals, respectively [419, 420].

Polyethersulfone composite membranes with nano-TiO_2 as additive showed higher fluxes and enhanced antifouling properties [421–423]. Ceramic composite membrane made of TiO_2 nanoparticles inside a tubular Al_2O_3 substrate showed improved water quality and flux compared to Al_2O_3 membranes [424]. 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 [425]. Nanostructured composite of TiO_2 and Fe_2O_3 incorporated into ultrafiltration membranes successfully reduced the fouling burden and improved the permeate flux [369].

Cellulose acetate membrane laden with nZVI was found effective in dechlorination of TCE [426]. A reactive membrane of bimetallic nZVI and Pt^0 nanoparticles was found to be very effective at reducing TCE [427]. Bimetallic 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 [428–430]. Polyvinylidene fluoride film containing Pd and Pd/Fe was used effectively for dechlorination of PCB's in one study [431] leading to the development of a membrane reactor for dechlorination of a wider range of compounds [391, 432]. Alumina-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 [273, 433]. Finally, ceramic composite membranes of TiO_2 and CNTs have resulted in enhanced membrane permeability and photocatalytic activity [434–436].

NF is reviewed for the removal of NOM and nitrates from ground/surface waters [326] and it has been reported to improve the water quality with a substantial reduction in organic contaminants [437]. 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 towards three different synthetic dyes [84]. An improved antifouling performance and flux increase was also observed in silica-incorporated membranes [438].

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 [439, 440]. 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 [89]. Finally, the addition of metal oxide nanoparticles including silica, TiO_2, alumina, and zeolites to polymeric ultrafiltration membranes has helped reducing fouling [441–445].

4. Conclusions and Perspectives

Safe water has become a competitive resource in many parts of the world due to increasing population, prolonged droughts, climate change, and so forth. Nanomaterials have unique characteristics, for example, large surface areas, size, shape, and dimensions, that make them particularly attractive for water/wastewater treatment applications such as disinfection, adsorption, and membrane separations. The review of the 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 by coupling with a second catalytic metal can result in enhanced water/wastewater quality when applied for this purpose by increasing the selectivity and reactivity of the selected materials. Surface modification may lead to the enhanced photocatalytic activity of the selected compounds due to the short lifetime of reactive oxygen species and increase the affinity of modified nanomaterials towards many emerging water contaminants. Bimetallic nanoparticles have also proved effective for remediation of water contaminants. However, further studies are required for understanding the mechanism of degradation on bimetallic nanoparticles responsible for the improved efficiency. For real field applications, however, an improved understanding of the process mechanism is very important for the successful applications of innovative nanocomposites for water/wastewater treatment.

Electrospinning offers the way to modify the surface properties of nanomaterials and different nanofibrous filters have successfully been used as antifouling water filtration membranes. They have extremely high surface-to-volume ratio and porosity, are 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, their 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 water/wastewater treatment is very limited and a stand-alone system (Figure 3) is proposed for removing all types of contaminants including bacteria/viruses, heavy metals and ions, and complex organic compounds.

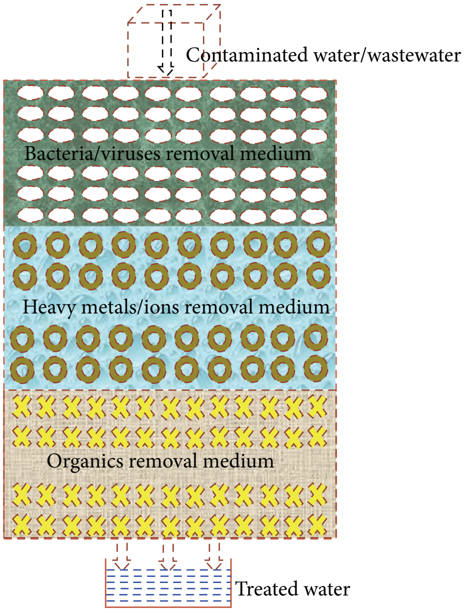


Figure 3: Schematic of a proposed composite nanofibrous media/membrane filters for complete removal of contaminants from water/wastewater.

The use of nanofibers and composite nanostructures membranes can help degrade a wide range of organic and inorganic contaminants in real field applications. The better understanding of the formation of nanocomposites membranes will certainly be a step towards improving the performance of multifunctional nanocomposites membranes. The pattern of nanoparticles within the host matrices of membranes and changes 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 water/wastewater treatment.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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