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REVIEW

Perspectives and applications of nanotechnology in water treatment

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Abstract Industrialization and excessive use of pesticides for boosting agricultural production have adversely affected the ecosystem, polluting natural water reserves. Remediation of contaminated water has been an area of concern with numerous techniques being applied to improve the quality of naturally available water to the level suitable for human consumption. Most of these methods, however, generate by-products that are sometimes toxic. Heterogenous photocatalysis using metal oxide nanostructures for water purification is an attractive option because no harmful by-products are created. A discussion on possible methods to engineer metal oxides for visible light photocatalysis is included to highlight the use of solar energy for water purification. Multifunctional photocatalytic membranes are considered advantageous over freely suspended nanoparticles due to the ease of its removal from the purified water. An overview of water remediation techniques is presented, highlighting innovations through nanotechnology for possible addressing of problems associated with current techniques.

Keywords Contamination · Water · Purification · Photocatalysis · Nanofiltration

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Introduction

The rapid pace of population growth has resulted in severe environmental contamination in air, water and soil. The world population is estimated to increase from the current figure of about 6.5 billion to an alarming 9 million by the middle of the twenty-first century. In order to boost agricultural production to cater to the huge population, farmers started relying on the extensive use of chemical pesticides, which has resulted in the contamination of groundwater (Sugunan et al. 2008; Baruah et al. 2009a, b, c, d, e). Water is an essential requirement for life, and its availability in pure form is important for different life-sustaining activities such as human consumption and agriculture, to name a few. Human activities have affected nature's very own water recycling and purification mechanism and have totally disturbed the balance between the consumption and natural purification processes resulting in a shortage of drinkable water. Almost all of the natural sources of drinking water such as surface water, groundwater, lakes and reservoirs, rivers and canals, and rainwater have been found to be contaminated with a wide variety of toxic materials and pathogenic microorganisms (Baruah et al. 2009a, b, c, d, e). Almost 12 million people die every year from water-related diseases, as per a World Health Organization (WHO) report (http://www.who.int/infectious-disease-report/pages/textonly.html). Impure water is the cause of about 90 % of all diseases occurring in developing. There are over 4 billion reported cases of diseases resulting from consumption of impure water globally. Pure low-carbon-content water through novel methods is of utmost necessity for the healthy existence of human being as well as the ecosystem.

Drinking water is currently being disinfected through the use of physical as well as chemical techniques.

Conventional water disinfection processes have certain restrictions resulting in apprehensions about their applications at a mass scale. Disinfection using UV light is effective against most of the harmful microorganisms but is incapable of inactivating certain disease-causing microbes like *Cryptosporidium* and *Giardia lamblia*. Chlorination is generally accepted as an effectual water disinfection technique as it is robust, cheap and has prolonged post-treatment outcome. However, chlorine readily reacts with natural organic materials present in water, thereby producing halogenated trihalomethanes (THMs) such as chloroform, bromoform, bromodichloromethane and haloacetic acids (HAA), which are carcinogenic in nature(Sadiq et al. 2004; Gopal et al. 2007; Sugunan et al. 2008; Baruah et al. 2009a, b, c, d, e). Ozonation is another alternative disinfection technique that effectively removes many of the disease-causing microbial contaminants from water but is not always viable as it requires on site ozone production and it is not always possible to guarantee water safety to the end users.

Heterogeneous photocatalysis shows promise as a water purification technique as compared to other conventional methods as this process does not generate harmful by-products (Sugunan et al. 2008; Baruah et al. 2009a, b, c, d, e). It can break up complex long-chained organic molecules, which are mostly toxic, into benign fragments as well as immobilize microbial cells by fracturing the cell walls. Nanotechnology is a disruptive technology that can make an impact in the area of water purification as nanostructures offer large surface to volume ratios ideal for surface reactions (Hornyak et al. 2008). The possibility of preparing photocatalytic membranes by growing semiconducting nanostructures on conventional membranes makes this technique even more attractive (Baruah et al. 2011; Baruah et al. 2008a, b). Replacement of fossil fuel with renewable energy like solar energy can present a cleaner and more efficient way of water purification, even in isolated rural sites. Point-of-use water purification systems can be designed using antimicrobial nanomaterials such as silver (Ag) and zinc oxide (ZnO) (Li et al. 2008). Membranes are increasingly being used in the fields of drinking water and wastewater treatment (Marcucci et al. 2003). Active functional membranes incorporated with antimicrobial or photocatalytic nanomaterials will be capable of accomplishing multiple treatment targets in a single course of action, at the same time minimizing fouling (Li et al. 2008). Loss of nanomaterials into water during treatment is a matter of concern for human health and the ecosystem, and therefore, proper attachment of nanomaterials to supports is of importance (Wiesner et al. 2006; Hirano 2009). This article is an abridged version of the chapter by Baruah et al. (2015) [Chapter 2: Nanotechnology in Water Treatment] published in the book series Environmental Chemistry for a Sustainable World (http://www.springer.com/series/11480).

Water remediation processes

Filtration

Filtration is the process of removal of solids from water by allowing the water to pass through a medium that blocks the particulate contaminants. The porous medium can be a porous physical barrier, a chemical or a biological process (Baker et al. 1981). This process is capable of removing mainly macroscopic particles. Microscopic particles and microbial specimens cannot be efficiently filtered out using standard filtration methods. However, innovative filtration technologies such as microfiltration, ultrafiltration and nanofiltration have emerged to handle these issues. In water treatment, filtration is also used to depict some biological processes in which undesirable constituents are removed by absorption into a biological film present in the filter medium.

Heavy metal adsorption

Industries are constantly dumping heavy metal ions into lakes, rivers and reservoirs, thereby polluting them. Heavy metals are broadly defined as materials whose density is above 5 g/cm^3 (Barakat 2011). Common heavy metals present in aqueous streams include chromium, mercury, lead and cadmium (Bailey et al. 1999). Table 1 (Kurniawan et al. 2006a, b) shown below describes the maximum contaminant level of heavy metals in surface water and their toxicities.

Heavy metal removal from water is crucial as these metals are non-biodegradable and can cause various health risks to both human and animal life (Babel et al. 2003; Argun et al. 2008). A variety of techniques can be applied to remove these metals from water which include chemical precipitation (usually used for inorganic effluents and not much effective for trace amount of solvents) (Bose et al. 2002; Wang et al. 2004), coagulation and flocculation (higher cost and lower efficiency) (Ayoub et al. 2001; Semerjian et al. 2003; Kurniawan et al. 2006a, b), reverse osmosis (effective but expensive) (Ozaki et al. 2002; Eddy 2004; Qdais et al. 2004), electrodialysis (effective for concentrated solution only) (Eddy 2004; Bhattacharyya et al. 2008), ion exchange (sophisticated and expensive) (Baes et al. 1996; Tiravanti et al. 1997; Bhattacharyya et al. 2008) and adsorption and filtration (efficient and cost effective) (Tran et al. 1999; Bhattacharyya et al. 2008). Heavy metal adsorption is a well-known process that utilizes mass transfer technique to remove adsorbates by

Table 1 Maximum contaminant level (MCL) of heavy metals in surface water and their toxicities

Heavy metals	Toxicities	Maximum effluent discharge standards (mg/L)		
		*EPA(2004) USA	**PCD(2004) (Thailand)	***EPD(2004) (Hong Kong)
Chromium (IV)	Headache, vomiting, diarrhea, nausea	0.05	0.25	0.05–0.10
Chromium (III)		0.10	0.75	
Zinc (II)	Lethargy, depression, neurologic signs	1.00	5.00	0.60–1.00
Copper (II)	Liver damage, insomnia, Wilson disease	0.25	2.00	0.05–0.1
Cadmium (II)	Renal disorder, kidney damage	0.01	0.03	0.001–0.05
Nickel (II)	Nausea, chronic asthma, coughing, dermatitis	0.20	1.00	0.10–0.20

* EPA (Environmental Protection Agency), USA

** PCD (Pollution Control Department), Thailand

*** EPD (Environmental Protection Department), Hong Kong

depositing them on the surface of adsorbent. It can be applied at lower concentrations for both continuous and batch operations. Ease of access and cost-effectiveness are other advantages of this technique (Mohanty et al. 2006; Bhattacharyya et al. 2008). Two kinds of forces may act during adsorption, namely physisorption and chemisorption (Rouquerol 1999). Physisorption is normally a weak force of attraction between molecules; it is non-specific, and molecules can move freely from one surface to another (Sawyer et al. 1994). This weak force of attraction can be dipole–dipole attraction and hydrogen bonding (Kelesoglu 2007). Chemisorption is based on very strong electrostatic forces, and a chemical bond forms between adsorbent and adsorbate, which is normally covalent or electrostatic bonding.

Many naturally occurring materials and industrial residue from different processes can be suitable adsorbents due to their cost-effectiveness (Bailey et al. 1999). A low-cost adsorbent is normally the material which is abundantly available, no further or very little purification is required or is a waste or by-product of some industrial process (Bailey et al. 1999). A lot of research is going on to study the properties of these adsorbents. Some common adsorbents used in water treatment to adsorb heavy metal ions are zeolites, alginates and activated charcoal.

Zeolites

Zeolites are naturally occurring materials which can also be produced synthetically (Ming et al. 1987). There are more than 40 natural zeolite-occurring species with clinoptilolite being the most abundantly available. Zeolite has three-dimensional crystal structure containing negative charge which is produced by replacement of Al^{3+} ions with Si^{4+} ions in a tetrahedron structure (Bailey et al. 1999). Enhanced adsorption capacities of zeolites are due to their higher ion-exchange capabilities. Zeolite structure consists of large channels and cavities where the ion exchange takes place and ion-exchange selectivity of zeolite results in charge separation. Figure 1 describes the selectivity of zeolite for both reactant and products depending on channel.

Potassium, sodium, calcium and other positively charged ions present in the channel are exchangeable and get replaced by heavy metal ions. Heavy metals present in wastewater (chromium, mercury, lead and cadmium) are effectively adsorbed on zeolites. Clinoptilolite is a widely used zeolite for wastewater treatment due to its higher selectivity and ion-exchange capability to remove heavy metal ions including strontium and cesium (Grant et al. 1987). Vaca Mier et al (2001) studied the selectivity of zeolite for the removal of various heavy metals and observed that zeolites show higher selectivity for lead ions followed by cadmium, copper and cobalt.

Alginates

Bioadsorbents are becoming increasingly popular for heavy metal removal from aqueous streams because of their effectiveness on treating dilute wastewater containing heavy metals. Presence of many functional sites, unlike ion-exchange resins (having only one functional site), and their relatively lower costs has led to widespread use of these materials (Wang et al. 2009; Fu et al. 2011). Biosorbents can be derived from many sources such as algal biomass, non-living sources including shrimp, squid and crab, and microbial mass such as yeast, fungi and bacteria (Apiratikul et al. 2008). Alginate is a polysaccharide that can be easily obtained from brown seaweeds. Alginates are biodegradable materials, and good biocompatibility makes them useful material for various applications (Gombotz et al. 2012; Pasparakis et al. 2006). Brown seaweed algae abundantly available in world has attracted the attention of many researchers due to its renewable natural biomass source and easy availability, low cost, higher metal affinity for adsorption and comparable

(a) Reactant Selectivity   **(b)** Product Selectivity
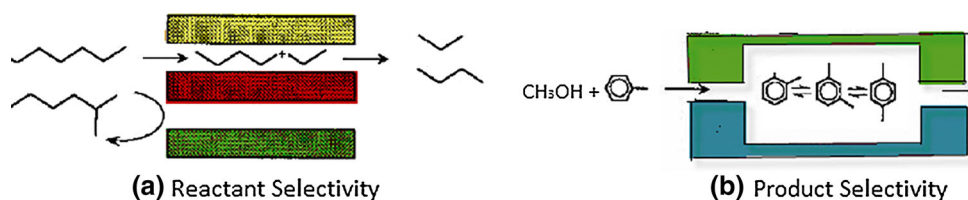
removal efficiency (Apiratikul et al. 2008; Fu et al. 2011). Vijaya et al. (2008) stated that at trivial conditions physiochemical properties, e.g., porosity and degradability of alginates, can be altered very easily, which can improve efficiency of heavy metal removal.

Alginates are biopolymers, and these have higher binding affinity for heavy metals, making them suitable material for higher metal uploads (Volesky 2003). Bailey et al. (1999) stated that alginates contain calcium ions which are replaced with heavy metals to form metal alginates by adsorption process. Cochrane et al. (2006) compared alginate (*Fucus vesiculosus*) and other commercially available adsorbents to study their selectivity and efficiency for copper removal from wastewater. Microalgae showed more than 95 % removal efficiency for copper which is comparable to other low-cost adsorbents and due to their low cast can prove to be a good alternative. Araujo et al. (1997) removed trivalent chromium using calcium alginate, where they studied the relationship between amount of chromium adsorbed and calcium replaced and the effect of initial concentration of alginate on adsorption processes. Park et al. (2004) used different types of alginates for removal of heavy metal from wastewater. Alginate gel, alginate beads and alginate capsules were used as adsorbent in the study. Alginate capsule showed higher adsorption of lead. Alginates capsule has high binding capacity to lead because of presence of xanthan gum in alginate solution. Aderhold et al. (1996) studied the ability of heavy metal removal of different alginates and also the effect of presence of more than one heavy metal on removal efficiency. Holan et al. (1993) stated that biosorbents can swell and disintegrate which can restrict their use for heavy metal removal. Thus, alginate composites can be a good solution. Alginate composite beads can be formed to remove heavy metals from wastewater streams. Composite beads of sodium and chitosan can be synthesized to increase adsorption capacity. Different composites remove different heavy metals depending upon their selectivity, and more work needs to be carried out to find a clear correlation (Ngah et al. 2008; Wan Ngah et al. 2011).

Activated charcoal/activated carbon

Water purification using charcoal is a very old practice. Use of charcoal to treat wastewater dates back centuries when charcoal was primary material to remove odor and taste from wastewater (Gupta et al. 2009). Water was kept in vessels open to sunlight and then filtered through charcoal. Modern use of charcoal is after oxidizing it, often called activated charcoal or activated carbon. Charcoal is oxidized at high temperature using different activation agents to increase surface area and porosity, which are essential for adsorption of heavy metals. Charcoal can be activated by both physiothermal and physiochemical methods. In physiothermal process, all the volatile matter removed by heating excessively at 500–600 °C and gasification is done at milder conditions to develop pores in the crystal structure. Activated carbon is a basic form of graphite and has an amorphous structure containing pores of various sizes (Mohan et al. 2006). Chemical activation is carried out by carbonization and metallic additives such as zinc chloride added prior to carbonization (Allen et al. 1998).

The major sources or raw material for activated carbon includes wood char, petroleum coke, sawdust, carbon black, peat and coconut shells (Pollard et al. 1992). Source of activated carbon plays a major role in its selection as adsorbent. High cost of activated carbon restricts its use for various environmental applications. Natural sourced activated carbons are low cost and widely used for heavy metal removal. Activated carbon can be classified into four types, i.e., granular, powdered, fiber and cloth activated carbon depending upon size and shape and type of raw material used (Kurniawan et al. 2006a, b). Activated carbon can be used for a variety of purposes and is a very good adsorbent material due to its porous structure for removing heavy metals from wastewater. Huang et al. (1977) and Di Natale et al. (2007) successfully removed chromium (IV) from water using activated carbon. Reaction mechanism involved reduction of chromium (IV) to chromium (III) and subsequent adsorption on activated carbon. pH of the water was noted to be one of the most important parameters for adsorption efficiency. Different research groups (Lee et al. 1995; Ranganathan 2000) have also used activated carbon to remove chromium from wastewater. The studies revealed that carbon could be reused after adsorption efficiently. Many researchers have reported the removal of Co(II), Cd(II), Ni(II), Pb(II), Cr(III), Cu(II) and Cr(VI) from wastewater using activated carbon with and without other chelating agents (Corapcioglu et al. 1987; Huang

et al. 1984; Chang et al. 1994; Anirudhan et al. 2011; Kobya et al. 2005). Paajanen et al. (1997) removed heavy metals using activated carbon synthesized from peat, coconut shell and coal. Uzun et al. (2000) compared efficiency of activated carbon with other low-cost adsorbent such as agar and chitosan for heavy metal removal from wastewater. Authors concluded that one material could be a good adsorbent for certain metals but it may not be suitable for another one. All adsorbent showed comparable efficiency, but order of selectivity for different materials was different depending on surface properties, solution pH and many other factors. Kurniawan et al. (2006a, b) observed that agriculture-based activated carbons after treatment are most effective for removal of heavy metals, i.e., hazelnut shell activated carbon, orange peel and citric acid-modified activated carbons have the highest removal capacity for heavy metals such Cr(IV), Ni and Cu(II) as compared to activated carbon synthesized from coal, calcined phosphate and other synthetic materials (Marshall et al. 1999; Ajmal et al. 2000; Kobya 2004).

It can be concluded that among all three types of adsorbents discussed above agriculture-based activated carbon after heat treatment has shown outstanding adsorption properties as compared to other low-cost adsorbents such as zeolites and alginates. It also should be noted that effectiveness of adsorbents also depends upon local conditions, e.g., countries with less agriculture cannot use agriculture-based adsorbent due to higher cost of logistics (Kurniawan et al. 2006a, b).

Disinfection

Microorganism present in water and wastewater can cause a variety of diseases to humans. Microorganisms that are responsible for diseases are known as pathogens. Various water treatment technologies are available to inactivate these pathogens. Inactivation of pathogen is usually called as disinfection of water (Sobsey 1989). Disinfection is the term used for removal of only pathogens, and it does not remove all the microorganisms present in water as few useful microorganisms also present in water (Ellis 1991). Different techniques are applied for disinfection including chlorination, ultraviolet light treatment and ozonation.

Chlorination

Chlorination is widely used method for water treatment for centuries. Initially chlorine was used for odor removal, but in late nineteenth century chlorination progressed as an effective disinfection technique (Tzanavaras et al. 2007). Chlorination is effective against bacteria and viruses, but it is not effective against protozoan cysts which restrict its use for some applications (Burch et al. 1998). Mechanism for chlorination involves damage to cell wall of microorganisms, where chlorine penetrates into its cell to unsettle respiration and DNA activity. Chlorination is normally carried out by liquefied chlorine gas, chlorine dioxide, calcium hypochlorite particles, sodium hypochlorite solution, etc. (2004). Use of chlorine gas can cause organic matters such as fulvic acids prevailing in water to form halogenated hydrocarbons or TMH which are health hazards if consumed (Lykins et al. 1986; Huang et al. 1997). Chlorine dioxide has better disinfection properties than chlorine because less organoleptic interference is produced. Chlorine dioxide (ClO_2) gas is neutral intricate of chlorine gas with IV^+ oxidation state. ClO_2 is a highly volatile compound and in aqueous solutions found as free radical (Tzanavaras et al. 2007). ClO_2 is soluble in water at very low temperatures, and due to its one electron transfer mechanism, it is one of the most versatile compound for disinfection (Hoehn et al. 1996). ClO_2 can be easily removed from water by deaeration. Huang et al. (1997) compared ClO_2 and Cl_2 for disinfection of bacteria in water and found that ClO_2 is better disinfectant than its counterpart chlorine gases. Disinfection by ClO_2 depends upon pH of solution, disinfectant loading and contact time.

Ultraviolet (UV) light treatment

Since the inception of UV treatment of water in the 1970s, (Bukhari et al. 1999), it has been widely used for microorganism reduction as it produces no harmful by-products (Hijnen et al. 2006). The UV treatment technique involves a low-pressure UV lamp at a wavelength from 200 to 300 nm (Zhou et al. 2002). UV lamps do not affect the biological stability of water as it happens with chemical treatment. UV dose is very important parameter for the efficacy of this process. UV dose is defined as the rate of total incident radiation per unit area from all the directions and at all wavelengths and the exposure time (Bolton 1999; Zhou et al. 2002; Najam Khan et al. 2015).

UV light treatment utilizes physical mechanism instead of addition of any chemicals used by other techniques. UV light penetrates the structure of microorganism by absorption. Dosage of UV light is very important, and at higher dosages of UV light, proteins absorb the light and damage the cell wall, leading to death of the cell. At lower concentration of UV light, it is absorbed by DNA and RNA to inactivate the cell (Zhou et al. 2002). Rate of inactivation depends upon amount of light absorbed by microorganisms, i.e., UV dosage which is the intensity of UV light and time of exposure (Gyürék et al. 1999). Another important parameter for UV treatment performance is the surface area of microorganisms and their distribution (Loge et al. 1999). The inactivation by UV varies considerably for different organisms (Karanis et al. 1992). The efficiency of

UV treatment system thus strongly depends upon type of water being used (Zhou et al. 2002; Mamane et al. 2010)

Ozonation

Ozone is a colorless and very unstable gas that consists of three oxygen atoms. Ozone is readily converted back to oxygen by forming one free oxygen atom or radical during transition stage. The free radical of oxygen is short-lived and very reactive. At ambient conditions, free radical of oxygen survives only for a few milliseconds. Due to its high oxidation potential, ozone is a good candidate for disinfection of water streams. Ever since the use of ozone for water disinfection in 1886 by De Meritens, considerable research has taken place especially in recent times for replacing chlorine with ozone as it does not form TMH and organochlorine during the disinfection process (Camel et al. 1998). Ozone when added in wastewater rapidly is consumed because organic matter and inorganic salts require it. After the initial ozone demand is satisfied, it leads to disinfection much faster than what is achieved with chlorination.

Ozone reacts directly with organic matter as oxidizing agent and also decomposes instantly to form a complex mechanism which produces free hydroxy radicals during the process further enhancing the disinfection process (Hoigné et al. 1976). The efficiency of ozonation process strongly depends upon type of water being disinfected. Ozone due to its highly unstable nature produces very low amount of by-products (Glaze et al. 1988). A major disadvantage of ozonation is the fact that ozone needs to be generated at the point of use because it cannot be transported due to its highly unstable nature (1999)

Figure 2 shows two pathways of ozonation of water competing for oxidation, direct oxidation is slower than hydroxy radical mechanism but concentration of aqueous ozone is higher as compared to hydroxy radical where ozone concentration is less in indirect oxidation process (1999).

DeMers et al. (1992) observed that ozonation is better than chlorination, but due to its inability to sustain left over in distribution system it should be used with chlorination or other disinfectants for thorough disinfection. Reaction

parameters such as pH, temperature and organic matter content can play a major role in increasing the efficiency of the process. Reports available in the literature indicate that pH and temperature do not affect the reaction efficiency (Farooq et al. 1977; Katz 1980; Kinman 1975). Ozonation is widely used for inactivation of bacteria (Domingue 1988), removing protozoan cysts (Domingue 1988), and virus inactivation (Bablon 1991). All the disinfection techniques stated above have certain advantages and disadvantages, but UV treatment and Ozonation have greater potential for being used as viable disinfection processes to replace chlorination. Chlorine gas is very effective disinfectant, but due to production of harmful by-products, its use as disinfectant is diminishing very fast.

Harmful effects of water remediation schemes

Disinfection by-products (DBPs) are generally formed when disinfectants, during water purification, react with naturally occurring organic matter and also anthropogenic contaminants such as bromides and iodides (Hebert et al. 2010) Chlorination is a well-accepted method for water treatment, but it has its share of disadvantages. Excessive amounts of chlorine can be toxic for human beings and can be a cause of irritation to the eyes, the nasal passage and respiratory system (Medina-Ramon et al. 2005). Use of chlorinated drinking water can be related to the risk of cancer, especially bladder and colorectal cancers, which can be attributed to the presence of THM (Cantor et al. 1998; Hildesheim et al. 1998; Cantor et al. 1999). Chlorination is not a useful disinfectant for the removal of protozoan cysts (http://www.inchem.org/documents/ehc/ehc/ehc216.htm#SectionNumber:1.3). The remarkable biocidal properties of chlorine are negated due to the formation of DBPs during the chlorination process, which are detrimental to human health. A drawback of chemical treatment, especially for drinking water, is the bad taste of the water. Further, the process is potentially harmful for people with thyroid disease or iodine allergy (http://www.who.int/water_sanitation_health/dwq/gdwq0506.pdf).

A report released by the national environmental health group Women's Voices for the Earth (WVE) (http://www.womensvoices.org/wp-content/uploads/2010/05/Disinfectant-Overkill.

Fig. 2 Oxidation reactions of ozone during water disinfection (Hoigné et al. 1976)
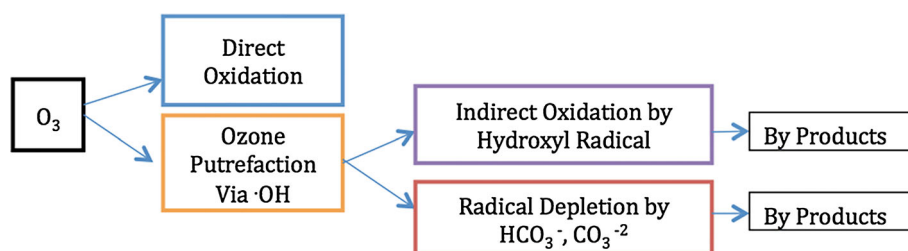
pdf) linked disinfectant chemicals with chronic illnesses and conditions such as asthma, hormone imbalance, and immune system problems in a report titled "Disinfectant Overkill: How Too Clean May Be Hazardous to Our Health," which cited more than 40 peer-reviewed reports and scientific studies. According to the report Triclosan and Triclocarban, two antibacterial disinfectants may have hormone-disrupting effects: Triclosan adversely affects communication between cells in the brain and the heart, while Triclocarban appears to amplify testosterone in the body (http://www.womensvoices.org/wp-content/uploads/2010/05/Disinfectant-Overkill.pdf). The physical and chemical properties of disinfectants and their by-products can influence their behavior in drinking water.

Ozone can react with bromide to form brominated ozone DBPs like bromate ion (BrO_3^-). In the presence of natural organic matter, ozonation leads to the formation of non-halogenated organic DBPs such as carboxylic acids, aldehydes and ketoacids. In the presence of both natural organic matter and bromide, ozonation forms hypobromous acid and this can form brominated organohalogen compounds. Halobenzoquinones (HBQs) have recently been considered disinfection by-products (DBPs) of toxicological relevance as it can be the cause of bladder cancer (Zhao et al. 2012) The problem with ultraviolet treatment is that even though it is capable of immobilizing a wide variety of disease-causing bacteria, the effect is temporary. UV-treated water should not be stored for long as the bacteria again revives after the UV source is removed (http://www.drinking-water.org/html/en/Treatment/Chemical-Disinfection-Oxidants-technologies.html). The flow rate of water is crucial in a UV treatment process as a high flow rate may lead to insufficient UV exposure and a slow rate may lead to heat buildup and subsequent damage to the bulb (Gadgil 1997)

Nanotechnology in water purification

Nanotechnology, which relates to materials and devices with physical dimensions comparable to or less than 100 nm, shows immense promise as a viable means of treating both persistent and emerging water contaminants (Brame et al. 2011; Baruah et al. 2009a, b, c, d, e). This emerging technology is capable of positively affecting technologies such as desalination of seawater to increase freshwater supply. Engineered nanomaterials could also have adverse effects on the ecosystem by contributing to water contamination (Baruah et al. 2009a, b, c, d, e; Najam Khan et al. 2014). Here, we discuss both the positive properties as well as implications of nanomaterials in water treatment.
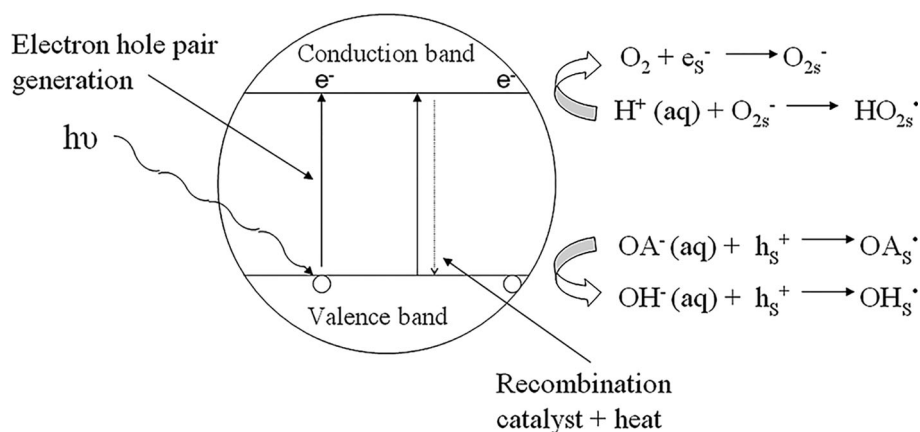
Photocatalysis

The Report from the Workshop on Nanotechnologies for Environmental Remediation[4] identifies solar photocatalysis as the main technology breakthrough for water treatment and purification, particularly in developing regions. Initial pilot projects are now being carried out. Photocatalytic systems may also complement existing techniques in the removal of trace contaminants. Such systems are commercially available, e.g., for the disinfection of swimming pools.

Photocatalysis, using nanostructures of metal oxide semiconductors such as zinc oxide (ZnO), titania (TiO_2), tungsten oxide (WO_3) and zinc stannate (Zn_2SnO_4) can be an attractive way of water purification as it is capable of removing chemical as well as biological contaminants (Baruah et al. 2012; Baruah et al. 2009a, b, c, d, e). A good photocatalyst should absorb light efficiently preferably in the visible or near UV part of the electromagnetic spectrum. Sufficient electron vacant states need to be present to inhibit recombination of electron hole pairs upon light exposure. As a lot of work is going on using photocatalysis in the agriculture and microbiology fields, it is important that the photocatalysts should be biologically inert and non-toxic. Nanostructured photocatalysts offer large surface to volume ratios allowing higher adsorption of the target molecules. Intensive research over the past decade for its implementation in the purification of drinking water can be found in the literature (Mahmood et al. 2011; Makhal et al. 2010; Sapkota et al. 2011; Fujishima et al. 2000; Bianco-Prevot et al. 2001; Cho et al. 2004; Aguedach et al. 2005; Chatterjee et al. 2005; Evgenidou et al. 2005; Adams et al. 2006; Chen et al. 2006; Benabbou et al. 2007; Baruah et al. 2008a, b; Baruah et al. 2009a, b, c, d, e; Baruah et al. 2010a, b). Efficacy of photocatalysis in the detoxification of a wide range of industrial and agricultural effluents is also well documented (Gaya et al. 2008). Another interesting aspect of photocatalysis is the potential utilization of sunlight, which could allow energy-efficient treatment in remote locations.

The underlying mechanism of heterogeneous photocatalysis is schematically represented in Fig. 3. It involves a wide band gap semiconductor photocatalyst, which upon irradiation with light of energy higher than the band gap energy of the material, electron–hole pairs (excitons) are created. The photogenerated electron moves up to the conduction band, while the hole drifts to the bottom of the valence band. Majority of these photogenerated charge carriers undergo wasteful recombination, while escape recombination and initiate redox reactions in molecules adsorbed at the surface of the photocatalyst and thereby degrading them. The photogenerated electrons and holes have been found to degrade almost all types of organic,

Fig. 3 Schematic diagram explaining photocatalysis on semiconducting surface (Baruah et al. 2009a, b, c, d, 2009e)
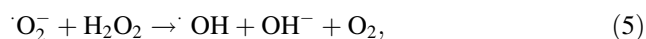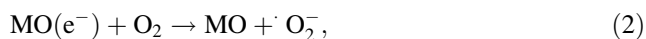

inorganic and microbial contaminants (Gaya et al. 2008), owing to their high redox potentials.

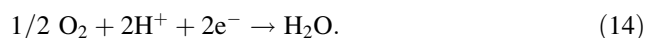The fundamental process during photocatalysis is given by

$$MO + h\nu \rightarrow MO(e^- + h^+) \tag{1}$$

where MO represents a metal oxide photocatalyst such as TiO_2 and ZnO. Photogenerated electrons lead to the formation of superoxide anions ($^.O_2^-$), hydrogen peroxide molecules (H_2O_2), hydroxyl radicals ($^.OH$), hydrogen dioxide anion (HO_2^-) and hydroperoxy radicals ($^.HO_2$) (Banerjee et al. 2006; Baruah et al. 2009a, b, c, d, e).

$$MO(e^-) + O_2 \rightarrow MO + ^.O_2^-, \tag{2}$$

$$MO(e^-) + ^.O_2^- + 2H^+ \rightarrow MO + H_2O_2, \tag{3}$$

$$MO(e^-) + H_2O_2 \rightarrow MO + ^.OH + OH^-, \tag{4}$$

$$^.O_2^- + H_2O_2 \rightarrow ^.OH + OH^- + O_2, \tag{5}$$

$$^.O_2^- + H^+ \rightarrow ^.OH_2, \tag{6}$$

$$MO(e^-) + ^.OH_2 \rightarrow MO + HO_2^-, \tag{7}$$

$$HO_2^- + H^+ \rightarrow H_2O_2, \tag{8}$$

$$2^.OH_2 \rightarrow O_2 + H_2O_2. \tag{9}$$

While the oxidation reactions initiated by the photogenerated holes are:

$$MO(h^+) + H_2O \rightarrow MO + ^.OH + H^+, \tag{10}$$

$$MO(h^+) + H_2O \rightarrow MO + ^.OH + H^+, \tag{11}$$

$$MO(h^+) + OH^- \rightarrow MO + ^.OH. \tag{12}$$

The reactions are terminated as:

$$^.OH + H^+ + 2e^- \rightarrow H_2O, \tag{13}$$

$$1/2\ O_2 + 2H^+ + 2e^- \rightarrow H_2O. \tag{14}$$

Nanostructures consist of a large number of low-coordination-number atoms at edge and corner sites of the crystal lattice providing numerous catalytically active sites.

A lot of work is going on to remove the harmful effects of chemical contaminants from groundwater mainly through photocatalysis using nanoparticles of metal oxides such as TiO_2 and ZnO (Herrmann et al. 2000; Bianco-Prevot et al. 2001). Another nanostructured semiconductor that is receiving attention from researchers as a stable photocatalyst is the ternary oxide zinc stannate (Baruah et al.; Foletto et al. 2010; Huang et al. 2012; Tian et al. 2012; Kurz et al. 2006; Lou et al. 2006; Lou et al. 2006). Rahman et al. (2005) studied the degradation kinetics of two pesticides dichlorvos and phosphamidon using Degussa 25 (commercially available TiO_2 nanoparticles) and observed that the addition of electron acceptors such as hydrogen peroxide (H_2O_2) enhances the degradation rates of the pollutants. Solar photocatalysis was also successfully used for degrading aldrin with three transformation products, dieldrin, chlordane and 1,2-hydroxy dieldrin (Bandala et al. 2002). Dichlorvos, which has been classed as a Restricted Use Pesticide (RUP) due to its toxicity, has been successfully degraded using photocatalysts such as TiO_2 and ZnO (Evgenidou et al. 2005). It was observed that the addition of electron acceptors such as hydrogen peroxide (H_2O_2) or potassium persulphate ($K_2S_2O_8$) increases the degradation rate in the presence of TiO_2 but retards it in the presence of ZnO. Mahalakshmi et al. (2007) has used total organic carbon (TOC) analyzer to confirm the complete mineralization of carbofuran which is an extremely toxic carbamate pesticide that is extensively used as an insecticide in a wide variety of field crops, including soybean and potatoes. Water-soluble pesticides have also been mineralized at the pilot plant scale using two well-defined systems exploiting solar UV light for heterogenous photocatalysis using TiO_2 nanoparticles (Oller et al. 2006). Nearly 100 % mineralization was obtained with total removal of pesticides such as cymoxanil, dimethoate, methomyl, oxamyl, pyrimethanil and telone (Oller et al. 2006).

Water purification agents should be capable of removing not only chemical, but also microbial contaminants such as

bacteria, fungi, virus and molds. Photocatalytic inactivation of microorganisms is a complex process, and the rate of inactivation varies with the type, concentration and the physiological state of the microbes (Rincon et al. 2004; Lonnen et al. 2005). The nature, morphology, concentration and state (slurry or immobilized) of the catalyst material also have a great influence on the microbial inactivation rates (Huang et al. 1998; Qi et al. 2004; Sondi et al. 2004; Adams et al. 2006). Among the various bacterial species, *Escherichia coli* (*E. coli*) which causes diarrhea has been extensively tested to optimize photocatalytic processes as well as for testing newly designed photoreactors (Krishna et al. 2008). Apart from *E. coli* in pure water, the photocatalytic inactivation of other *coliform bacteria* has also been reported in the literature (Gelover et al. 2006). TiO$_2$ nanoparticles (Degussa P25) have been used to successfully inactivate different genera of bacteria including *Escherichia coli*, *Pseudomonas aeruginosa*, *Salmonella typhimurium* and *Enterobacter cloacae* (Ibanez et al. 2003). Reports of photocatalytic inactivation of model microbes such as *Escherichia coli*, *Staphylococcus aureus*, *Saccharomyces cerevisiae* and *Aspergillus niger* spores have been reported for palladium (Pd) doped TiO$_2$ and tin dioxide (SnO$_2$) films grown on glass substrates (Erkan et al. 2006). The use of TiO$_2$ nanoparticles (Degussa P25) to inactivate bacteria (*E. coli*, *Pseudomonas aeruginosa*), fungi (*Candida albicans*, *Fusarium sloani*), protozoa (the trophozoite stage of *Acanthamoebe polyphaga*), spores (*Bacillus subtilis*) and cysts under solar light irradiation are also available in the literature (Lonnen et al. 2005).

Nanofiltration

Nanofiltration, a relatively new entrant in the group of membrane filtration processes, used mostly with surface water and fresh groundwater having fewer amounts of dissolved solids. The objectives of nanofiltration are the softening (polyvalent cation removal) of water and removal of DOB precursors such as natural and synthetic organic matter (Letterman 1999; Hillie et al. 2007). The type of materials that can be filtered out depends upon the pore sizes of the filtration membranes. Nanofiltration, which is a cross-flow filtration technology, can be placed in between ultrafiltration and reverse osmosis. The pore size of the nanofiltration membrane can go down to about 1 nm. Figure 4 shows some contaminants that can be removed using membranes with different pore sizes ranging from 0.5 to 1000 nm (http://www.techneau.org/fileadmin/files/Publications/Publications/Deliverables/D5.3.4b.pdf). Figure 5 shows a schematic representation of water softening using nanofiltration membrane where the water is forced through the membrane using high pressure.
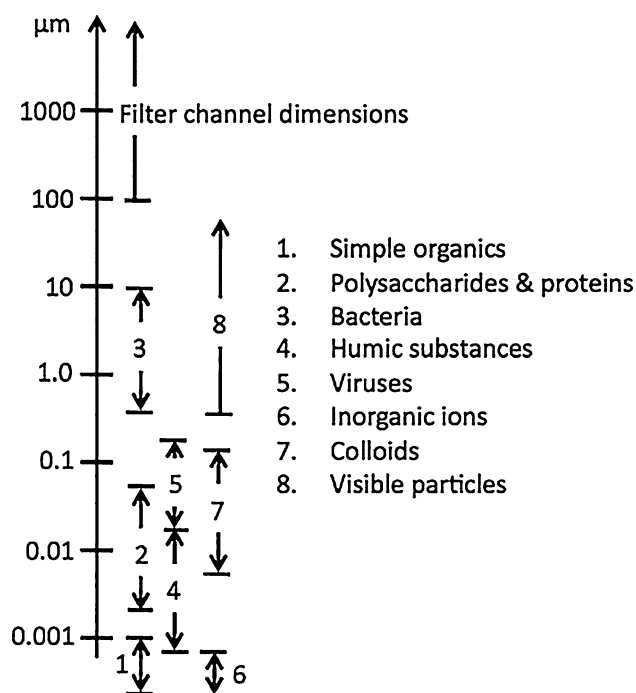

Fig. 4 An overview of membrane filtration. The pores sizes of the filtration membranes lie in the range between 0.5 and 1000 nm

The selectivity of a nanofiltration membrane is governed by two different parameters: retention and permeability. The retention is a function of the solute size. In nanofiltration membranes, retention and permeability are also a function of electric charge and the valency of the salts and compounds in the solution (http://www.fumatech.com/EN/Membrane-technology/Membrane-processes/Nanofiltration/). Monovalent ions of mild concentrations can mostly
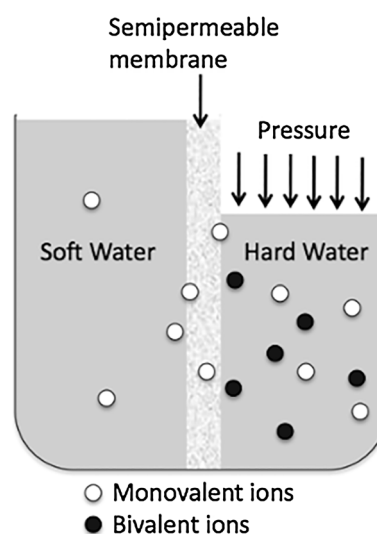

Fig. 5 Schematic representation of water softening using nanofiltration membrane

Table 2 Properties of different cross-flow filtration techniques

Method	Pore size (nm)	Molecular weight	Pressure (bar)	Permeation
Reverse osmosis	<0.6	<500	30–70	Water
Microfiltration	50–5000	>500 kDa	0.5–2	Water + low molecular solutes + macromolecules + colloids
Ultrafiltration	5–50	2–500 kDa	0.5–10	Water + low molecular solutes + macromolecules
Nanofiltration	0.6–5	500–2000 kDa	10–40	Water + low molecular solutes

pass through a nanofiltration membrane unobstructed, while most of the multivalent ions such as sulfates and carbonates are blocked. Cation retention using a nanofiltration is minimum for protons, and for other commonly present cations, retention increases following an order as sodium, potassium, calcium, magnesium, copper and iron. As for anions, the retention capability of nanofiltration membranes increases in the following order: nitrates, chlorides, hydroxides, sulfates, carbonates and phosphates (http://www.fumatech.com/EN/Membrane-technology/Membrane-processes/Nanofiltration/).

The membrane pore size is a major factor that determines whether a particular solute will pass through the membrane or not. Nanofiltration is a type of cross-flow filtration where the majority of the feed flow travels tangentially across the surface of the filter media, rather than through it (Koros et al. 1996). An organization of cross-flow filtration techniques is given in Table 2, which shows that membranes are available with pore sizes in the range of about 0.5 nm to about 5 μm. Dalton is a general unit of molecular weight in membrane filtration and expressed in g/mole (http://www.techneau.org/fileadmin/files/Publications/Publications/Deliverables/D5.3.4b.pdf).

From Table 2, it is obvious that the nanofiltration process is capable of removing almost all types of solutes from natural surface waters. However, if the source water is seawater, brackish water or groundwater, reverse osmosis is a better option. Even then, nanofiltration techniques are widely used for softening and natural organic matter may be a matter of apprehension here. Natural organic matter can add undesirable color to the water and also foul the membranes. Nanofiltration and reverse osmosis techniques are efficient in softening hard water for which high operating pressures are needed. To address scaling and fouling issues, modifiers such as antiscalants become necessary (http://www.techneau.org/fileadmin/files/Publications/Publications/Deliverables/D5.3.4b.pdf).

Future prospects

Nanotechnology can usher in a revolution in the domains of water treatment and distributed water reuse. It is capable of precluding concerns related to the formation of harmful disinfection by-products associated with conventional water treatment methods. Nanomaterials are endowed with unique properties like high surface to volume ratios, enhanced surface-related activities like catalysis and antimicrobial properties, property of self-assembling on substrates to form films, high conductivity that can be effectively used in capacitive deionization method for desalination, and high fluorescence for detection. Nanomaterials can be engineered to effectively act as a visible light photocatalyst so that water purification can be carried out using solar energy, which is available freely. Another area of concern that can possibly be addressed by nanotechnology is the degradation of the quality of water as it moves through distribution networks. Using nanotechnology, point-of-use water purification systems utilizing solar energy could be designed, which can be ideal for disaster-prone areas (Baruah et al. 2012). Nanotechnology is capable of exploiting alternative water sources for drinking and agriculture, keeping energy consumption to a bare minimum. Nanotechnology can especially impact the developing countries, which are more prone to degradation of water quality. High-performance innovative water treatment technologies have now become a necessity. Future water treatment systems in developing countries will most likely opt for nanotechnology-based water monitoring, treatment and reuse systems that can efficiently immobilize a wide variety of water pollutants coupled with affordability and ease of operation.

Conclusions

Nanotechnology is likely to make a tremendous impact in the area of drinking and wastewater purification and reuse. Nanomaterials possess unique properties as compared to their bulk counterparts such as increased surface area to volume ratios, higher surface reactivity and band tunable semiconductivity, to mention a few. Wideband semiconducting nanostructures can be used to degrade harmful contaminants and microbes through photocatalysis using solar radiation. Nanofiltration, using membranes with pore sizes in the nanometer regime, can successfully convert hard water into soft water by blocking mono- and bivalent ions as the water passes through the semipermeable membranes. Nanotechnology can potentially improve all

the current disinfection, purification and desalination techniques and usher in an era of point-of-use water purification systems harnessing solar energy.

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