

CHAPTER 1

Decontamination Using Nanotechnology

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1.1 Introduction

Nanotechnology can be defined as the synthesis, characterization, and application of materials science and engineering and devices whose smallest functional organization in at least one dimension is on the nanometer scale (one billionth of a meter).^{1,2} The nanotechnology has grown to multidisciplinary scientific fields in a decade. Nanotechnology has shown huge potential in areas as diverse as drug development, water decontamination, information and communication technologies, and the production of stronger, lighter materials and human health care.^{3,4} Water and air are two vital components of life on Earth; the existence of life on Earth is made possible largely because is owed by the presence of clean water and air in habitual form because of their importance to metabolic processes within the body. Clean and fresh water and air are essential for the very existence of life. Contamination of natural water sources by pathogenic microorganisms, heavy metal and groundwater with organic pollutants are worldwide public health problems, leading to waterborne outbreaks of infectious hepatitis, viral gastroenteritis, and cancer. Other contaminants of water are microorganisms such as *Escherichia coli* (*E. coli*), amoebas, *Cryptosporidium*, cholera, viruses, and bacteria.

Long-term drinking of water containing high levels of heavy metals can cause serious disorders, such as anemia, cancer, kidney disease, and mental retardation.⁵ Decontamination is the reduction or removal of chemical and biological agents by means of physical,

chemical neutralization, or detoxification techniques.⁶ The recent development of nanotechnology has raised the possibility of environmental decontamination through several nanomaterials, processes, and tools.⁷ To keep pace with this extremely rapid growth in nanotechnology in the field of remediation, it is necessary to critically assess the current knowledge of environmental decontamination where prominent scientists and researchers have provided insight in their areas of expertise, thus offering an overall picture of state-of-the art of the field.⁸ This chapter summarizes the expertise of the bottom-up approach of decontamination for the successful realization of remediation in drinking water from microbes, heavy metals, industrial toxic chemicals, chemical warfare agents, microbes, and biowarfare agents through nanotechnology.⁹⁻¹² A recent study on nanotechnologies in water, drinking water, and wastewater worldwide for 2007 was projected to be US\$1.6 billion and US\$6.6 billion for 2015.¹³

1.2 Drinking Water Decontamination

The presence of natural organic materials, heavy metals as well as industrial pollutants, could cause pollutants to infiltrate drinking water.¹⁴ Drinking water should be free from metals (Cd, Zn, Cu, Pb, Zn, As, Al, Be, and Ag), nutrients (PO_4^- , NH_3 , NO_3^- , NO_2^- , TP, and TN), algae (cynobacterial toxins), and biological agents (*E. coli*, viruses, bacteria, parasites), and organics (e.g., antibiotics, chloroacetic). Chlorine, ozone, chlorine dioxide, and chloramine are the most common disinfectants in use today, and each produces its own suite of chemical disinfection by-products (DBPs) in drinking water. Although the major concern has been focused in chlorinated DBPs, brominated DBPs have been considered to be even more hazardous than their chlorinated counterparts. Formation of iodinated- and nitrogen-containing DBPs have been studied more intensively.¹⁵ In fact, chlorine can react with organic material to produce chlorinated hydrocarbons. Ozone, among the strongest oxidizers known, is commonly used worldwide for disinfection and decontamination.

Several important processes that have been adopted for purification of drinking water, including UV photolysis, ozone-based applications: $\text{O}_3/\text{H}_2\text{O}_2$, O_3/UV , $\text{O}_3/\text{H}_2\text{O}_2/\text{UV}$, and $\text{O}_3/\text{H}_2\text{O}_2/\text{TiO}_2$; heterogeneous photocatalysis: TiO_2/UV , catalytic oxidation where aqueous H_2O_2 decomposes over heterogeneous catalyst including metals (e.g., Fe, Cu, Pt, Ti, and Ni) and metal oxides immobilized on various support materials such as sand, silica, zeolites, and alumina; electrochemical oxidation and ultrasound irradiation.¹⁶⁻³⁰ We know that the germicidal effect of UV light on microorganisms is primarily due to the dimerization of adjacent thymine bases in the DNA molecule through covalent bonding. In addition, photocatalytic

inactivation using TiO_2 has the ability to destroy pathogens. When a specific photocatalyst, such as titanium dioxide (TiO_2), having a band-gap energy of 3.2 eV, is illuminated with near-UV light with a wavelength less than 385 nm, photons are absorbed and electron-hole pairs are produced in this material. The holes are very oxidizing and can be used to destroy organisms.

Reverse osmosis (RO) is commonly used to purify water and remove salts and other impurities in order to improve water's color, taste, or other properties. RO uses a semipermeable water purifier membrane, allowing the highly purified water to pass through it while leaving certain unwanted or harmful substances behind. These substances are rinsed away leaving pure, clear, fresh-tasting water. RO units with carbon filters may also reduce the level of some soluble organic compounds (SOCs) such as pesticides, dioxins and volatile organic compounds (VOCs) (such as chloroform and petrochemicals). It is necessary to establish feedwater quality guidelines to optimize system performance and prevent the three main problems associated with RO: scaling, fouling, and degradation of RO membranes. These problems tend to decrease system productivity because they reduce the purity of wastewater. In fact, the low solubility products such as calcium carbonate, barium sulphate, reacted silica are difficult to remove from RO membranes. Besides, the RO filter has been used to perform desalination. It is well known that the separation of hydrocarbons and the removal from various liquid mixtures are important objectives of chemical and petrochemical industries. Interestingly, the metal-ligand-coordinated multilayer membrane showed a certain separation capability for aromatic-aliphatic hydrocarbons.³¹⁻⁴⁰

1.2.1 Water Decontamination Using Carbon Filters

Organic contaminants such as benzene, endrin, lindane, methoxychlor, 1,2-dichloroethane, 1,1-dichloroethylene, 1,1,1-trichloroethane, total trihalomethanes (TTHMs), toxaphene, trichloroethylene, 2,4,5-TP (silvex), para-dichlorobenzene, mercury (Hg^{2+}) inorganic and organic mercury complexes, arsenium, and chromium have been filtered from drinking water to acceptable standards by AC filtration.⁴¹⁻⁴⁴ The activated carbon (AC) has been used as filter which interacts strongly with organic molecules.

AC has been widely used for many purposes both in laboratory and industrial settings, due to its ability to absorb organic compounds and inorganic metal complexes. Since its introduction in analytical chemistry, enrichment of trace metals using AC has been favorably performed with very high concentration factors in different matrices. The mechanism of sorption on AC is still under investigation. Adsorption equilibrium studies have revealed that pH is the dominant parameter controlling adsorption. Increasingly

stringent regulations related to the treated water quality drive a worldwide interest in membrane technology applications, whether for removal of particles or dissolved matters. Microfiltration (MF) as well as ultrafiltration (UF) are viewed as particle removal processes. Use of MF or UF processes is expanding rapidly as an alternative from water to conventional filtration processes to meet increasingly stringent regulations related to the treated water quality and, in particular, to pathogens such as *Giardia* and *Cryptosporidium*. The membranes used in these processes do not remove color, natural organic matter, or synthetic organic chemicals. MF and UF must be combined with other conventional technologies such as AC adsorption and coagulation to overcome some of these limitations. Table 1.1 shows water contaminants that can be reduced to acceptable standards by AC filtration. There is also a problem with the AC whereby adsorption capacity decreases by a number of factors, including a decrease in the number of available

Primary Drinking Water	Standards Contaminant MCL*	mg/L
Inorganic Contaminants		
Organic arsenic complexes	0.05	
Organic chromium complexes		0.05
Mercury (Hg ⁺²) inorganic	0.05	
Organic Contaminants		
Benzene	0.005	
Methoxychlor		0.1
1,2-Dichloroethane	0.005	
1,1-Dichloroethylene	0.007	
1,1,1-Trichloroethane		0.200
Total trihalomethanes (TTHMs)		0.10
Trichloroethylene	0.005	
2,4-D		0.1
2,4,5-TP (silvex)	0.01	
Para-dichlorobenzene	0.075	

*MCL—Maximum Contaminant Level

TABLE 1.1 Water Contaminants Reduced to Acceptable Standards by AC Filtration

adsorption sites and an increase in the negative surface charge caused by adsorption of some material (i.e., humic acid) with the AC.

1.2.2 Carbon Nanotube as Filter Material

Carbon nanotubes (CNTs),⁴⁵⁻⁴⁸ a new form of carbon, are attracting great research interest due to their exceptional adsorption and mechanical properties and unique electrical property, highly chemical stability, and large specific surface area mainly because of their extremely small sizes, uniform pore distribution, and large specific surface area.^{45,49} Their well-defined porosities and functionalities render CNTs' superior adsorbents to other adsorbents like AC. Nanotube surfaces are chemically modified to enhance the adsorption of specific ions or molecules. Due to their porous structure, CNTs are found to have much higher adsorption than that of a carbon pack, graphitized carbon black with the same surface area as CNTs. This is also confirmed by first principles calculations that water, carbon dioxide, and methane adsorb weakly to the surface of carbon nanotubes, whereas oxygen and other molecules behaving as charge acceptors have adsorption energies four to five times larger than that of water. Of course, adsorption sites in a single-walled carbon nanotube (SWCNT) bundle can be inside of the tubes (pore), the interstitial triangular channels between the tubes, the outer surface of the bundle, or the grooves formed at the contact between adjacent tubes on the outside of the bundle. In other words, certain functionalities or defect sites on the surface of the nanotube may play a role in the adsorption of molecules and ions. Adsorption of 2,3-dichlorophenol was found to be dependent on the mass of multi-walled carbon nanotubes (MWCNTs), concentration, solution pH, and adsorption temperature. MWCNTs were found to be an excellent adsorbent for removing 2,3-dichlorophenol from aqueous solutions in a very short period of time.⁵⁰ The purified multiwalled carbon nanotubes are also used as a trap-packing material to adsorb VOCs purged from a spiked water sample. For MWCNTs, adsorption can occur in the aggregated pores, inside the tube, or on the external walls. In this latter case, the presence of defects, as incomplete graphene layers, has to be taken into consideration. Although adsorption between the walls has been proposed in herringbone-type graphite nanofibers, it is unlikely to occur in the case of MWCNT due, for many molecules, to a steric effect and should not prevail for small molecules due to long diffusion paths. It has been reported that CNTs had significantly higher dioxin removal efficiency than that of AC. CNTs are good fluoride absorbers and their fluoride removal capability is superior to that of an AC filter.

1.2.3 Functionalization of Carbon Nanotubes for Adsorption

Oxidation of CNTs with oxidized acid can also introduce many functional groups, such as hydroxyl ($-\text{OH}$), carboxyl ($-\text{COOH}$), and carbonyl ($>\text{C}=\text{O}$), on the surface of CNTs.^{51–53} Adsorption properties of raw MWCNT were greatly improved by oxidation, as well as by amino functionalization. It was found that adsorption capacities change with increasing temperature, whereby the amino-functionalized MWCNT had the best adsorption capacity for Cd^{2+} . This gives the possibility that functionalized MWCNTs could be used to produce filtration membranes for the removal of heavy metals from industrial waters at higher temperatures and for preconcentration of heavy metals in analytical chemistry and environmental protection.⁵⁴ CNT functionalization is considered to be one of the distinct properties of CNTs. The noncovalent method of functionalization is preferred over the covalent method because pore textural properties of CNTs are not affected. The primary goal of CNT functionalization is to increase its water solubility so that it is possible to use them in many practical applications.^{55,56} Removal of viruses increased at higher ionic strengths (NaCl) due to the suppression of repulsive electrostatic interactions between viruses and SWCNTs.⁵⁷

These functional groups attached to the surfaces of CNTs improve their adsorption capabilities of metal ions and organic pollutants in solution. Heavy metal adsorption on CNTs has been shown to depend on surface functional groups, specific surface area, and solution components. The most important factor is the surface functional group, which generates oxidized acids. It is well known that oxidation treatment by nitric acid causes an increase in cation-exchange capacity. Different functional groups with an acidic nature introduced on the surfaces of CNTs can dissociate at different pH values.

1.3 Photocatalytic (TiO_2) Interaction with Organic and Inorganic Compounds

In line with current environmental problems, the applicability of the aforementioned combination of adsorption and TiO_2 -based heterogeneous photocatalysis has been studied as an environmentally friendly water treatment technology on a laboratory scale.^{58–73} Nanocomposite titania nanoparticles are chemically functionalized to increase their quantum yield. These nanostructured photocatalysts are to be used for water treatment, as well as for decontamination of environmental pollutants.

The photocatalytic material such as TiO_2 has been used extensively for environmental decontamination purposes.^{58–73} Several researchers^{63–73} have tried to understand the decontamination mechanism of TiO_2 based nanomaterials in a membrane, filter or colloidal

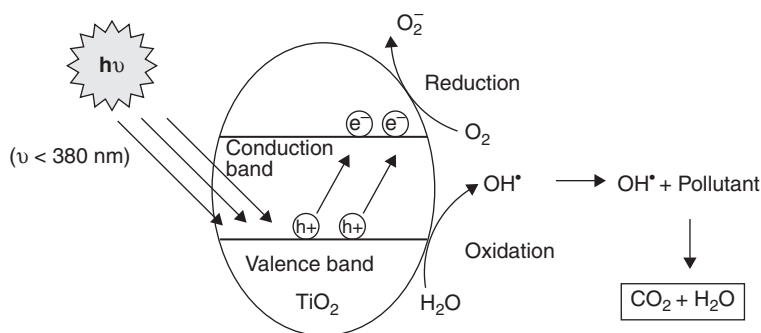


FIGURE 1.1 Mechanism of the photocatalytic effect of TiO₂.

with organic or biomolecules. Figure 1.1 shows the mechanism of photocatalytic effect of TiO₂ nanomaterials with organic pollutant. TiO₂ is a semiconductor with a bandgap energy of 3.2 eV. When electrons are excited by the light of energy equal or exceeding its bandgap energy, they are promoted from the valence band to the conduction band, leaving positive holes in the valence band. These electrons and holes are capable of reducing and oxidizing compounds at the TiO₂ surface, respectively. If electrons and holes do not recombine to produce heat, they can follow the reductive and oxidative pathways indicated by the reactions. In addition, holes often react with water or hydroxyl ions adsorbed to TiO₂, producing hydroxide radicals, which then oxidize adsorbed organics. The TiO₂ occurs in rutile, anatase and brookite phases but the anatase phase of TiO₂ is photocatalytic active.



where $h\nu$ = light energy, h^+ = hole, e^- = electron, M^{n+} = oxidized compound, and $\cdot\text{R}_{(\text{ads})}$ = the adsorbed organic species.

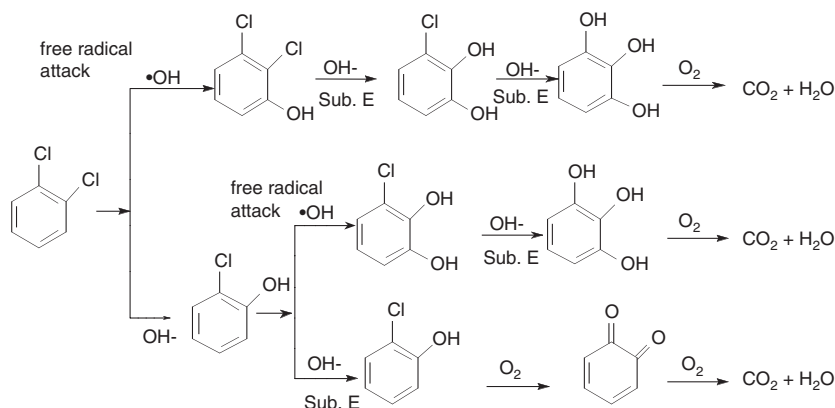


FIGURE 1.2 Photocatalytic oxidation and remediation of dichlorobenzene.

Typically, a photocatalytic reaction is conducted in a suspension of submicrometer semiconductor material and, therefore, requires an additional separation step to remove the catalyst from the treated water. Removing such fine particles from large volumes of water involves further expense and presents a major drawback to applying photocatalytic processes for treating wastewaters. To minimize these problems, research has been carried out by immobilizing titania onto various substrates, such as glass beads, sand, silica gel, quartz optical fibers, and glass fiber, in the form of mesh or on a glass reactor wall. Although this approach provides a solution to the solid–liquid separation problem, slurry-type reactors offer significant advantages over immobilized catalyst-type reactors in terms of catalyst surface availability and superior mass-transfer properties.

It is always important to understand the leaching of TiO_2 nanoparticles while it is used in water purification purposes. TiO_2 has been shown to precipitate in surface photochemical processes resulting in the oxidation of the adsorbed substrate, such as C_{60} . This implies that fullerene derivatives may act as sensitizers when they attach to the surface of TiO_2 . Due to this effort, there will be a photo-induced charge transfer from fullerene moiety to TiO_2 . Self-assembly materials containing C_{60} covalently attached to TiO_2 nanoparticles. Figure 1.2 shows the photocatalytic oxidation and remediation of dichlorobenzene using TiO_2 nanoparticles.

1.4 Enzymatic Decontamination

Enzymes that are capable of hydrolyzing and detoxifying such agents are of significant use. The gene for the bacterial phosphotriesterase was subcloned into several expression systems, including *E. coli*, insect cells, and *in vitro* compartmentalization. An appropriate mixture of enzymes and other natural products offers considerable

advantages over other decontaminants.⁷⁴⁻⁹⁰ Because enzymes are catalytic, they are highly efficient and can detoxify many times their own weight of agent in seconds or minutes. Unlike most chemical catalysts, many different types and specificities of enzymes can be mixed together in a single formulation. Because enzymes function best at pH values near neutrality, there are few, if any, compatibility or corrosion concerns as long as the material being decontaminated can tolerate water. There will not be any flammability concerns using a water-based system. Similar to enzymes in commercial laundry detergents, an enzyme-based decontaminant, being biodegradable will pose little or no health or environmental danger and leave no hazardous products that would need to be dealt with. Potentially, an enzyme-based decontaminant will be benign enough to be used directly on the skin of personnel and casualties. Another major advantage is that an enzyme-based decontaminant would be provided as a dry granulated powder that would be added to whatever water-based spray or foam system the user has available. This provides a significant reduction in logistical burden as well as using existing equipment, both military and civilian. The immobilization of enzymes has been extensively studied for various large-scale applications due to potential advantages including product purification, catalyst recycling, and continuous operation. The limiting factor of the application of enzyme immobilization is due to the rather poor biocatalytic efficiency of the immobilized enzymes. The most toxic organophosphorus compounds known are the lethal nerve agents tabun (GA), sarin (GB), soman (GD), and VX, shown in Fig. 1.3. The toxicity of these compounds is due primarily to the practically irreversible inhibition of acetylcholine esterase (AChE), the enzyme responsible for hydrolysis of the neurotransmitter acetylcholine.

The reaction mechanism for the enzymatic hydrolysis of acetylcholine by AChE is shown in Fig. 1.4. During this transformation, an active-site serine residue initiates a nucleophilic attack on the carbonyl carbon of acetylcholine to form a covalent acetyl-acyl-enzyme intermediate, concurrent with the release of choline from the active site. The free enzyme is regenerated in the second step via a hydrolytic attack by water and the release of acetate.

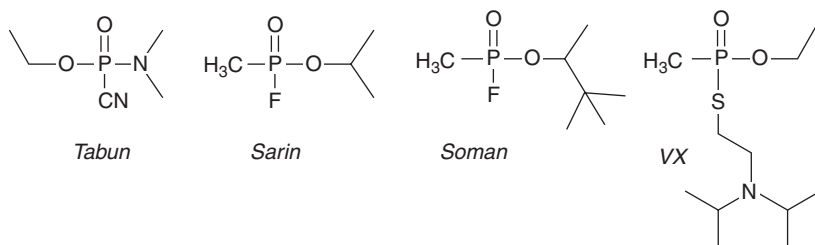


FIGURE 1.3 Schematic of nerve agents, tabun (GA), sarin (GB), soman (GD), and VX.

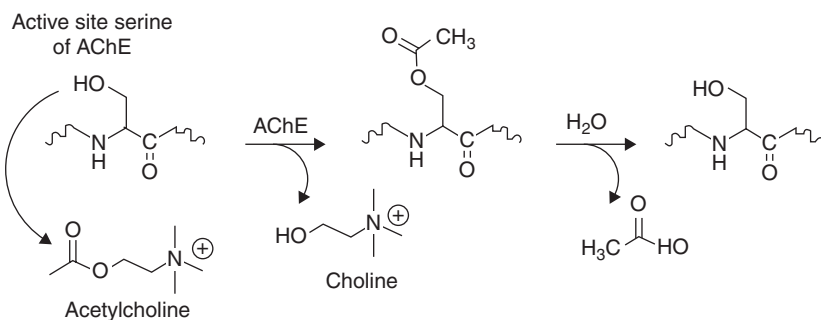


FIGURE 1.4 Reaction mechanism for enzymatic hydrolysis of acetylcholine by AChE.

Phosphotriesterase also possesses the ability to catalyze the hydrolysis of a wide spectrum of organophosphate insecticides including parathion, methyl parathion, and Metafos, among others. In addition, phosphotriesterase is capable of hydrolyzing nerve agents sarin, soman, and VX, but the catalytic efficiencies for these substrates are significantly lower than the less toxic insecticides which include tabun (GA), sarin (GB) and soman (GD). Hydrolysis in basic media works well for these agents, but less well with sulfur mustard (H or HD). Direct-base hydrolysis is not effective for V agents, an example of which is VX. However, oxidation of the sulfur in VX in an aqueous acid medium is rapidly followed by hydrolysis to nontoxic products. An acidic medium also causes protonation of the amine nitrogen, both increasing the solubility of VX and enhancing oxidation on sulfur.

1.5 Nanomaterial for Chemical Warfare Agents

Currently, many advances have been made to neutralize harmful chemical agents.⁹¹⁻¹⁰¹ To ensure their general effectiveness, military decontamination solutions have traditionally been highly aggressive chemicals. The Navy decontamination (decon) solution uses hypochlorous acid, whereas the Army decon solution (DS2) uses sodium hydroxide and diethylenetriamine. DS2 is a mixture of diethylenetriamine (70%), diethylene glycol monomethyl ether, household bleach (5%), and supertopical bleach (>5%). Calcium hypochlorite (HTH) is an important decontamination solution available on the market. Nanoparticles such as nanotubes can incorporate biochemical catalysts that will lead to an arsenal of smart nanoparticles for specific remediation applications. The key point is that such smart nanoparticles will allow for reduced use of highly aggressive chemical systems.

Metal oxides like silica, alumina, α - Fe_2O_3 , MgO , and La_2O_3 having particles of larger size were observed to have decontamination properties and unique surface chemistry of promising chemical warfare agents (CWAs). Decontamination of 2-chloroethyl ethylsulfide using titanate nanoscrolls reactions could render them harmless. The reaction chemistry involving detoxification of CWAs is shared by some organic pollutants and pesticides. Recent experiments showed that powders of metal oxides MgO , CaO , Al_2O_3 , and TiO_2 , when penetrated by CWAs, initiate reactions found in liquid decontamination schemes in addition to binding agents to their surfaces.^{91–103} Certain chemical reactions facilitate chemical warfare decontamination. Nucleophilic substitution of the C—X or P—X bond, elimination of H—X (X=Cl, F and CN), hydrolysis of C—Cl or P—X (X=Cl, F, and CN) bond, oxidation of —C—S— or —P—OR bond, and formation of surface-bound products of CWA such as alkoxides or phosphonates are some reactions that assist in the decontamination of chemical warfare. Hydrolysis reactions observed for GD, VX, and HD are attributed to the basic MgO or CaO or Al_2O_3 surface and are strictly analogous to the known solution chemistry of the agents. Moreover, catalysis was suspected as a means of sustaining some of the reactions. It was expected that nanocrystal powders would be more effective than powders of conventional microcrystals due to larger surface areas and larger numbers of available reactive sites due to surface, corner, and edge defects. In reality, much of that surface area would not be accessible to adsorbates due to the aggregation of nanocrystallites, rendering many facets of nanocrystallites inaccessible. Sulfur mustard initially reacts with surface of ZnO to form sulfonium ion; soon after that, the sulfonium ion interacts with water molecules present within ZnO nanostructures to form thiodiglycol and hemisulfur mustard (Fig. 1.4). Moreover, sulfur mustard reacts with the ZnO surface by the elimination reaction and releases hydrochloric acid and further yields elimination products like divinyl sulfide, vinyl chloroethyl sulfide, and vinyl hydroxyl ethyl sulfide (Fig. 1.5). Released hydrochloric acid may react with the ZnO surface and can form minute amounts of zinc chloride species thereby poisoning the active surface. ZnO nanowires were grown by dipping the Si n-type substrates upside down in the growth solution at 80°C for 2 h.

1.5.1 Decontamination due to Zero Valent Ions

Use of nanoscaled zero-valent iron particles (nZVI) to remediate contaminated soil and groundwater has received increasing amounts of attention within the last decade,^{103–107} primarily due to its potential for broader application, higher reactivity, and cost effectiveness compared to conventional zero-valent iron applications and other in

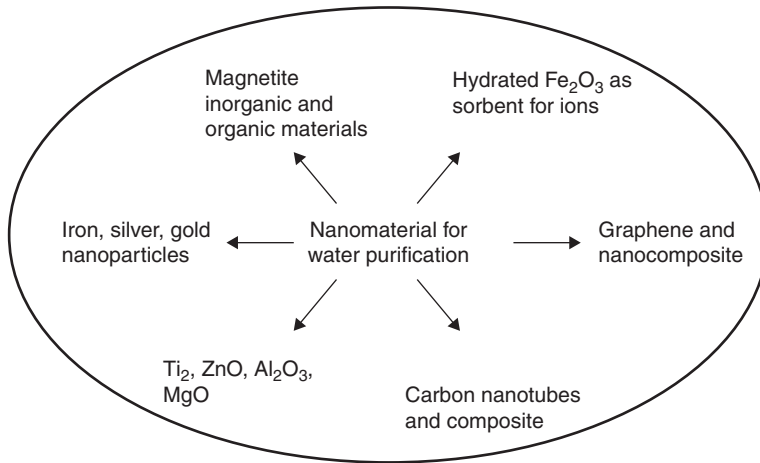


FIGURE 1.5 Application of nanomaterials in areas of contaminant destruction/sorption.

situ methods. However, the potential environmental risks of nZVI in situ field scale applications are largely unknown at present, and traditional environmental risk assessment approaches are not yet able to be completed. Nanoparticles may produce ROS through photocatalytic events or if the metal ion can exist in different redox states (e.g., Fe, Mn, Cu, and Co) by the Fenton reactions.

The development of nZVI particles are also questioned regardless of its immense applicability of decontamination.¹⁰⁷ This is, in part, due to extensive and profuse uncertainties regarding the potential health and environmental risks of nZVI, similar to cases of other engineered nanoparticles. In fact, only a few studies to date have investigated the potential for nZVI to pose an environmental risk including a very small number of toxicity and ecotoxicity studies of nZVI's potential to cause adverse effects in laboratory studies. In addition, previous cases that involved substituting one set of risks by another set of risks, termed "risk-risk trade-offs" such as methyl tertiary-butyl ether as a fuel additive, and other examples that involved costly cleanup efforts illustrate the need to thoroughly analyze technology options before their full-scale introduction and particularly within early stages of development.

Among the many applications of nanotechnology that have environmental implications, remediation of contaminated groundwater using nanoparticles containing nZVI is one of the most prominent examples of a rapidly emerging technology with considerable potential benefits.¹⁰⁴⁻¹⁰⁷ The focus here is the use of nano-scale reactive iron (NZVI) particles for treatment of dense nonaqueous phase liquid (DNAPL) contamination. However, it

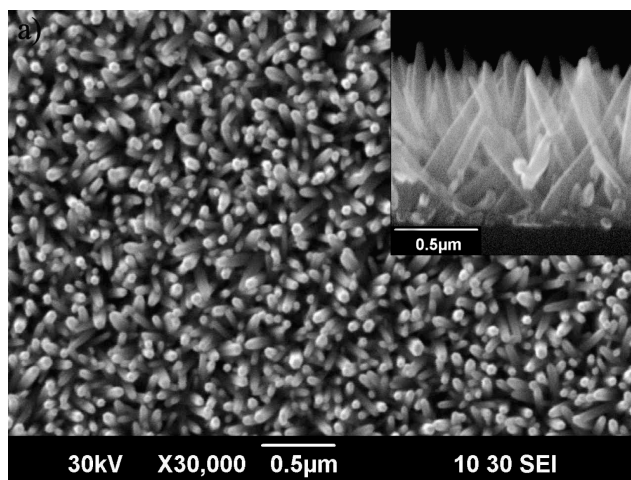


FIGURE 1.6 SEM image of the sample as grown nanowires. Inset: cross section of the sample with nanowires.¹⁰²

is recognized that a range of materials are viable for treating a range of contaminants. The current and future challenges of using NZVI for subsurface remediation are not from material performance but stem more from environmental factors and operational aspects. All laboratory studies have been done in homogeneous and well-defined batch or column experiments. However, for remediation applications, NZVI is applied to physically and chemically heterogeneous and poorly defined subsurfaces. A heterogeneous layer structure of aquifer materials on a subsurface can create preferential flow that controls the transport and emplacement of NZVI. Although both solution-phase synthesis and vapor-phase synthesis can be used to produce NZVI, most of the synthesis approaches of NZVI particles, which are commercially available or normally used in laboratory routines, are solution-phase approaches. Vapor-phase synthesis normally yields relatively small quantity of particles; therefore, for remediation that requires a large quantity of NZVI, the vapor-phase approach might not be appropriate. Emulsification of NZVI is another novel modification of NZVI for active in situ remediation. This approach is adapted from emulsified oil flushing for DNAPL remediation by magnetic separations. Magnetic materials, when scaled down to nanosize, show different magnetization behavior than bulk magnetic material does. Because of this, the magnetic separation characteristics would be affected. If scaling down the size generates hard magnetic material, then magnetic filter regeneration would be difficult. Soft magnets with low magnetic permanence and superparamagnetic materials with

zero magnetic remanence are preferred for magnetic separation applications. Magnetic nanoparticles could be separated based on their nanostructures, as the ease of direction of magnetization would vary depending on the ordering of atoms in the magnetic structure.

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