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Rhizobiont in Bioremediation of Hazardous Waste

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Remediation of Toxic Environmental Pollutants Using Nanoparticles and Integrated Nano-Bio Systems

Amrita Kharangate-Lad and Nezlyn Cressida D'Souza

Abstract

Rapid industrialization and urbanization have led to the release of many toxic contaminants in the environment. Environmental pollution is a major concern as it affects health and well-being of all organisms. The toxicity and health hazards associated with the environmental pollutants requires clean-up from the soil and water. Remediation using nanotechnology is an effective alternative to conventional physicochemical methods as it is eco-friendly and economical. In this chapter we emphasize the use of nanoparticles in the remediation of toxic environmental pollutants such as heavy metals, hydrocarbons, antibiotics, hormones, dyes, and recalcitrant agro-based and organohalide compounds. Nanoparticles are synthesized biogenically which involves its synthesis using plants and microorganisms. Nanobioremediation is a technology that integrates microbial bioremediation and nanoremediation. This chapter provides a brief outlook on applications of nanobioremediation through integrated nano-bio systems in removal of organic or inorganic toxic pollutants and discusses the implications of using these integrated systems on a larger scale. The benefit of this collaboration is the development of more efficient, cost-effective, environmentally friendly solutions in remediation of toxic environmental pollutants.

Keywords

Nanoparticles · Microbes · Synthesis · Toxic pollutants · Remediation

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19.1 Introduction

Environmental pollution is undeniably a major issue that the society faces. The environment gets polluted with toxic compounds released due to industrialization and unplanned urbanization. Toxic pollutants like heavy metals, particulate matter, industrial effluents, sewage, pesticides, fertilizers, oil spills, and other organic compounds are a few examples of environmental pollutants. The use of conventional physical and chemical technologies in the treatment of contaminated soils and water is expensive as they have high energy requirements and usually leave behind hazardous waste residues. Therefore, cost-effective bioremediation technologies that use microorganisms to detoxify and transform pollutants have been developed for the treatment of polluted soils and water (Abatenh et al. 2017; Kumar et al. 2018; Tomei and Daugulis 2013). Another technology that is being explored currently is the use of nanoparticles in cleaning up toxic pollutants from contaminated sites. Nanoparticles have highly desirable properties that favour their application in remediation of recalcitrant pollutants from soil and water.

19.2 Properties of Nanoparticles

Nanoparticles possess unique physicochemical properties and therefore, nanotechnology has gained a lot of attention in the last decade. Nanoparticles are nanoscopic particles having dimensions within the range of 1–100 nm. Due to their nanoscale size these particles exhibit exceptional physicochemical properties such as high energy, large specific surface area, and unique visible properties (Fig. 19.1). Nanoparticles by virtue of their small size have properties that differ from the bulk metal. They exhibit physical and chemical properties that are a virtue of their nano-size. The nano-size of the particles imparts them with unique chemical properties such as large surface per unit area and high reactivity. Nanoparticles exhibit plasmon resonance as owing to their small size they have the ability to confine their electrons and produce quantum effects. The media used in the synthesis of nanoparticles and the number of bioactive compounds present in the media directly influence the chemical and morphological properties of nanoparticles (Yadav et al. 2017).

Based on the chemistry, the nanoparticles are broadly classified into organic and inorganic. Organic nanoparticles include carbon nanoparticles (like carbon nanotubes (CNT)), while inorganic nanoparticles include metals (such as nanoscale zerovalent iron (nZVI), palladium, cadmium), noble metals (gold and silver), semi-conductors (copper, zinc oxide, and titanium oxide), and magnetic nanoparticles (maghemite, magnetite) (Guerra et al. 2018).

Nanoparticles have different ways of interacting with each other, some tend to remain as individual moieties, while others tend to group together, depending upon the attractive or repulsive forces acting between them. Agglomeration of nanoparticles is often linked with oxidation-induced-instability, which leads to decrease in the surface area and reactivity of the particles. Coating of nanoparticle with suitable stabilizers increases their adsorbing capacity while decreasing their

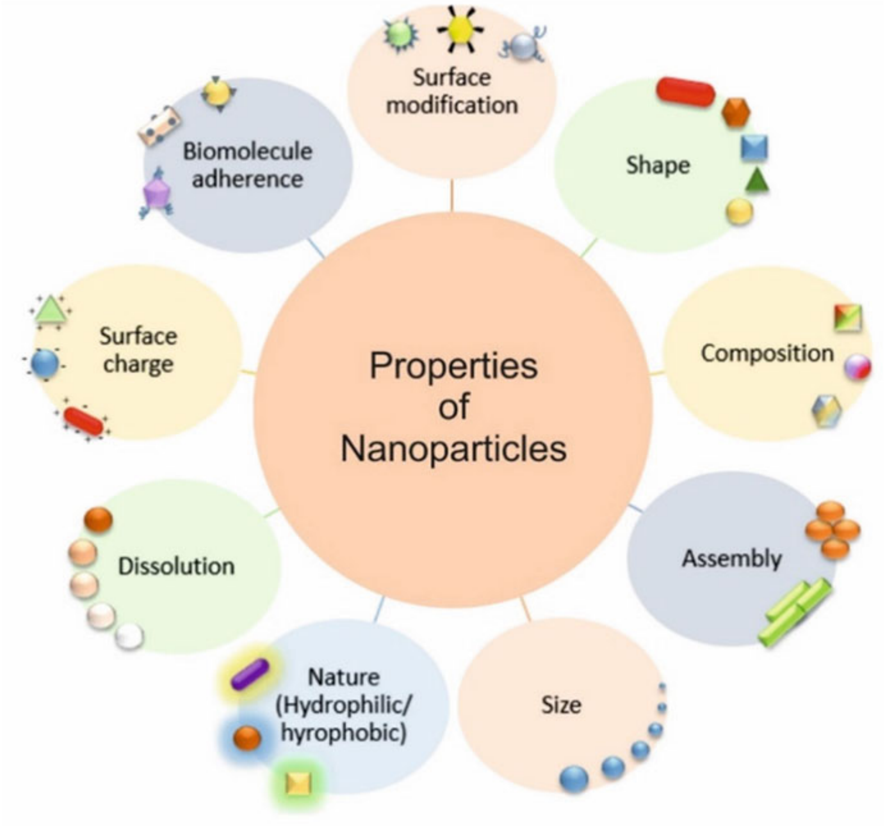

Fig. 19.1 Properties of nanoparticles

tendency to agglomerate. Immobilization with organic polymers such as lactate, gaur gum, gellan gum, etc. enhances their interaction with the toxic compounds by improving nanoparticle mobility, stability, and reactivity. Nanoparticles can be designed to exhibit specific physical properties and chemical composition in order to target specific pollutants (Sakulchaicharoen et al. 2010).

19.3 Nanoparticles and Nanomaterials

Morphologically, nanoparticles can be spheres, sheets, cylinders, and tubes (Pal et al. 2019). However, they differ from nanomaterials owing to their dimensions. Nanomaterials have been defined as "a manufactured or natural material that possesses unbound, aggregated, or agglomerated particles where external dimensions are between 1 and 100 nm size range", according to the EU Commission (Jeevanandam et al. 2018). Nanomaterials therefore have a length of 1–1000 nm in at least one dimension. Based on the shape, size, structure, and composition, nanomaterials can be classified into nanoclusters, nanofibres, nanotubes, nanoshells, and nanocomposites.

Nanomaterials have successfully demonstrated their ability to degrade environmental pollutants from soil and ground/surface water. Metal-based nanomaterials composed of silver (Ag) nanoparticles have been used for disinfection of water and soil from *Escherichia coli* and *Pseudomonas aeruginosa*. Titanium dioxide (TiO_2) nanoparticles have been proven efficient against *Escherichia coli,* MS-2 phage and hepatitis B virus. Other titanium-based nanoparticles have been reportedly used for remediation of aromatic hydrocarbons (phenanthrene), chlorinated compounds (2-chlorophenol), organic pollutants (endotoxins), microorganisms (*Escherichia coli* and *Staphylococcus aureus*), dyes (Rhodamine B), and biological nitrogen (nitric oxide) (Guerra et al. 2018). Binary mixed oxides, iron nanoparticles, and bimetallic nanoparticles have been used for the treatment of water containing methylene blue, heavy metals, and chlorinated and brominated compounds, respectively (Jeevanandam et al. 2018). Due to their minute size and surface properties, they can enter very small spaces in the soil and attain access to contaminants well below the subsurface level. Since movement of these nanoparticles is governed by Brownian motion, they can remain suspended in groundwater facilitating wider transport of the particles and more efficient interaction with the contaminant (Tratnyek and Johnson 2006).

19.4 Synthesis of Nanoparticles

The modification of surface properties of the nanoparticle improves its interaction and compatibility with the pollutant of interest. Nanoparticles, in general, are synthesized using two approaches: top-down and bottom-up. The top-down approach centres on gradual degradation of bulk materials to nanosized particles, while the bottom-up approach involves assembly of atoms and molecules into a nanoparticle (Narayanan and Sakthivel 2010). The former approach involves usage of expensive and energy-intensive physical methods, while the latter can be further sub-classified into chemical and biological methods. The employment of high-priced toxic solvents and extreme physical conditions (such as high pressure and temperature) in chemical synthesis of nanoparticles renders the chemical process as environmentally unfriendly (Narayanan and Sakthivel 2010). Biological methods, which utilizes plants and microbes (algae, bacteria, yeasts, actinomycetes, and fungi) serve as an eco-friendly and less expensive alternative to top-down synthesis of nanoparticles (Fig. 19.2) (Sastry et al. 2003).

19.4.1 Biogenic Production of Nanoparticles Using Plants

The biosynthesis of nanoparticles generally entails oxidation–reduction reactions. Plants and microorganisms have enzymes and produce antioxidizing and reducing

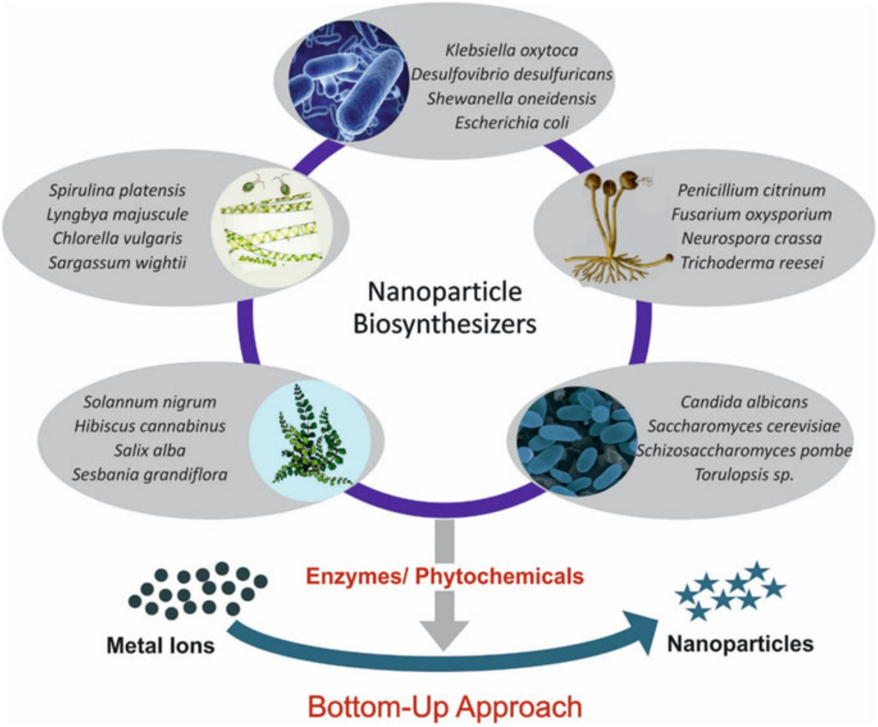

Fig. 19.2 Biogenic synthesis of nanoparticles using bottom-up approach

phytochemicals, which facilitate reduction and detoxification of accumulated metal ions. In the synthesis of metal/metal oxide nanoparticles, plant extracts (from roots, leaves, flowers, stems) are reported to be effective as they contain phytochemicals such as amides, aldehydes, ascorbic acids, carboxylic acids, flavones, ketones, phenols, and terpenoids. These compounds have been reported to have ability to reduce metal salts into metal nanoparticles. Plants such as Alfalfa, oats, Tulsi, lemon, Aloe vera have been reported to be used in production of silver and gold nanoparticles (Singh et al. 2016). Through optimization of process parameters, nanoparticles of desired sizes and morphologies can be biosynthesized (Iravani 2014). The application of whole plants or their phytochemical extracts in synthesis of nanoparticles has been demonstrated through numerous studies (Hussain et al. 2016). Biogenic production using plants is a preferred method as it is easier to scale-up (Megharaj et al. 2011).

19.4.2 Biogenic Production of Nanoparticles Using Microorganisms

Microbes possess highly versatile and effective catalytic mechanisms that enable degradation of various toxic compounds (Watanabe 2001). Additionally, microbes are ubiquitous, much easier to culture and manipulate, have high growth rates, and require low cost investment. Silver nanoparticles have been biogenically produced by using bacteria such as *Arthrobacter gangotriensis, Aeromonas* sp. SH10, *Bacillus amyloliquefaciens, Bacillus cecembensis, Bacillus indicus, Bacillus cereus, Corynebacterium* sp. SH09, *Escherichia coli, Enterobacter cloacae, Geobacter* sp., *Lactobacillus casei, Phaeocystis antarctica, Pseudomonas proteolytica,* and *Shewanella oneidensis.* Gold nanoparticles have been reported to be extensively produced using *Bacillus subtilis* 168, *Bacillus megaterium* D01, *Desulfovibrio desulfuricans*, *Escherichia coli* DH5a, *Plectonema boryanum* UTEX 485, *Rhodopseudomonas capsulate,* and *Shewanella alga*. Fungi have been used to synthesize zinc, gold, silver, and titanium metal/metal oxide nanoparticles. Biosynthesis of silver and gold nanoparticles by *Saccharomyces cerevisiae* has been reported (Singh et al. 2016).

Microorganisms synthesize inorganic nanoparticles by assimilating metal ions from the surrounding environment and converting them to their elemental form using enzymes. Heterotrophic sulphate reducing bacteria have been reported to reduce gold-(I)-thiosulphate complex (Au $(S_2O_3)_2^{3-}$) to elemental gold (Au^0) (Lengke and Southam 2006). Similarly, *Stenotrophomonas maltophilia* SELTE02 mediated transformation of selenite (SeO_3^{2-}) to elemental selenium (Se^0) (Gregorio et al. 2005). Microbial biosynthesis can be achieved through intracellular and extracellular routes. The intracellular method consists of transporting metal ions into the cell, followed by enzymatic reactions, whereas the extracellular method involves adsorption of the metal ions on the cell surface, followed by enzyme-moderated reduction (Zhang et al. 2011). Microorganisms are known to synthesize various types of nanoparticles including metallic nanoparticles (gold, silver, cobalt, chromium, and other metals), magnetic, non-magnetic nanoparticles, and alloy nanoparticles (Li et al. 2011).

The biogenic method of nanoparticle production is an enzyme-mediated "green" synthesis of nanoparticles (Fig. 19.2). It allows elimination of costly, inefficient methods that involve use of expensive, hazardous solvents (such as sodium borohydride, ammoniacal silver nitrate) and produce toxic by-products. Furthermore, the particles generated by biogenic processes have greater surface area and higher catalytic reactivity (Mohanpuria et al. 2008). Table 19.1 lists microorganisms involved in the biogenic synthesis of various nanoparticles.

19.5 Nanoparticles: Mechanism of Action

Nanoparticles exhibit surface and chemical properties that are distinct from the bulk metal due to their nano-size. The nano-size increases the mechanical, magnetic, electrical, and optical properties of the nanoparticles. Nanoparticles catalyse processes such as adsorption and chemical reactions such as photocatalysis, precipitation, reduction, oxidation, and hydrogen peroxide dependent oxidation (Fenton reactions). The adsorption capacity of nanoparticles is being used to develop sorbents like carbon nanotubes and zeolites that have demonstrated efficient removal

Table 19.1 Biogenic production of nanoparticles by microorganisms

Biogenic production of nanoparticles by microorganisms		
Nanoparticle	Microorganism	References
Iron (Fe)	*Pleurotus* sp.	Mazumdar and Haloi 2011
	Bacillus subtilis	Sundaram et al. 2012
	Escherichia coli	Arcon et al. 2012
	Klebsiella oxytoca	Anghel et al. 2012
	Chaetomium globosum	Elcey et al. 2014
Zinc (Zn)	*Desulfobacteriaceae*	Labrenz et al. 2000
	Lactobacillus sp.	Selvarajan and Mohanasrinivasan 2013
	Streptomyces sp.	
	Aspergillus terreus	Raliya and Tarafdar 2013
	Sphingobacterium thalpophilum	Raliya and Tarafdar 2014; Rajabairavi et al. 2017
Palladium (Pd)	*Bacillus sphaericus*	Creamer et al. 2007
	Desulfovibrio desulfuricans	Gurunathan et al. 2015
	Bacillus benzeovorans	Omajali et al. 2015
	Shewanella sp.	Zhang and Hu 2017
	Geobacter sulfurreducens	Yates et al. 2013
Copper (Cu)	*Penicillium aurantiogriseum*	Honary et al. 2012
	Penicillium citrinum	Cuevas et al. 2015
	Penicillium waksmanii	Tiwari et al. 2016
	Stereum hirsutum	Parveen et al. 2016
	Bacillus cereus	Parveen et al. 2016
	Pseudomonas sp., *Serratia* sp.	Parveen et al. 2016
Silver (Ag)	*Bacillus licheniformis*	Kalimuthu et al. 2008
	Trichoderma reesei	Nanda and Saravanan 2009
	Cladosporium cladosporioides	Balaji et al. 2009
	Candida sp.	Kumar et al. 2011
	Streptomyces naganishii	Alani et al. 2012
Gold (Au)	*Rhodococcus* sp., *Streptomyces* sp.	Ahmad et al. 2003
	Aspergillus fumigatus, A. flavus	Gupta and Bector 2013
	Rhodopseudomonas capsulata	He et al. 2007
	Stenotrophomonas maltophilia	Nangia et al. 2009
	Penicillium brevicompactum	Mishra et al. 2011
Titanium (Ti)	*Lactobacillus* sp.	Prasad et al. 2007
	Bacillus subtilis	Vishnu Kirthi et al. 2011
	Aspergillus flavus	Rajakumar et al. 2012
	Aspergillus terreus	Raliya and Tarafdar 2014
	Bacillus amyloliquefaciens	Khan and Fulekar 2016
Cadmium (Cd)	*Escherichia coli*	Sweeney et al. 2004
	Rhodopseudomonas palustris	Bai et al. 2009
	Fusarium sp.	Reyes et al. 2009
	Bacillus amyloliquefaciens	Singh et al. 2011
	Bacillus licheniformis	Shivashankarappa and Sanjay 2015

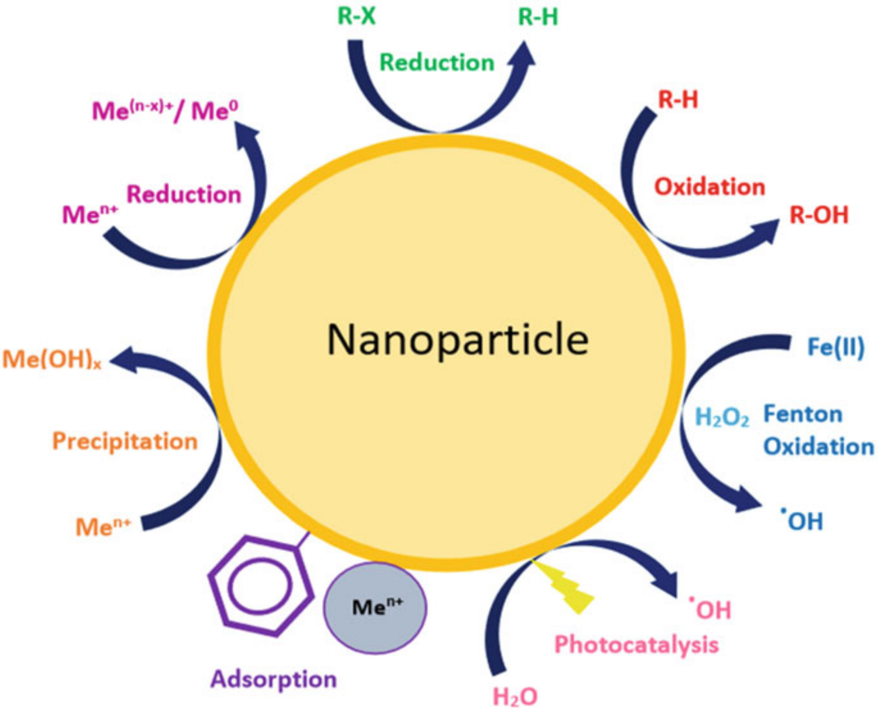

Fig. 19.3 Mechanisms catalysed by nanoparticles in degrading toxic pollutants

of anions and metals from drinking water (Yaqoob et al. 2020). Nanoparticles mediate the transformation of toxic metals into their respective hydroxides by precipitation. Nanoparticles play a crucial role in nanobioremediation by catalysing reactions and generating hydrogen ions that aid the microorganisms to further detoxify and transform the compound. The high reactivity and larger surface area for interaction offer the nanoparticles the characteristic property of having high oxidation potential. Metal oxide nanoparticles have been shown to catalyse oxidation reactions that can transform toxic pollutants such as hydrocarbons. Nanoparticles can absorb photons and act as nanophotocatalysts. The larger surface area of nanoparticles increases the oxidation of organic pollutants at the surface of the nanoparticles resulting in their degradation. TiO_2 is the most commonly used nano-photocatalyst in treatment of wastewater, due to its low cost, chemical stability, easy availability, and zero toxicity. Composite nanomaterials like copper oxide doped (CuO-doped) zinc oxides (ZnO) have been used to catalyse the Fenton oxidation. Fenton oxidation is a H_2O_2-assisted photo-decomposition of toxic pollutant in the presence of solar radiation (Banik and Basumallick 2017). Figure 19.3 summarizes the mechanisms catalysed by nanoparticles in degradation of toxic pollutants.

19.6 Remediation of Toxic Metals Using Nanoparticles

Heavy metals constitute a group of metals and metalloids that have atomic density higher than 4000 kg/m^3 (Edelstein and Ben-Hur 2018). Although plants need various heavy metals such as molybdenum, zinc, iron, etc. for basic metabolic activities, at higher concentrations, these metals are harmful to the plant (Vardhan et al. 2019). Heavy metals are toxic to humans even at low concentrations. Due to urbanization, population explosion, and rapid industrialization, soil and water resources end up as heavy metal sinks. Bioremediation measures thus play an important role in restoring balance within these contaminated environments, failure of which would lead to bioaccumulation and biomagnification of these toxic metals at the cellular level.

Nanomaterials such as bimetallic nanoparticles, metal oxides, nanoscale zeolites, and carbon nanotubes have been considered for metal remediation. Iron oxide nanoparticles have been extensively studied in metal detoxification owing to their ability to be separated easily from the reaction media and due to their low toxicity. Immobilization of accumulated heavy metals in soil plays a crucial role in their transformation and detoxification. Nanoparticles such as nano-alginate, bentonite-nZVI, zerovalent iron nanoparticles, nanocarbon, and dendrimers have been used to immobilize the heavy metals in soil (Helal et al. 2016).

Nano-hydroxyapatite, a phosphate-based nanomaterial has demonstrated significant results in clean-up of Pb contaminated soils. It has also been reported to immobilize other heavy metals such as Cr, Zn, Cd, and Cu. Studies on both graphene oxide nanoparticles and nZVI nanoparticles have been reported in relation to increasing the bioavailability of arsenic and other metals in polluted soils (Baragaño et al. 2020). Similar studies using graphene as the adsorbent showed removal of arsenic by 80% from wastewater. The advantages of this nanomaterial are its ability to be recycled and reused. A nanocomposite with reduced graphene oxide and Fe_3O_4 has been studied for the removal of Pb^{2+} ions from water by co-precipitation and has been found to be efficient. Other graphene-based nanomaterials have been successfully used for the removal of heavy metals such as Cu (II), Fe (II), Mn (II), and Pb (II). Activated carbon nanomaterial has been successfully applied in the removal of heavy metals such as Zn^{+2} and Al^{+3} ions and has demonstrated an efficiency up to 92%. In wastewater treatment, it showed an absorption capacity of up to 50% for Cu^{+2} ions. Other activated carbon nanomaterials have been successfully employed in remediation of heavy metals such as Pb^{+2}, Cd^{+2}, Cu^{+2}, Ni^{+2}, and Zn^{+2} (Baby et al. 2019). The role of various environmental factors affecting the remediation of heavy metals by iron-based nanoparticles has been studied. This study also focused on the toxicity of these nanoparticles on the living organisms (Latif et al. 2020). Hydrous iron oxide-based nanomaterials have been found to remove heavy metals like Pb (II), Cu (II), Cd (II), and Ni (II). However, they have been found to be most effective in removal of arsenic (V) from drinking water. Nanomaterials based on manganese oxides have been used for the treatment of metals such as Tl (I), U, Pb (II), Zn (II), Hg (II), Cd (II), and Cu (II) and have now been considered promising in removal of heavy metals from polluted water. Polyvinylpyrrolidone-coated magnetic nanoparticles (PVP-Fe_3O_4 nanoparticles) have shown significant efficiency (100%) in the removal of Cd, Cr, Ni, and Pb. Hexavalent chromium (VI) is a prevalent inorganic pollutant that is known to be toxic, mutagenic, and carcinogenic (Valko et al. 2005). Remediation of Cr (VI) involves its reduction to trivalent Cr (III), a less toxic, non-carcinogenic form that can be precipitated into hydroxides. This reaction serves as a key step for the removal of Cr (VI) from aqueous solutions (Mabbett et al. 2004). Iron nanoparticles such as ferromagnetic carbon-coated nanoparticles have been used to remove 95% of Cr (VI) from aqueous solutions. Table 19.2 shows the use of nanoparticles in heavy metals remediation.

19.7 Remediation of Hydrocarbons Using Nanoparticles

Hydrocarbon pollution refers to the contamination of the environment by organic polycyclic aromatic hydrocarbons (PAH) or crude oil (petroleum hydrocarbons) compounds. Oil pollution has severe environmental and ecological consequences. Crude oil and its derivatives are considered as some of the most pervasive environmental pollutants. The leakage of crude oils into aquatic systems results in wide-ranging contamination. Oil sludge is a thick complex emulsion made up of various petroleum hydrocarbons, heavy metals, sediments, and water. It is composed of alkanes, aromatics, resin, and asphaltenes and contains volatile and semi-volatile organic compounds, which are reported to be genotoxic (Srivastava et al. 2019). Unintentional and deliberate spillage of oil into the environment leads to transfer of toxic organic materials into the food chain.

Due to their unique physicochemical properties, nanoparticles have gained tremendous interest in oil biodegradation. Nanoparticles can decrease the hydrophobicity of these hydrocarbon compounds and thus increase their bioavailability for bioremediation. Graphene-doped TiO_2 nanoparticles have demonstrated adsorption and photodegradation of phenanthrene, fluoranthene, and benzopyrene to phthalic and benzoic acid; oxygenated and hydroxylated PAHs; acyclic hydrocarbons and alcohols (Bai et al. 2017). Similarly, iron hexacyanoferrate (FeHCF) nanoparticles have been employed in degradation of anthracene, chrysene, and fluorine. It was also established that aromatic hydrocarbon adsorption by the FeHCF followed a molecular weight dependent trend: anthracene > phenanthrene > fluorene > chrysene > benzopyrene (Shanker et al. 2017). Bimetallic magnetic Pt/Pd nanoparticles supported on magnetic silica reduced anthracene to non-toxic partially hydrogenated species. Furthermore, compared to monometallic nanoparticles, the bimetallic particles displayed enhanced activity post recycling (Zanato et al. 2017). Nanoparticles are efficient in degrading organic compounds such as benzophenone and phenol. Benzophenone-3 is an ultraviolet absorber, and thus they can be degraded using catalytic ozonation. Manganese oxide and cobalt oxide nanoparticles immobilized on ceramic membrane displayed remarkable

Table 19.2 Nanoparticles used in remediation of heavy metals

Nanoparticles used in remediation of heavy metals		
Pollutant	Nanoparticle	References
Copper (Cu)	Amino-functionalized magnetic nanoparticles	Hao et al. 2010
	Polydopamine nanoparticles	Farnad et al. 2012
	Maghemite nanoparticles	Rajput et al. 2017
	nZVI	Boente et al. 2018
	Graphene oxide–manganese oxide nanoparticles composite	Wan et al. 2018
Chromium (VI) (Cr)	Chitosan-nZVI (nanoscale zerovalent iron) nanoparticles	Geng et al. 2009
	Magnetite–maghemite nanoparticles	Chowdhury and Yanful 2010
	Cerium oxide nanoparticles	Contreras et al. 2015
	CTAB modified magnetic nanoparticles	Elfeky et al. 2017
	Magnetic alkaline– Lignin−dopamine nanoparticles	Dai et al. 2019
Arsenic (As)	Magnetite–maghemite nanoparticles	Chowdhury and Yanful 2010
	Iron oxide nanoparticles	Shipley et al. 2011
	Cupric oxide nanoparticles	Reddy et al. 2013
	γ-Aluminium oxide nanoparticles	Ghosh et al. 2019
	Magnetic iron-titanium binary oxide composite	Deng et al. 2019
Cadmium (Cd)	Cerium oxide nanoparticles	Contreras et al. 2015
	Iron phosphate (vivianite) nanoparticles	Qiao et al. 2017
	EDTA functionalized iron oxide nanoparticles	Huang and Keller 2015
	Silica-coated iron oxide nanoparticles	Ghafoor and Ata 2017
	Graphene oxide–manganese oxide nanoparticles composite	Wan et al. 2018
Lead (Pb)	Iron phosphate (vivianite) nanoparticles	Liu and Zhao 2007
	Cerium oxide nanoparticles	Contreras et al. 2015
	EDTA functionalized iron oxide nanoparticles	Huang and Keller 2015
	L-cysteine stabilized nZVI nanoparticles	Bagbi et al. 2017
	Maghemite nanoparticles	Rajput et al. 2017
Mercury (Hg)	Carboxymethyl cellulose stabilized iron sulphide nanoparticles	Gong et al. 2012
	Citrate-coated gold nanoparticles	Ojea-Jiménez et al. 2012
	Silica-coated magnetite nanoparticles	Mohmood et al. 2016
	Inorganic magnetic sulphide nanoparticles	Patel et al. 2018

catalytic ozonation of benzophenone-3 (Guo et al. 2016). Reduced graphene oxide silver nanoparticles (rGO-Ag) were demonstrated to successfully breakdown phenol (Bhunia and Jana 2014). ZnO and magnetite-zinc oxide (Fe_3O_4-ZnO) hybrid nanoparticles have been studied in the degradation of phenols and the hybrid nanoparticles were found to be more efficient (Feng et al. 2014). UV-irradiated semiconductor catalysts such as titanium dioxide have been used in photodegradation of organic contaminants (Banik and Basumallick 2017). Calcium peroxide nanoparticles have been reported to exhibit remarkable degradation of benzene and toluene (Mosmeri et al. 2017; Qian et al. 2013). Removal of benzene from aqueous solutions has also been carried out using magnetite nanoparticles. The nano-magnetic particles have been reported to decompose 98.7% and 94.5% of the benzene in batch and continuous conditions, respectively. Furthermore, these particles can easily desorb benzene and be reused for its removal (Amin et al. 2013). Magnetite nanoparticles catalysed decomposition of p-nitrophenol to aromatic intermediates such as benzoquinone and hydroquinone, which were further oxidized by hydrogen peroxide. The nanoparticles exhibited the ability to be magnetically separated from the sludge and reused (Sun and Lemley 2011). Magnesium doped magnetite nanoparticles have been reported to effectively decompose hexachlorobenzene, a precursor of carcinogenic and mutagenic compounds (Su et al. 2014).

The remediation of petroleum-based hydrocarbons using nanoparticle-stabilized surfactants has been explored (Ali et al. 2020). Iron nanoparticles produced biogenically from *Vaccinium floribundum* showed a significant removal of up to 86% and 88% in the total petroleum hydrocarbons from polluted soil and water, respectively (Murgueitio et al. 2018). Raw and distillate forms of petroleum oil were treated with nanocarbon scavengers which efficiently sequestered these hydrocarbons by 80% and 91%, respectively (Daza et al. 2017). Polyvinylpyrrolidone (PVP) coated magnetite nanoparticles (NP) have demonstrated remediation of lower chain alkanes by 70% and higher chain alkanes by 65% after incubation of 1 h. However, the efficiency of degradation of these alkanes increased significantly on incubation of the nanoparticles with oil degrading bacteria (Alabresm et al. 2018). Table 19.3 enlists integrated systems studied for hydrocarbon degradation. Other studies have shown significant results in degradation of hydrocarbons using nanobioremediation, which has been discussed further in this chapter.

19.8 Remediation of Hormones, Antibiotics, and Medicinal Drugs Using Nanoparticles

Antibiotics and hormones are emerging pollutants. Modern healthcare and their extensive use in farming results in their release in effluent and wash waters leading to contamination of groundwater and soil. Their recalcitrant nature makes them persist in the soil and water and lead to serious effects on the environment and human health.

Table 19.3 Nanoparticles used in remediation of hydrocarbon pollutants

Nanoparticles used in remediation of hydrocarbons		
Pollutant	Nanoparticle	References
Polycyclic aromatic hydrocarbons (PAHs)		
Pyrene	Haematite nanoparticles	Jorfi et al. 2016
Phenanthrene, fluoranthene, benzopyrene	Titanium dioxide nanoparticles	Bai et al. 2017
Anthracene, phenanthrene, chrysene, fluorene, and benzopyrene	Iron hexacyanoferrate nanoparticles	Shanker et al. 2017
Anthracene	Bimetallic magnetic Pt/Pd nanoparticles	Zanato et al. 2017
Benzophenone-3	Manganese oxide and cobalt oxide nanoparticles coated on ceramic membrane	Guo et al. 2016
Phenol	Magnetite and zinc oxide hybrid nanoparticles Reduced graphene oxide silver nanoparticles (rGO-Ag)	Feng et al. 2014; Bhunia and Jana 2014
Petroleum-based hydrocarbons		
Total petroleum hydrocarbons	Iron nanoparticles	Murgueitio et al. 2018
Raw and distillate forms of petroleum oil	Nanocarbon nanoparticles	Daza et al. 2017
Alkanes	Polyvinylpyrrolidone (PVP) coated magnetite nanoparticles	Alabresm et al. 2018

19.8.1 Remediation of Antibiotics

Nanoparticles have been reported to clean up even low concentrations of these compounds. Many nanoparticles like zerovalent iron nanoparticles have exhibited the ability to remove antibiotics such as sulfacetamide from the aqueous solution by 61% (Table 19.4). Bimetallic $CuFe20_4$ nanoparticles have been efficiently used for the removal of amoxicillin from aqueous solutions. Photocatalytic degradation of ciprofloxacin has also been reported using graphene oxide/nanocellulose composite. Similar degradation studies have also been reported on ciprofloxacin using zerovalent iron nanoparticles. Tetracycline degradation was successfully carried out by nanoscale zero-valent iron (nZVI) and Fe/Ni bimetallic nanoparticles in aqueous solution. Although Fe/Ni nanoparticles performed better that nZVI, the removal efficiency of both the particles was reported to decrease with increase in pH, thus asserting that the degradation process is pH-dependent (Dong et al. 2018b). UiO-66, a zirconium based nanoparticle doped with cobalt was reported to adsorb and degrade tetracycline via photocatalysis. Factors affecting the adsorption process included dosage of the nanoparticles, coexisting ions, pH, and initial concentration of the antibiotic (Cao et al. 2018). The Oxone process has been utilized to degrade organic compounds. In this process, Oxone [peroxymonosulfate (PMS)] is

Table 19.4 Nanoparticles used in remediation of antibiotics, medicinal drugs, and hormones

Nanoparticles used in remediation of antibiotics, medicinal drugs, and hormones		
Pollutant	Nanoparticle	References
Antibiotics		
Amoxicillin	Copper-iron bimetallic nanoparticles	Malakootian et al. 2019
Ceftriaxone	Fe_3O_4 nanoparticles	Malakootian et al. 2019
Sulfacetamide	Zerovalent iron nanoparticles	Malakootian et al. 2019
Tetracycline	Fe/Ni bimetallic nanoparticles Cobalt-doped UiO-66 nanoparticles Martite nanoparticles Cu/Fe bimetallic particle ZnS nanoparticles Titanium oxide nanocomposite	Dong et al. 2018b; Cao et al. 2018; Soltani et al. 2018; Malakootian et al. 2019
Ciprofloxacin	Zinc oxide nanoparticles Fe-doped zinc oxide nanoparticles Titanium oxide nanoparticles Cupric oxide nanoparticles Graphene oxide/nanocellulose composite Zerovalent iron nanoparticles	Gharaghani and Malakootian 2017; Das et al. 2018; Malakootian et al. 2020
Levofloxacin	Zn oxide nanoparticles, Graphene oxide sheets	El-Maraghy et al. 2020
Hormones		
Estrone	Polystyrene nanoparticles Titanium dioxide nanoparticles	Akanyeti et al. 2017; Czech and Rubinowska 2013
17-ß-Estradiol	Manganese oxide nanoparticles Magnetic biochar nanoparticles	Han et al. 2014; Dong et al. 2018a
Medicinal drugs		
Paracetamol	Activated carbon nanoparticles	Baby et al. 2019
Carbamazepine	Haematite nanoparticles Magnetite nanoparticles	Rajendran and Sen 2018; Liu et al. 2018
Ibuprofen	Nanoscale zerovalent iron (nZVI)	Machado et al. 2013

combined with a source of transition metals to generation sulphate ions, which catalyse the redox process. Martite (α-Fe_2O_3) nanoparticles have been found to activate the Oxone process to degrade the recalcitrant antibiotic (Soltani et al. 2018).

The degradation of ciprofloxacin, another commonly used antibiotic, was proved both spectrophotometrically and microbiologically by the loss of antibiotic activity, using Fe-doped-ZnO nanoparticles (Das et al. 2018). Immobilized nanoparticles allow easy placement and removal of aqueous contaminants. Moreover, it resolves complications related to separation of catalysts from the solution, as well as allows reusability of nanoparticles. ZnO immobilized on a glass plate exhibited nano-photocatalysis of ciprofloxacin with high removal efficiency (Gharaghani and Malakootian 2017). Similarly, TiO_2 nanoparticles immobilized on a glass plate were shown to adsorb and photodegrade ciprofloxacin and also resulted in decreased sludge production (Malakootian et al. 2019). Other antibiotics such as ampicillin, amoxicillin, and penicillin were reduced by bentonite-supported Fe/Ni nanoparticles (Weng et al. 2018).

19.8.2 Remediation of Medicinal Drugs

Medicinal drugs such as paracetamol, carbamazepine, and ibuprofen not just pollute water bodies, but pose a significant threat to aquatic communities. As in case of other organic compounds, a variety of nanoparticles have been researched for their applicability in degrading these recalcitrant compounds. Ibuprofen reduction can be carried out by nZVI nanoparticles. While effective on lab scale, simulation of the nanoremediation with ibuprofen contaminated soil concluded that the degradation process is slower in aqueous systems, but without any significant impact on contaminant degradation efficiency (Machado et al. 2013). Haematite nanoparticles were demonstrated to be effective adsorbents of carbamazepine and thus offer a potential remediation method for the adsorptive elimination of drugs from contaminated aquatic systems (Table 19.4) (Rajendran and Sen 2018). Magnetite nanoparticles were also confirmed to effectively remove carbamazepine (Liu et al. 2018). Nanoparticles have also been proved effective in reducing chlorhexidine, a commonly used antiseptic component. Photocatalytic degradation of chlorhexidine was successfully concluded using titanium dioxide nanoparticles as photocatalyst (Das et al. 2014). Activated carbon nanoparticles have been reportedly used for the bioremediation of methylene and paracetamol from hospital wastewaters (Baby et al. 2019).

19.8.3 Remediation of Hormones

Naturally synthesized and synthetic hormones are one of the most potent trace contaminants released into soil and water bodies. Apart from excretion and disposal of hormones, application of animal faeces as manure to fields allows transmission of endogenous hormones into soil and ultimately in water bodies. These hormones and endocrine disruptors are known to affect the reproductive health of various animals by disrupting sexual function and promoting abnormal sexual development. As with other organic compounds, nZVI, under oxidative conditions, was found to adsorb and degrade 17β-estradiol (Table 19.4) (a popular endocrine disrupting chemical) and 17α-ethinylestradiol (a synthetic birth control drug) into less bioactive estrone (Jarosova et al. 2015). Biochar, a pyrogenic black carbon product of biomass pyrolysis can be used to derive nanosized biochar particles due to their nanoporous structure, good stability, multiple functional groups, and easy availability. Magnetic biochar nanoparticles (Mag-BCNPs) are a composite of nanosized biochar nanoparticles and magnetic iron nanoparticles (Fe_3O_4). This composite was demonstrated to effectively reduce 17β-estradiol. Post remediation, the Mag-BCNPs can be recovered using a magnet, and the biochar can be desorbed by purging with ozone, thus allowing easy regeneration of the nanoparticles (Dong et al. 2018a). Carboxymethyl cellulose (CMC) stabilized manganese oxide nanoparticles have also been studied for their 17β-estradiol degrading activity (Han et al. 2014). Estrones can be removed from wastewater using TiO_2-assisted photocatalysis (Czech and Rubinowska 2013). A hybrid polystyrene nanoparticle-ultrafiltration system with renewable nanoparticles acts as a lucrative solution to filtering estrone from contaminated water. However, due to the higher permeability of the filtration system compared to that of nanofiltration/reverse osmosis systems, estrone removal capacity of 40% was achieved. However, replacing the filtration system with a finer one can increase success of the designed system (Akanyeti et al. 2017). Table 19.4 shows the use of nanoparticles in remediation of antibiotics, medicinal drugs, and hormones.

19.9 Remediation of Dyes and Organic Solvents Using Nanoparticles

With the onset of industrialization, the synthesis and utilization of various organic compounds have increased exponentially. These organic compounds include dyes which are released into the environment through anthropogenic activities. These pollutants are known to have neurotoxic, mutagenic, and carcinogenic effects on biological systems (Das and Chandran 2011). Various studies using nanoparticles have been carried out on degradation of the dyes. Nanomaterials such as reduced graphene oxide, (rGO)-TiO_2 has been successfully applied for the bioremediation of Alizarin Red S. Other studies have also reported the use of graphene oxide nanomaterials in association with TiO_2 or ZnO in photodegradation of methylene blue. Bimetallic nanoparticles of iron (Fe) and nickel (Ni) showed significant results in cleaning up of the azo dye Orange G from wastewater (Foster et al. 2019). Similarly, remarkable efficiency was exhibited when absorbent made of ZnO nanoparticle was used to remove azo dyes, amaranth, and methyl orange from aqueous solution (Zafar et al. 2019). Tara and others (Tara et al. 2020) have summarized the nanoparticles used in the removal of Azo dyes and the efficiency of removal of these

Table 19.5 Nanoparticles used in remediation of dyes and organic solvents

Nanoparticles used in remediation of dyes, and organic solvents		
Pollutant	Nanoparticle	References
Dyes		
Dye containing effluents	Nickel nanoparticles	Kale and Kane 2017
Azo dyes	ZVI nanoparticles Cerium loaded copper oxide Iron nanoparticle	Tara et al. 2020
Methylene blue, Congo red	Copper oxide nanoparticles Copper oxide nanoneedles Graphene oxide sheets	Tara et al. 2020
Cationic and anionic dyes	Nickel doped zinc oxide nanoparticle	Tara et al. 2020
Coomassie brilliant blue, Amido black 10B	Copper oxide nanoneedles Graphene oxide sheets	Tara et al. 2020
Malachite green, methyl violet	Magnetic NPs coated with poly (dimethylaminoethyl acrylamide) and methyl acrylate on modified with Fe_3O_4 nanoparticles	Tara et al. 2020
Methylene blue	Copper oxide nanoparticles activated with montmorillonite clay	Tara et al. 2020
Methylene blue, methyl orange, rhodamine B, organic dyes	Silver nanocomposite, Silver nanoparticles	Tara et al. 2020; Bhakya et al. 2015
Amaranth and methyl orange	ZnO nanoparticles	Zafar et al. 2019
Orange G	Fe and nickel nanoparticles	Foster et al. 2019
Organic solvents		
TCE	Zerovalent iron nanoparticles	Ibrahem et al. 2012
p-Nitrophenol	Magnetite nanoparticles	Sun and Lemley 2011
Hexachlorobenzene	Magnetic micro/nano Fe_xO_y-CeO_2 composite Mg-doped iron oxide nanoparticles	Jia et al. 2010; Su et al. 2014
Benzene	Magnetite nanoparticles Calcium peroxide nanoparticles	Amin et al. 2013; Mosmeri et al. 2017
Toluene	Calcium peroxide nanoparticles Tungsten doped titanium dioxide nanoparticles	Qian et al. 2013; Poorkarimi et al. 2017

dyes by the respective nanoparticles. Table 19.5 summarizes different nanoparticles that are involved in remediation of dyes and organic solvents.

Nanoscale iron nanoparticles have shown significant efficiency in removal of chlorinated organic solvents contaminating groundwater (Zhang and Elliott 2006). Zerovalent iron nanoparticles have been used for the remediation of trichloroethylene (TCE). It was considered an efficient reducing catalyst and resulted in fast degradation of TCE from contaminated soils (Ibrahem et al. 2012).

19.10 Remediation of Agro-Based Compounds Using Nanoparticles

Agro-based compounds such as herbicides, fungicides, pesticides, weedicides are extensively used in agriculture. These compounds leach and contaminate the surrounding soil and water bodies. Due to the complex chemical structure of these compounds they are recalcitrant in nature and tend to persist in the environment for a long time. Nanoparticles have been found to be efficient in removal of pesticides from contaminated sites by adsorption and photodegradation of these compounds (Table 19.6). Nanomaterials such as zinc oxide, titanium dioxide, and silver have been reported to photocatalyse the degradation of pesticides. Metal nanocomposites and polymer matrix nanocomposites have been successfully used in the removal of organic and inorganic pollutants. Iron nanoparticles have been successfully used in remediation of carbamate pesticides such as methomyl. Fe_3O_4 nanoparticles have

Table 19.6 Nanoparticles used in remediation of agro-based compounds

Nanoparticles used in remediation of agro-based compounds		
Pollutant	Nanoparticle	References
Aldrin	Fe_3O_4 nanoparticles	Sahithya and Das 2015
Alachlor	Nanoscale zerovalent iron (nZVI)	Bezbaruah et al. 2009
Eldrin	Fe_3O_4 nanoparticles	Sahithya and Das 2015
Lindane	Fe_3O_4 nanoparticles, silver nanoparticles	Sahithya and Das 2015
	Nanoscale zerovalent iron (nZVI)	Elliott et al. 2009
Organophosphate pesticides	TiO_2, ZnO, Au-TiO_2, CuO nanoparticles	Sahithya and Das 2015
Atrazine	Nanoscale zerovalent iron (nZVI)	Bezbaruah et al. 2009
Chlorpyrifos	Silver and gold nanoparticles	Bootharaju and Pradeep 2012
DDT	Mixed metal oxides nanoparticles	Manav et al. 2018
Profenofos (organophosphorous pesticide)	Fe/Ni bimetallic nanoparticles	Mansouriieh and Khosravi 2015
Malathion	Iron oxide nanoparticles and iron oxide/ gold nanoparticles Magnetite nanoparticles and gold coated magnetic nanoparticles	Fouad et al. 2015
Acetamiprid	Fe/Ni bimetallic nanoparticles	Marcelo et al. 2016

been applied for their efficiency in clean-up of organochlorine pesticides such as aldrin, endrin, and lindane. Silver nanoparticles have also been efficient in remediation of pesticide lindane. Organophosphate pesticides such as monocrotophos, malathion, chlorpyrifos, dichlorvos have been photo-catalytically degraded using nanoparticles such as titanium dioxide, zinc oxide, gold-titanium dioxide, and copper oxide, respectively.

Gold-titanium dioxide (Au-TiO_2) nanoparticles have been reported to photocatalyse the degradation of chloridazon (Sahithya and Das 2015). Rapid anoxic degradation of pesticide alachlor (2-Chloro-2′,6′-diethyl-*N*-(methoxymethyl)-acetanilide) has been reported using nZVI. The reaction resulted in dechlorinated alachlor, which was significantly more biodegradable in comparison to its parent compound (Bezbaruah et al. 2009) (Table 19.6). The nZVI nanoparticles have been documented to degrade lindane (γ-hexachlorocyclohexane), a commonly used organochlorine pesticide, into benzene, diphenyl, and di/tetra/penta forms of the compound (Elliott et al. 2009). Silver and gold nanoparticles have exhibited remarkable ability to decompose chlorpyrifos, an organophosphorothioate pesticide, to 3,5,6-trichloro-2-pyridinol and diethyl thiophosphate at room temperature. These nanoparticles have also demonstrated an ability to be regenerated and reused (Bootharaju and Pradeep 2012). Catalytic degradation of Profenofos (an organophosphorous pesticide) and acetamiprid (a neonicotinoid insecticide) using Fe/Ni bimetallic nanoparticles has been reported (Marcelo et al. 2016; Mansouriieh and Khosravi 2015). $MgCeO_3$ and $MgFe_2O_4$ are mixed metal oxides that have been studied for degradation of DDT (dichlorodiphenyltrichloroethane), a well-known harmful organochlorine insecticide. Both mixed oxides degraded DDT to less toxic compounds DDD (dichlorodiphenyldichloroethane), DDE (dichlorodiphenyldichloroethylene), and simpler compounds within the first reaction hour (Manav et al. 2018). Zerovalent metal nanoparticles, semiconductor nanoparticles, and some bimetallic nanoparticles have been documented to treat environmental pollutants such as azo dyes, pesticides (chlorpyrifos and organochlorines), nitroaromatics, hormones, and antibiotics (Foster et al. 2019).

19.11 Remediation of Organohalide Compounds Using Nanoparticles

Organohalide compounds such as polybrominated diphenyl ethers (PBDEs), dioxins, polychlorinated biphenyls (PCBs), and chlorinated aliphatic hydrocarbons (CAH) are omnipresent, highly toxic fat-soluble compounds. Being insoluble in water, their low bioavailability hinders biodegradation by aquatic microbes. PCBs are toxic organohalide compounds classified as persistent organic pollutants. Various nanoparticles have been studied for their remediation. Pd/Fe bimetallic nanoparticles and nZVI have been reported to catalyse hydro-dechlorination of hexachlorobiphenyl (Chen et al. 2014). Carbon-modified titanium dioxide (CM-*n*-TiO_2) nanoparticles were reportedly used in photocatalytic degradation of PCB mixtures Aroclor 1254 and Aroclor 1260 (Shaban et al. 2016). Surfactants enhance

Table 19.7 Nanoparticles used in remediation of organohalide compounds

Polychlorinated biphenyls (PCBs)		
Pollutant	Nanoparticle	References
2-chlorobiphenyl	Palladium–aluminium (Pd/Al) bimetallic nanoparticles	Yang et al. 2013
Penta, hexa, hepta, and octobiphenyls	Nanoscale zerovalent iron (nZVI)	Gomes et al. 2014
2,2′,4,4′,5,5′-hexachlorobiphenyl	Nanoscale zerovalent iron (nZVI)	Chen et al. 2014
Aroclor 1254 and Aroclor 1260	Carbon-modified titanium dioxide (CM-*n*-TiO_2) nanoparticles	Shaban et al. 2016

solubilization of hydrophobic organic compounds, hence their integration with nanoparticles may lead to increase in bioremediation efficiency. In a combined electro-remediation set-up consisting of electrodialysis, non-ionic detergents (Saponin and Tween-80), and nZVI nanoparticles, degradation of up to 76% PCBs was reportedly achieved within 5 days. However, in a set-up with just surfactant and nanoparticles, the remediation efficiency was low, thus supporting the assumption that the surfactant will not always enhance nanoparticle activity (Gomes et al. 2014). Similar observations were made pertaining to remediation of 4-chlorophenol and 2-chlorobiphenyl using palladium-aluminium (Pd/Al) bimetallic nanoparticles (Table 19.7). While the nanoparticles by themselves were capable of degrading the PCBs, addition of ionic and non-ionic surfactants led to decrease in hydrodechlorination of the organic compounds (Yang et al. 2013). Polychlorinated aliphatic compounds such as trichloroethylene can be remediated using nanoparticles. Polymethyl methacrylate coated nZVI have also been reported capable in reducing trichloroethylene (Wang and Chiu 2009). Bimetallic Ni/Fe nanoparticles immobilized in hydrophilic nylon-66 membrane have been reported to completely dechlorinate trichloroethylene within 25 min. The immobilization of nanoparticles improved the reactivity and longevity of the nanoparticles (Parshetti and Doong 2009).

19.12 Nanobioremediation Using Integrated Nano-Bio Systems

Microbial bioremediation or nanoremediation, by itself offers multiple advantages over conventional treatment methods, but also has multiple noteworthy limitations. Microbial degradation of toxic compounds is a time-consuming process that can take over a year. Its effectiveness varies with conditions such as pH, nutrients, temperature, and other environmental conditions (Abatenh et al. 2017; Kumar et al. 2018). Integration of nanotechnology with microbial bioremediation can overcome these limitations and provide a better alternative to pollutant remediation. The reaction of the toxic pollutant with nanoparticles transforms the pollutant into compounds more easily metabolized by microorganisms. Therefore, nanobioremediation is a promising alternative in remediation technologies involving detoxification of toxic pollutants.

Nanobioremediation can be described as a combination of nanotechnology and bioremediation systems, wherein the former enhances the effectiveness of the latter. Therefore, nanobioremediation offers the benefits of both nanotechnology and bioremediation. The application of nanobioremediation can be done using two approaches. The sequential approach involves subjecting the contaminant to nanoparticles followed by exposure to the bioagent. The nanoparticles break down the contaminants to simpler compounds, allowing the bioagent to metabolize it further. In the concurrent or combined approach, the nanoparticle and biological agent is injected into the system simultaneously (Tratnyek and Johnson 2006; Karn et al. 2009). While both these methods seem to be practical alternatives to conventional technologies, more research and development is needed for their execution on a larger scale.

The most significant advantage of using nanoparticles in remediation is their application in both in situ and ex situ conditions. In ex situ remediation, the contaminated soil is excavated, while the contaminated groundwater is extracted and treated with nanoparticles on-site or off-site. In in situ treatment methods, nanoparticles are directly injected at the site of contamination where it degrades the contaminant. Compared to conventional methods, nanobioremediation is best suited for in situ treatment of contaminated soil and groundwater as the exceptional properties and colloidal nature of nanoparticles allow deep penetration up to the water table, a feat that is not possible to achieve with ex situ methods. Furthermore, nanoparticles enhance the reaction rates of microorganisms, thus allowing better degradation of pollutants (Karn et al. 2009). The ability of nanoparticles to degrade pollutants in association with biological systems presents opportunity for development of more efficient nano-bio integrated technologies. Apart from treatment and remediation of contaminated sites, nanoparticles can be applied to pollution sensing and pollution prevention.

19.13 Nanobioremediation of Toxic Heavy Metals

Integrated systems have great scope in metal bioremediation. Nanoparticles in combination with microorganisms have been used successfully to remediate heavy metals. Nanoscale zerovalent iron (nZVI) is smaller and more reactive than microscale zerovalent iron (ZVI) and therefore offers elevated reactive surface area, higher reactivity, and better injectability into aquifers, thus making them valuable in bioremediation of polluted sites (Li et al. 2006). However, due to their tendency to agglomerate and form larger particles, the reactivity of the nanoparticles decreases. Thus, direct application of free nZVI in remediation of contaminated soil and water could lead to overall decrease in efficiency of nanoparticles and leaching of iron into aquatic bodies. The immobilization of nZVI onto biodegradable, porous materials (beads) helps to overcome agglomeration. The toxic pollutant diffuses into beads and comes in contact with the entrapped nanoparticles. Similar studies on

Table 19.8 Nano-Bio integrated systems for remediation of Chromium (VI)

Integrated systems for remediation of toxic Chromium (VI)			
Metal pollutant	Nanoparticle	Bioagent	References
Bacteria			
Chromium (VI)	CNTs	*Shewanella oneidensis* MR-1	Yan et al. 2013
	CNTs	*Pseudomonas aeruginosa*	Pang et al. 2011
	Bio-Pd	*Clostridium pasteurianum* BC1	Chidambaram et al. 2010
Consortium			
Chromium (VI)	nZVI	Whey microbiota	Nemecek et al. 2016
Chromium (VI)	nZVI	Consortia of *Bacillus subtilis*, *Escherichia coli,* and *Acinetobacter junii*	Ravikumar et al. 2016

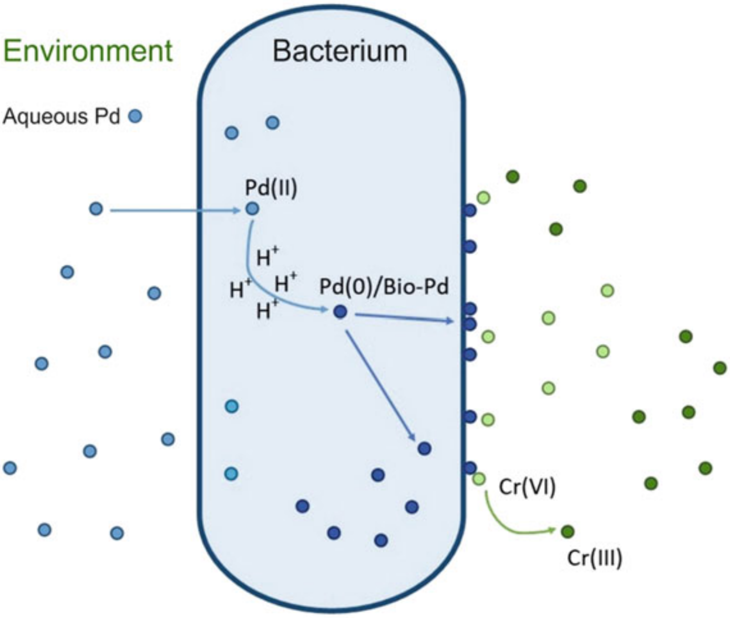

Fig. 19.4 Integrated Bio-Pd system in chromium remediation. The anaerobic hydrogen-producing bacterium accumulates aqueous palladium and reduces it to Bio-Pd, which further reduces toxic chromium (VI) to its less toxic form, Cr (III)

immobilization of nZVI on calcium alginate (nZVI−C − A beads), in presence of microbial biofilm has been shown to be effective in reducing Cr (VI) to Cr (III) (Ravikumar et al. 2016). Table 19.8 depicts various studies carried out using integrated nano-bio systems for remediation of chromium (VI).

The Bio-Pd (bio-reduced-Pd) integrated system is an innovative method for in situ remediation of toxic metals (Fig. 19.4). Bio-Pd generation is a novel system wherein bacterium, in the presence of hydrogen, reduces and precipitates accumulated palladium, Pd (II) into Pd nanoparticles (Pd (0)). The two-step remediation system involves synthesis of Pd nanoparticles (bio-Pd) followed by reaction with the contaminant. Although this two-step process is suitable for ex-situ remediation, hydrogen supply acts as a technical and commercial challenge. *Clostridium pasteurianum* BC1, an anaerobic hydrogen producer, allows coupling of in situ production of H_2 with in situ synthesis of nanoparticles, which further catalyses the degradation of contaminants. The bio-Pd synthesized by *Clostridium pasteurianum* BC1 successfully demonstrated the reduction of Cr (VI) to Cr (III) (Chidambaram et al. 2010).

Nanomaterials have been used to augment the metal-reduction efficiency of microorganisms. Compared to suspended cells, immobilized microorganisms allow continuous and stable bioreduction of Cr (VI). However, the alginate used for immobilization can lower the reduction efficiency due to its effect on electron and mass transport. Impregnation of organic nanomaterial such as carbon nanotubes (CNTs) has proved to enhance the mechanical strength and bioreduction capacity. The immobilization of *Shewanella oneidensis* MR-1 in alginate/CNT beads was shown to yield a higher reduction efficiency compared to the controls, possibly due to accelerated electron transfer (Yan et al. 2013). CNT-modified polyvinyl alcohol and sodium alginate matrix using immobilized *Pseudomonas aeruginosa* also enhanced bioreduction efficiency of the microorganism (Pang et al. 2011). Additionally, these modified beads can be reused effectively multiple times. This technique of CNTs impregnated immobilization can be applied to various other biological reduction processes and can be tested on various other pollutants.

19.14 Nanobioremediation of Polycyclic Aromatic Hydrocarbons (PAHs)

Hydrocarbons and organic pollutants are persistent in nature. The chances of degrading these compounds increase on application of nano-bio integrated systems. Hydrophobic polycyclic aromatic compounds (PAH) such as carbazole and indenopyrene are toxic and carcinogenic environmental pollutants. *Sphingomonas* sp. XLDN2–5 cells magnetically immobilized in Fe_3O_4 nanoparticle containing gellan gum matrix were proved to be effective in degrading carbazole. The addition of Fe_3O_4 nanoparticles to immobilized bacterial cells significantly enhanced its biodegradation activity (Wang et al. 2007). Yeast strains of *Candida tropicalis* NN4 have been reported to enhance indenopyrene biodegradation activity of zerovalent iron nanoparticles by producing glycolipid biosurfactant (Ojha et al. 2019). *Rhodococcus erythropolis* IGTS8 combined with magnetite (Fe_3O_4) nanoparticles have been found to effectively desulphurize dibenzothiophene (DBT), a recalcitrant organo-sulphur compound released on combustion of fossil fuels, to 2-hydroxybiphenyl (2-HBP). The nanoparticles have been reported to enhance cell growth rate, in comparison with the controls containing either the bacteria or the nanoparticles (Karimi et al. 2017). Immobilized *Pseudomonas*

Table 19.9 Integrated nano-bio systems for remediation of polycyclic aromatic hydrocarbons

Integrated nano-bio systems for remediation of polycyclic aromatic hydrocarbons			
Nanoparticle	Bioagent	Pollutant	References
Polycyclic aromatic hydrocarbons			
Fe^0 nanoparticles (ZVI)	*Candida tropicalis* NN4	Indenopyrene (InP)	Ojha et al. 2019
Pd/nFe bimetallic nanoparticles	Laccase enzyme from *Trametes versicolor*	Triclosan	Bokare et al. 2010
Magnetite (Fe_3O_4) nanoparticles	*Pseudomonas delafieldii*	Dibenzothiophene	Shan et al. 2005
Fe_3O_4 nanoparticles + gellan gum gel beads	*Sphingomonas* sp.	Carbazole	Wang et al. 2007
Magnetite nanoparticles	*Rhodococcus erythropolis* IGTS8	Dibenzothiophene (DBT)	Karimi et al. 2017
Nitro-aromatic compound			
Fe^0 nanoparticles (ZVI)	Methanogenic anaerobes	RDX (hexahydro-1,3,5-trinitro-1,3,5-triazine)	Oh et al. 2001

delafieldii coated with magnetite nanoparticles are also effective in degrading DBT (Shan et al. 2005). Successful degradation of a pollutant depends on its chemical structure. Some pollutants can be degraded completely, while others are broken down to less toxic moieties.

Triclosan, a halogenated aromatic hydrocarbon compound, is a commonly used antibacterial and antifungal component of soaps, toothpaste, and deodorants. Although triclosan is not considered as chemical pollutant of high concern currently, its degraded by-products have been detected in soil, wastewater, drinking water, and even human breast milk. Triclosan has been reported to bioaccumulate in various aquatic species, raising concern of it being an emerging toxic contaminant (Dhillon et al. 2015). Complete biotransformation of triclosan to non-toxic oligomers has been reported via sequential nanobioremediation. Pd/nFe bimetallic nanoparticles, under anaerobic conditions, dechlorinated triclosan to 2-phenoxyphenol, which, in the absence of nanoparticles, was oxidized into non-toxic products by laccase enzyme isolated from *Trametes versicolor* (Bokare et al. 2010). Table 19.9 lists some of the integrated nano-bio systems used for bioremediation of polycyclic aromatic hydrocarbons.

19.15 Nanobioremediation of Petroleum-Based Hydrocarbons

Although the studies conducted on nanobioremediation of oil sludge are few, researchers have demonstrated successful degradation of aliphatic and aromatic hydrocarbons of oil sludge using an integrated system of nanoparticles and microorganisms. The nanoparticles decrease the hydrophobicity of the hydrocarbons allowing them to be accessible to the microorganisms for degradation. The bacteria,

Table 19.10 Integrated systems for remediation of petroleum-based hydrocarbon pollutants

Integrated systems for remediation of petroleum-based hydrocarbon pollutants			
Nanoparticle	Bioagent	Pollutant	References
Fe_2O_3 and Fe_3O_4 nanoparticles	*Thalassospira* sp.	Oil sludge	Rizi et al. 2017a
Fe_2O_3 and Fe_3O_4 nanoparticles	*Chromohalobacter* sp.	Oil sludge	Rizi et al. 2017b
Consortium			
ZnO nanoparticles	*Bacillus, Pseudomonas, Achromobacter,* and *Microbacterium*	Crude oil	Ismail et al. 2013
NiO nanoparticles	*Halomonas xianhensis* A, *Halomonas zincidurans* B6, *Pseudomonas stutzeri* ATCC 17588, and *Halomonas salifodinae* BC7	Crude oil	El-Sheshtawy et al. 2017

Thalassospira sp. and *Chromohalobacter* sp. isolated from an oil site in the Persian Gulf has been studied to successfully degrade oil sludge into various non-toxic compounds. The addition of Fe_2O_3 and Fe_3O_4 nanoparticles, at low concentrations, further enhanced degradation of the compounds, without negatively impacting bacterial growth rate (Rizi et al. 2017a, b).

The interaction between nanoparticles and bacteria may not always be synergistic; nanoparticles can have toxic effects on the microbe and thus reduce its bioremediation potential. Crude oil degradation by the isolated bacteria: *Bacillus, Pseudomonas, Achromobacter,* and *Microbacterium sp.* was deemed successful; but addition of ZnO nanoparticles to *Pseudomonas* decreased its growth rate and lowered bioremediation potential (Ismail et al. 2013). This reduction can be attributed to inhibitory effect of the nanoparticles on the bacterium.

Low bioavailability of crude oil and their poor aqueous solubility obstructs microbial degradation. Nevertheless, evolved mechanisms allow many aquatic microbes to utilize these hydrophobic compounds as a source of carbon. Crude oil degrading bacteria produce biosurfactants and bioemulsifiers that permit bacterial adhesion to hydrocarbons and increase aqueous solubility of crude oil components, thus enhancing its biodegradation. Microcosms containing consortia of *Halomonas xianhensis, Halomonas zincidurans, Pseudomonas stutzeri,* and *Halomonas salifodinae*, along with nickel oxide (NiO) nanoparticles and biosurfactants were found to degrade different membered ring polyaromatics, and 90% of the crude oil sample, compared to microcosms with either biosurfactants or nanoparticles (El-Sheshtawy et al. 2017). Table 19.10 lists some of the integrated nano-bio systems used for remediation of petroleum-based hydrocarbons.

Nanoparticle assisted microbial degradation of hydrocarbon plays a significant role in hydrocarbon decontamination. However, many aspects of nanobioremediation need to be studied such as the innate properties of the nanoparticles, their impact on the microorganisms, the reaction conditions and whether sequential or combined treatment systems need to be applied. Also, the effect of biosurfactants and bioemulsifiers on the biodegradation of hydrocarbon contaminated sites needs to be evaluated prior to large-scale application of the integrated system.

19.16 Bioremediation of Organic Solvents and Antibiotics

The organic solvent, trichloroethylene (TCE), is an industrial solvent and is a common soil contaminant which is toxic at low levels. Due to its mobility in groundwater and high density, remediation of TCE can be challenging. Combination of nanoscale zerovalent iron (nZVI) along with bacteria *Dehalococcoides* sp. (Table 19.11) has demonstrated successful dechlorination and degradation of TCE to non-toxic compounds in comparison to controls that either contained the microorganism or the nanometal. It was also reported that the combination enhanced the rate of degradation as the nZVI stimulated bacterial growth by providing the bacteria with hydrogen which was utilized to dehalogenate the chlorinated compound (Xiu et al. 2010).

Antibiotics find their way into soil and aquatic systems due to their incessant use in farming and poultry. Due to their continual input into the environment, antibiotics have been classified as pseudo-persistent organic pollutants (Ma and Zhai 2014). These compounds resist biodegradation due to their antimicrobial properties. While nanoparticles and a few microorganisms are known to individually degrade these compounds to certain extent, integrated remediation can offer a cost-effective and more efficient alternative. A combined nano-bio system using MgO nanoparticles has been reported to accelerate the rate of degradation of the antibiotic Cefdinir, by enhancing the cell membrane permeability of yeast *Candida* sp. (Table 19.11) (Adikesavan and Nilanjana 2016). Table 19.11 also enlists some of the other contaminants that have been studied for their remediation using an integrated system of nanoparticles and microbes. Thus, nanoparticles are known to assist microbial activities, but their effect on microbial reaction rates is yet to be studied.

19.17 Bioremediation of Organohalide Compounds

Organohalide compounds are the most persistent toxic compounds that contaminate the soil and water ecosystems. Under aerobic conditions, nZVI has been reported to enhance the degradation of polybrominated diphenyl ethers (PDBE) such as tetrabromo-diphenyl-ether (BDE-47) by *Pseudomonas stutzeri* with increase in incubation time (Huang et al. 2016). PDBEs such as di-, tri-, and deca-BDEs can be degraded into bromophenols by subjecting it to nZVI followed by biological treatment using *Sphingomonas* sp. PH-07. The combination of *Sphingomonas* sp. PH-07 and nZVI is also effective in degrading tetrachlorodibenzo-p dioxin (TCDD), the most toxic dioxin compound (Kim et al. 2012). Nanoparticles integrated with biosurfactant producing bacteria can potentially remediate PCBs. The sequential treatment of PCBs with Pd/Fe nanoparticles followed by bioremediation with

Table 19.11 Nano-bio integrated systems for remediation of recalcitrant organic pollutants

Nano-bio integrated systems for remediation of recalcitrant organic pollutants			
Nanoparticle	Bioagent	Pollutant	References
Organic solvents			
Fe^0 nanoparticles (ZVI)	*Methanosarcina thermophila*	Carbon tetrachloride and chloroform	Novak et al. 1998
nZVI	*Dehalococcoides* sp.	Trichloroethylene (TCE)	Xiu et al. 2010
Antibiotics			
MgO nanoparticles	*Candida* sp. SMN04	Cefdinir (antibiotic)	Adikesavan and Nilanjana 2016
Organohalide compounds			
nZVI	*Sphingomonas* sp. PH-07	Polybrominated diphenyl ethers (PBDEs)	Kim et al. 2012
Pd/nFe bimetallic nanoparticles	*Sphingomonas wittichii* RW1	2,3,7,8-Tetrachlorodibenzo-p dioxin	Bokare et al. 2012
Carboxymethyl cellulose (CMC)-Pd/nFe bimetallic nanoparticles	*Sphingomonas* sp. NM05	γ-Hexachlorohexane (insecticide)	Singh et al. 2013
Magnetite nanoparticles	*Enterobacter aerogenes*	Organophosphate (pesticide)	Daumann et al. 2014
Pd/nFe bimetallic nanoparticles	*Burkholderia xenovorans*	Polychlorinated biphenyls (PCBs)	Le et al. 2015
Pd^0 nanoparticles	*Shewanella oneidensis* MR-1	Polychlorinated biphenyls (PCBs)	Windt et al. 2005
nZVI	*Dehalobacter* sp., *Sedimentibacter* sp., *Dehalogenimonas* sp.	Chlorinated aliphatic hydrocarbons (CAH)	Koenig et al. 2016
nZVI	*Pseudomonas stutzeri*	Polybrominated diphenyl ethers (PBDEs)	Huang et al. 2016
nZVI	*Bacillus* sp.	Polychlorinated biphenyls (PCBs)	Zhang et al. 2016

biosurfactant producing *Burkholderia xenovorans* resulted in formation of less toxic and harmless compounds (Le et al. 2015). The Bio-Pd-*Shewanella oneidensis* MR-1 system serves as a self-sustaining model for remediation, wherein the bacteria adsorb palladium nanoparticles, reduce it to zerovalent state, and precipitate it. These bio-reduced Pd (bio-Pd) nanoparticles were able to dechlorinate a wide range of chlorinated PCB congeners, without any selectivity (Windt et al. 2005). Culture medium dominated by organochlorine respiring bacteria *Dehalobacter* sp., *Sedimentibacter* sp., and *Dehalogenimonas* sp. when combined with nZVI completely reduce CAH such as dichloroethane and trichloroethane to ethane (Koenig et al. 2016). Carboxymethyl cellulose (CMC) stabilized bimetallic nanoparticles (CMC-Pd/nFe0) integrated with *Sphingomonas* sp. have been demonstrated to degrade γ-hexachlorohexane (γ-HCH), a commonly used insecticide. The stabilized nanoparticles demonstrated to have a biostimulatory effect on the bacterial cells (Singh et al. 2013). Likewise, zerovalent iron (ZVI) has been reported to enhance *Methanosarcina thermophila* mediated dechlorination of compounds such as chloroform and carbon tetrachloride, by serving as an electron donor (Novak et al. 1998). Table 19.11 lists nano-bio systems that are used for remediation of recalcitrant organic compounds.

19.18 Fate of Nanoparticles

While it is implicit that nanoparticles have various applications in the field of bioremediation, the extensive use of these nanoparticles and their subsequent release into the environment is a major concern, especially in cases of in situ remediation. Therefore, the zerovalent iron nanoparticles are currently the only nanoparticles on field application (Kharangate 2015). These particles, once released into the environment, may find channels to enter the food chain, deposit in various organisms, and manifest toxic effects. Nanoparticles have been studied to have toxicity affects that can be attributed to its size. Even nanoparticles of inert metals such as gold have shown to have toxic effects on biological systems (Jeevanandam et al. 2018). Due to the shortage of investigations of the toxicological effects of these nanoparticles, a sub-speciality of nanotechnology, known as nanotoxicology has been developed (Colvin 2003; Niazi and Gu 2009).

Compared to plants, invertebrates, and vertebrates, microorganisms exhibit widespread and diverse reactions to nanoparticles (Oberdorster et al. 2009). The toxic effects depend on the nature of the nanoparticle and its interaction with the microbial species. In case of rhizosphere resident bacteria *Pseudomonas chlororaphis* O6, CuO nanoparticles were demonstrated to neutralize the charge on the EPS colloids and release ions that disrupt the homeostatic control within the cell. Similar mechanisms of disrupting homeostatic mechanisms of the cell have been observed with ZnO nanoparticles (Dimpka et al. 2011).

Application of nanoparticles in remediation processes results in the discharge of these nanoparticles in the soil and aquatic ecosystems wherein they encounter and interact with various microorganisms, which are sensitive to these particles. Due to their nano-size, large surface area, and high reactivity, nanoparticles can destroy the microbial membrane and generate reactive oxygen species (ROS) that are known to disrupt cellular processes (Cecchin et al. 2016). Studies conducted on nanoparticles released in natural aqueous environments confirm that the nanoparticles react and conjugate with biological molecules such as DNA, RNA, proteins as well and lysophospholipids which facilitate their solubilization in water (Ke and Qiao 2007; Niazi and Gu 2009). Since microbes can assimilate nanoparticles, it can lead to accumulation in the food chain and ultimately build-up to toxic levels in humans.

Recovery of released nanoparticles from natural systems seems impossible, extensive research is needed to understand their potential toxic effects on various biological systems, prior to their large-scale application in nanobioremediation of contaminated sites.

Furthermore, soil microorganisms play an important role in regulating biogeochemical cycles and also possess natural mechanisms to degrade contaminants. Drastic reduction in populations of these beneficial organisms can lead to ecological imbalance. Despite their adaptability, nanoparticles used for in situ remediation can have a bactericidal effect on the inherent microbiota. Thus, determining the interactions between native microorganisms, the contaminant and the nanoparticle to be used prior to application is imperative for sustainable remediation of polluted sites.

19.19 Conclusion

Integrated systems have a lot of potential in remediating various organic and inorganic contaminants from polluted environments. Nanobioremediation has the ability to reduce the overall cost for environmental clean-up on large-scale application. While ex situ nanobioremediation is still at an infant stage, in situ application of nanoremediation and nanobioremediation to soil and ground/surface water can reduce contaminant to near-zero levels. Microbe-assisted intracellular/extracellular synthesis of biogenic nanoparticles acts as a novel, environment friendly, cost-effective strategy to supply tailored nanoparticles for remediation projects. Additionally, the possibility of intentionally tuning nanoparticles opens up new avenues in nanoremediation. Although this approach is a favourable alternative to conventional methods, nano-toxicity associated environmental risks are still unclear and need further research. Moreover, the effect of parameters like pH, temperature, ionic strength, presence of competing or inhibitory substances, etc. on remediation efficiency of the nanobioremediation method needs to be evaluated. But once these gaps are filled, integrated nano-bio systems can be applied on a larger scale.

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