



Nanoremediation: Nanomaterials and Nanotechnologies for Environmental Cleanup

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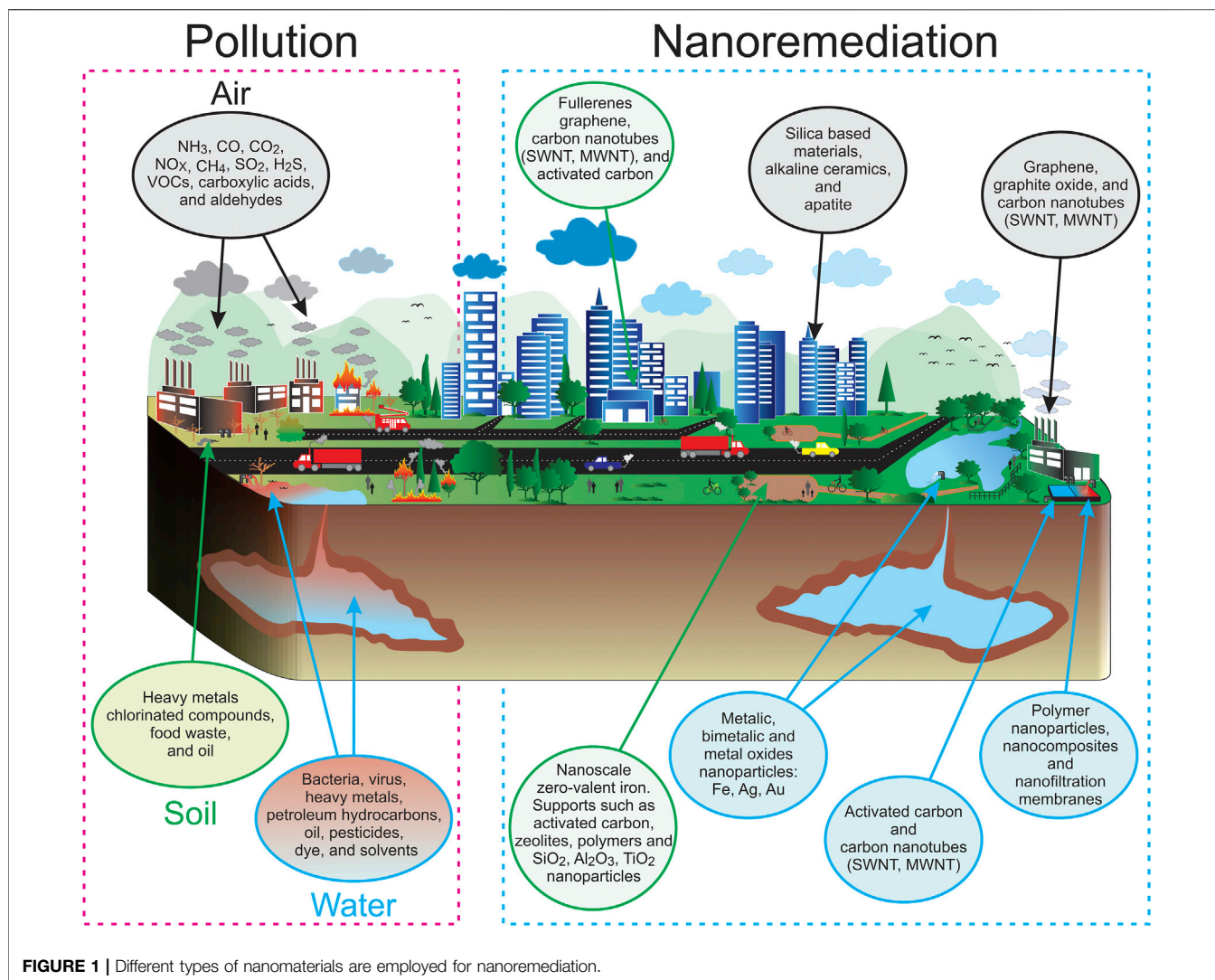
Different global events such as industrial development and the population increment have triggered the presence and persistence of several organic and inorganic contaminants, representing a risk for the environment and human health. Consequently, the search and application of novel technologies for alleviating the challenge of environmental pollution are urgent. Nanotechnology is an emerging science that could be employed in different fields. In particular, *Nanoremediation* is a promising strategy defined as the engineered materials employed to clean up the environment, is an effective, rapid, and efficient technology to deal with persistent compounds such as pesticides, chlorinated solvents, halogenated chemicals, or heavy metals. Furthermore, nanoremediation is a sustainable alternative to eliminate emerging pollutants such as pharmaceuticals or personal care products. Due to the variety of nanomaterials and their versatility, they could be employed in water, soil, or air media. This review provides an overview of the application of nanomaterials for media remediation. It analyzes the state of the art of different nanomaterials such as metal, carbon, polymer, and silica employed for water, soil, and air remediation.

Keywords: nanoremediation, nanomaterials, bioremediation, nanotechnology, environmental ecotoxicity

INTRODUCTION

Contaminated water, soil, and air represent a critical world problem involving extreme environmental and human health risks. Several developed techniques for remediation include conventional methods such as thermal treatment, pump-and-treat, chemical oxidation, and emerging technologies such as “nanoremediation” (Ganie et al., 2021; Mukhopadhyay et al., 2021). Nanoremediation uses engineered nanomaterials to clean up polluted media, and this technique is less costly and more effective than most typical methods.

In addition to its cost-effectiveness, the interest in applying nanomaterials for environmental remediation relies on the nanostructure’s characteristics. Nanoparticles (NPs) present sensitivity, high surface-area to mass ratio, exceptional electronic properties, and catalytic behavior (Corsi et al., 2018). Catalysis and chemical reduction can be regarded as the primary mechanisms for remediation by NPs. Moreover, NPs have been employed in the removal process based on adsorption because NPs present a random distribution of active sites in their high surface area and a wide possibility of coating modifications (Guerra et al., 2018). In addition, NPs can diffuse in small spaces, enhancing



their application in soil and water remediation. Also, membranes based on nanomaterials have been used in water nanofiltration (NF) since the membrane pores potentially retain big components in water effluents. Moreover, the interaction with the membrane selectively separates the more minor compounds. Nanomaterials employed for water, soil, and air remediation include metal oxides, carbon nanotubes, quantum dots, and biopolymers.

This review aims to discuss the applications of different types of nanomaterials in the context of water, soil, and air treatment, presenting current studies and approaches related to nanotechnology application for environmental remediation.

NANOREMEDIATION OF WATER

Over the last decade, the study of nanomaterials for application in water and wastewater treatment has been widely spread (Figure 1). As clean water is fundamental for living organisms to sustain life, contaminated groundwater is a problem that

concerns environmental researchers due to the extreme risks that it represents to different ecosystems (Schweitzer and Noblet 2018). Water sources are susceptible to pollution by many ions, heavy metals, petroleum hydrocarbons, pesticides, radioactive materials, as well as emerging pollutants such as pharmaceuticals and personal care products (Jadhav et al., 2015; Zamora-Ledezma et al., 2021).

In this context, research and development of efficient methods for water remediation are imperative. In recent years, different technologies based on nanomaterials have been employed in the remediation of water due to their properties, including the selectivity to certain pollutants and their absorption capacity (Table 1). The predominant nanomaterials employed in water remediation are metallic nanoparticles, biopolymeric membranes, and carbon-derived materials (Saikia et al., 2019).

Metal and Metal-Based Nanomaterials

Several types of metal oxide nanoparticles such as iron oxide (Fe₂O₃/Fe₃O₄), zinc oxide (ZnO), and titanium dioxide (TiO₂)

TABLE 1 | Advantages of Nanomaterials employed in media remediation.

Type of nanomaterial	Remediation mechanism	Media	Advantages	Limitations/Risks
Metal-based	Adsorption; oxidation; reduction; photodegradation, photocatalysis	Soil, water	<ul style="list-style-type: none"> • High specific surface area • Removal of diverse pollutants (chlorinated organic solvents, polychlorinated biphenyls, organochlorine pesticides) • Compatibility with other treatments 	<ul style="list-style-type: none"> • NPs can have adverse effects on pure cultures of bacteria • Research of the risks in human and environmental health is missing
Carbon-based	Adsorption	Soil, water, air	<ul style="list-style-type: none"> • High surface area • Microporosity • Sorption properties • Eco-friendly nature • Compatibility with other treatments 	<ul style="list-style-type: none"> • Different cell toxicity effects (Reactive oxygen species production, lysosomal and DNA damage) • Rapid saturation • High cost
Polymer-based	Nanofiltration	Water	<ul style="list-style-type: none"> • Employment of polymer derived from waste materials • Compatibility with other treatments 	<ul style="list-style-type: none"> • Denaturation by extreme temperature
Silica-based	Catalysis; adsorption	Air, water	<ul style="list-style-type: none"> • Versatility on surface modification • Adaptable pore size • Compatibility with other treatments 	<ul style="list-style-type: none"> • Performance depends on pH • Scattered size distribution

are utilized for water purification due to their high reactivity, photolytic characteristics as well as adsorbent properties derived from their massive surface area and affinity to different chemical groups (Aragaw et al., 2021). For instance, iron nanoparticles have been employed to treat dyes in wastewater from textile, paint, and paper industries due to their stability in suspension medium and high adsorption capacity. In recent years, these NPs have been highly efficient in the adsorption of dyes such as methyl orange and methylene blue, two of the most utilized dyes in industry, which present the most inharmonious effects on the environment and human health (Mashkoo and Nasar 2020). In this context, the methyl orange and phenol removal efficiency of magnetic iron oxide NPs in combination with carbon has been examined, revealing that the nanocomposites present stronger interactions with the dye, being the carbon concentration a decisive parameter in the NPs adsorbent behavior (Istratie et al., 2019). Besides dyes, heavy metals like chromium (VI) are another critical type of pollutants in water. Current researches suggested that the environmental risk by chromium (VI) could be lessened by the presence of iron oxide or zero-valent iron NPs and organic acids (such as citric acid) (Yang et al., 2017; Zhou et al., 2018). Titanium dioxide NPs are widely employed as photocatalyst for micropollutants removal in water, and it is an effective alternative for emerging contaminants such as pharmaceuticals (Mahmoud et al., 2017).

Carbon-Based Nanomaterials

Nanoporous carbon-based materials such as activated carbons, carbon nanotubes (CNTs), including multi-walled nanotubes (MWCNTs) and single-walled nanotubes (SWCNTs), and graphene and its oxide, present physicochemical characteristics that make them suitable for water treatment operations to remove contaminants like heavy metals, fluorides, textile dyes or

pharmaceutical products. For instance, a study evaluated the adsorption of hexavalent chromium by MWCNTs in contaminated groundwater (Mpouras et al., 2021). The authors analyzed the adsorption efficiency effect of parameters such as pH and adsorbent concentration. Their results suggested that at pH values higher than 7, the adsorption decreased. MWCNTs have also been applied in water gasoline removal projects (Lico et al., 2019). Due to the great environmental concern that represents fluoride, different alternatives based on carbon have been employed to achieve defluoridation of wastewater. In this context, there are reports of the fluoride removal capacity of chemical and bio-reduced graphene oxide, exposing that the first one presented an 87% of reduction; meanwhile, the bio-reduced presented 94% of capacity (Roy et al., 2017). Similarly, activated carbon has been widely explored in removing pharmaceutical products due to their low cost, large pore size, and high porosity. For instance, the comparison of carbamazepine and sildenafil citrate adsorption onto powdered activated carbon and granular activated carbon was reported in 2019 (Delgado et al., 2019). The results revealed that approximately 90% of the compounds were removed in 10 h using powdered activated carbon, whereas the granular activated carbon achieved just 40% of removal after 70 h, which is related to the greater surface area of the powdered. Likewise, the evaluation of caffeine, ibuprofen, and triclosan adsorption employing powdered activated carbon was reported, observing an important effect of pH (Kaur et al., 2018).

Polymer-Based Nanomaterials

Different alternatives based on polymer nanotechnology could be employed in water treatment, such as nanoparticles, nanocomposites, or NF membranes (Abdelbasir and Shalan 2019; Bassyouni et al., 2019). Particularly, polymeric

nanomembranes are employed to eliminate unwanted nanoparticles in the aqueous phase by detouring particles in the membrane pores and by the chemical interaction between the pollutants and the membranes, provoking the pollutant's immobilization. In this context, chitosan is a widely employed polymer for NF membranes elaboration based on facile manufacturing techniques such as solvent casting. These membranes are a strategy to clean textile wastewater (Long et al., 2020), revealing a lower rejection to electroneutral and negatively charged dyes than the positively charged. However, the dyes' physical size also plays a key role in NF efficiency (Weng et al., 2017). The stability and effectiveness of these nanofiltration membranes could be enhanced using the membranes as matrix or support to other types of materials, constituting a composite. Recently, synthetic and natural polymers such as polyamide, cellulose, and chitosan have been employed as membrane matrices and modified by different components such as triethanolamine, metal oxide nanoparticles, and carbon nanotubes (Yan et al., 2016; Lakhota et al., 2018). For example, it has been reported that by employing carboxylated MWCNTs in polyamide membranes, an increment in salt rejection rate can be observed, which is very useful to remove the industrial salts from textile effluents (Al-Hobaib et al., 2017). In addition, polyethersulfone membranes functionalized with MWCNTs, graphene, or other polymers exhibited excellent heavy metals and dyes rejection in aqueous media (Vatsha et al., 2014; Ma et al., 2017; Peydayesh et al., 2020).

NANOREMEDIATION OF SOIL

The settlement of *Homo sapiens* during the transition from hunter-gatherer to farmer resulted in an irreversible impact on nature. The dominance of the wheat business, first as a form of subsistence, later as a style of economic exchange, had consequences in the disappearance of animal species, plants, diversion of river courses, and soil erosion and contamination. Subsequently, the appearance and increase of industrialization and excessive urbanization have accelerated the deterioration and contamination of soil (Kumar et al., 2021). Recently, the use of nanomaterials for the remediation of soil has been attractive due to its high reactivity, high surface-to-volume ratio, surface functionalization, and modification of physical properties such as size, morphology, porosity, and chemical composition. The set of these properties allows the selectivity and efficiency in the capture of pollutants. The intercalation of nanoparticles in the soil allows the cleaning of extensive areas and reduces costs and time due to the application *in situ*. Nanoremediation for soil contamination has predominated with metallic and magnetic nanoparticles, carbon nanotubes, and nanoscale zero-valent iron (Mukhopadhyay et al., 2021).

Metal and Metal-Based Nanomaterials

Nanoscale zero-valent iron (nZVI) is an electron donor with a negative reduction potential. The use of nZVI is one of the most frequent in pilot trials (Cheng et al., 2021) because it allows the removal of chlorinated organic solvents, polychlorinated

biphenyls, and organochlorine pesticides through oxidation-reduction transformation strategies sequestration (Stefaniuk et al., 2016). nZVI has also been shown to be effective in the remediation of trichloroethene, hexavalent chromium, nitrate, lead, cadmium, and DDT with high cleaning percentages (Guerra et al., 2018). There are different nZVI synthesis methods such as carbothermal reduction, ultrasound-assisted, electrochemical, and green synthesis. Although nZVI possesses reactivity as a reducing agent, it lacks agglomeration dispersion stability, difficulty separating it from the remediated soil, and limited mobility. Modifications to the surface are a technological option to preserve its function, and the most frequent strategies include mixing with other noble metals in the form of an alloy such as Pd, Pt, Ag, Cu, and Ni. Other strategies include coating the surface with biopolymers like starch, carboxymethyl cellulose, guar gum, or synthetic polymers like poly (ethylene glycol). While the incorporation of nZVI on the surface of supports such as silica, activated carbon, zeolites, or polymer membranes facilitates the separation of the nanomaterial from the purified soil. Additionally, nZVI can be immobilized utilizing a "trapping" strategy in emulsions or dispersions of particles in biopolymers such as calcium alginate, chitosan, and gum arabic. Other metal-based nanomaterials include applying SiO₂, Al₂O₃, TiO₂, iron phosphate, goethite, and magnetic nanoparticles (Stefaniuk et al., 2016).

Carbon-Based Nanomaterials

Carbonaceous nanomaterials exhibit unique characteristics such as large surface area, high microporosity, excellent sorption capacities, and eco-friendly nature. Some architectures embrace fullerene C₆₀, fullerene C₅₄₀, SWCNTs, MWCNTs, graphene, and activated carbon nanoparticles (Matos et al., 2017; Marcon et al., 2021). Moreover, activation or functionalization of carbon-based nanomaterials represents additional advantages as in other environmental remediation applications. Recently, there has been a greater preference for CNTs because they offer greater adsorption capacity than graphene, graphene oxides, biochar, and granular activated carbon. The adsorption is determined by the exposure area and functional groups on the surface, such as -COOH and -OH. The adsorption capacity can be increased by coupling functional groups such as -NH₂, -SH, oxidation processes, nonmagnetic metal oxide coating, and grafting of magnetic iron oxides. The increase in surface area, high surface-to-volume ratio, and therefore its high reactivity favor flocculation and decrease its properties for nanoremediation. The use of the surfactant poloxamer 407 has allowed an adequate stabilization of multi-walls carbon nanotubes (Matos et al., 2017). CNTs can remove heavy metal ions such as Pb²⁺, Cu²⁺, Ni²⁺, and Zn²⁺; however, the immobilization of heavy metals depends on pH, organic matter content, and the presence of silt and clay particles. CNTs can also remediate the soil of total petroleum hydrocarbons, crude oil, Cr (VI), Cd, DDT, hexachlorocyclohexane, increasing the microbial population and plant growth (Shan et al., 2015). CNTs application techniques comprise their incorporation into membrane filtration, separation columns, and an aqueous dispersion.

NANOREMEDIATION OF GAS PHASE

Air pollution is one of the most significant problems that the world is facing this century since it impacts climate change and public health. The six most common and harmful outdoor air pollutants include particle matter (PM10 and PM2.5), nitrogen oxides (NO_x), sulfur dioxide (SO₂), carbon monoxide (CO), lead, and ground-level ozone, which is formed by chemical reactions between NO_x and volatile organic compounds (VOCs) (Manisalidis et al., 2020). NO_x, SO_x, VOCs, and ammonia (NH₃) are considered secondary particulate matter precursors. Carbon dioxide (CO₂) is not a pollutant; however, it is the most important greenhouse gas emitted through human activities. In order to overcome this problem, several options have been investigated, including the use of graphene oxides (GOs), graphite oxides and CNTs with highly reactive surface sites, and mesoporous silica materials with ordered and tunable porous structure, high surface area, large pore volume and thermal stability (Guerra et al., 2018).

Carbon-Based Materials

The benefits of nanotechnology in air pollution control are remediation and treatment, pollution prevention, and detection and sensing. The surface of graphite oxide is rich in oxygen-containing functional groups, which can be controlled by changing the reaction temperature with the addition of water (Luo et al., 2018). This material has been used for ammonia gas sensors operating at different temperatures (Bannov et al., 2017; Luo et al., 2018). Carbon-based nanomaterials also offer the possibility of combining other types of nanomaterials to form nanocomposites, merging different properties in a single new material (Scida et al., 2011). GO, and zirconium hydroxide/graphene composites (Seredych and Bandosz 2010; Babu et al., 2016) have been applied as an environmental remediation tool through the adsorption of SO₂. GO was also partially reduced via photoreduction under ultraviolet light irradiation and used as a photocatalyst to degrade VOCs (Tai et al., 2019). Furthermore, a GO membrane with a large specific surface area and a continuous pore structure was used to capture PM2.5 (Jung et al., 2018; Zou et al., 2019). There have been numerous studies on CNTs in order to enhance their adsorption properties. CNTs typically must be modified or coated with other reactive materials having appropriate functional groups or charges (Guerra et al., 2018). Modified MWCNTs or SWCNTs have been utilized to detect H₂S and SO₂ (Zhang et al., 2012), CO and NH₃ (Dong et al., 2013), NO₂ and NH₃ (Kim et al., 2016), NO₂ (Park et al., 2019), VOCs (Amade et al., 2014), NO_x and CO₂ (Su et al., 2009).

Silica-Based Materials

Silica-based nanomaterials exhibit high versatility because of their numerous advantageous properties, including wide surface area, adjustable pore size, and easily adaptable surfaces (Shukla et al., 2020). Furthermore, the ability of these nanomaterials for catalysis and adsorption has led to a growing interest in recent years for the remediation of polluted air and the elimination of contaminants in the gas phase (Guerra et al., 2018). The superficial modification of

silica nanomaterials may enhance their physicochemical properties. For example, incorporating hydroxyl groups on the surface of the silica nanomaterials may facilitate some surface phenomena, including gas adsorption and wetting. This approach is effective in designing novel catalysts and adsorbents. One of the first studies analyzing the adsorbent capability of modified mesoporous silica demonstrated that the existence of amine groups on its surface promotes the effective capture of H₂S and CO₂ from natural gas (Huang et al., 2003). According to the authors, the material quickly removed up to 80% of the total H₂S (35 min) and CO₂ (30 min); thus, that material is highly efficient in removing those gases. Similarly, another report revealed that aminosilicates have the potential to eliminate CO₂ from ambient air, which suggests that these materials may help mitigate climate changes (Choi et al., 2011). In addition to CO₂, these amine-modified silicates also effectively eliminate other organic contaminants such as aldehydes and ketones (Nomura and Jones 2013, 2014). Thus, they could be applicable for removing pollutants in an industrial environment. On the other hand, atmospheric contamination by lead (Pb) is an emerging environmental and health problem worldwide, and eliminating Pb from the air represents a challenging question. Concerning this, Yang et al. (Yang et al., 2013) developed silica nanoparticles to tackle this environmental problem. The results demonstrated that their silica nanoparticles could remove atmospheric Pb in polluted air. Therefore, silica-based nanoparticles might represent attractive environmental agents against industrial pollution by Pb and other heavy metals.

CONCLUSION

The high surface-to-volume ratio is the basic strategy offered by nanomaterials to adsorb contaminants. However, the increase in surface area is one of the main disadvantages of nanomaterials, and therefore, the appearance of the flocculation phenomenon and possible particle coalescence. Therefore, a challenge is to find the balance between physical stability and adequate surface activity that favors interaction with pollutants. While stabilization with non-ionic surfactants allows a decrease in flocculation, possibly the addition of functional groups to increase the removal of pollutants such as -COOH and -NH₂ can prevent the agglomeration of nanoparticles under specific pH conditions through a simultaneous mechanism of repulsion of electrical charges. With this proposal, the use of non-ionic surfactants would not be necessary. In addition, another challenge is the complexity of the different media. The formation of a corona can overshadow sophisticated nanomaterials on the nanoparticle's surface with ligands from the contaminated medium; therefore, nanoremediation may be favored with previous cleaning steps.

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Conceptualization, GL-G, JM and MDP-A; investigation, LE-G, JR-G, IGK, and GL-G; writing—original draft preparation,

LE-G, JR-G, MDP-A, and GL-G; writing—review and editing, IGK, MDP-A, and GL-G; visualization, GL-G; supervision, MDP-A, and GL-G; project administration, MDP-A, JM and GL-G.

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