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Nano-bioremediation of heavy metals from environment using a green synthesis approach

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ABSTRACT

The quality of human life is compromised due to the increased concentration of toxic heavy metals in air, water, and soil which is directly interacted with living life. Exceed levels of Cr, Cd, Cu, As, Zn, Pb, and Hg influence the living chain and not only causes human damage but also greatly effects animals, plants, and microorganisms. The consistent increase in drawbacks of traditional methods makes them a poor choice for the remediation of heavy metals. In comparison to that, the use of advanced technology at nano levels gives promising results. Many nanomaterials such as carbon nanotubes, nanofibers, nanoflowers, and nanoadsorbents of different metals such as copper, titanium, zinc, gold, silver, iron, cerium, and manganese use along with different biological materials increase the nano-bioremediation rate in the field of science and pose industrial and environmental applications. Being a cost-effective, eco-friendly, controllable nature of nano-bioremediation technology, they lack background knowledge, and handling at the commercial level. This review highlights different types of nanomaterials, how they are implemented in different application, their green synthesis approach, and the boon and bane of using nano-bioremediation technology in real-time.

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1. INTRODUCTION

The toxic nature of heavy metals Hg, Pb, Cr, Cd, and As become a global environmental issue which exceeds their limit and triggers the greater level of contamination [1]. Contamination of heavy metals increases due to the discharge from chemical intensive industries. Accumulation of heavy metals in human bodies as a result of presence of heavy metals in food chain which may enter due to their aqueous solubility which travels through living organisms [2]. Due to the excessive discharge of heavy metals in the environment, the bioremediation potential of different microbes reduces which limits the functionality of microbes and increases the pollutants' concentration [3]. Many human activities are disrupted by the presence of heavy metals such as an imbalance of ions in the body, increased toxicity, respiratory disturbance, gastrointestinal irritations, and even breakdown of the nervous system [4], [5].

The global requirement for clean water, soil, and air has been increasing by leaps and bounds making it impossible for industries to cope up with a better and more effective treatment of these environmental issues caused by heavy metals. There are different conventional methods present which were designed by different industries but these methods become less active and time costly and these physiochemical methods are chemical precipitation of heavy metals, reduction in less toxic form, electrodialysis of heavy metals into ions adsorption by different adsorbents [6], [7]. Other remediation process is also present such as membrane infiltration, leaching, electro-kinetics procedures, and solvent extraction though commonly used but have certain limitation most probably the extravagance of these methods which sidetrack the industries from adopting this remediation process [8].

Microbes and many plants play a significant role in remediation processes. Such as Bacillus sp. helps to improve the removal of different toxic compounds and heavy metals in association with phytoremediation as they secrete different enzymes which help them to rise in contaminated areas [9]. Bacteria use both passive (biosorption) and active (bioaccumulation) mechanisms for cost-effective remediation, using less energy consumption along with better air quality [10], [11]. Plants use different organs and parts of itself for bioremediation and one of them is the use of leaves for phytovolatilization of heavy metals which convert them into volatile and less toxic forms in the environment [12]. A combination of bacteria and plants is also used for the remediation of heavy metals such as lead which help plants to grow in a contaminated area [13]. Different bio-adsorbents are also used for enhancing the removal of heavy metals from the environment such as sawdust, sugarcane bagasse, and green tea leaves which is a cost-effective remediation strategy[14].

Nanoparticles are considered to be excellent adsorbent due to its various characteristics such as its size, surface area, catalytic activity, solubility, and surface of chemical composition [15]. Besides its positive impact, nanoparticles also have some drawbacks like nanotoxicity and nano-nutrition to some extent toward human life which in turn effect the whole process of bioremediation but there is no reconcilable conformation for using nanoparticles with bioremediation technologies [16], hence the main aim of this review article is to scrutinize the close relation of bioremediation with nanotechnology which is used for the enhancement of remediation process of heavy metals and its interlinkage toward the environment.

2. NANOMATERIALS

Materials that are classified as 1-100 nm in size are considered nanomaterials. They confine exclusively visible properties in various applications due to its size such as in biomedicine, synergistic, and boosting of chemical reactions [17]. The structural properties of nanoparticles such as high adsorption capacity and selectivity make them unique to be used for the elimination of contaminants from wastewater at low concentrations [18]. Nanoparticles offer atomic-level modification which opens up many further novel approaches that are not obtained from bulk materials [19].

2.1. Categorization of nanomaterials

2.1.1. Silver based nanomaterial

Metallic materials have the potential to remove heavy metals from contaminated water. Various studies and reports suggested that Ag based materials can be used for the remediation of heavy metals as they have the ability to take up the contaminant and removed it. With the addition of biological material such as leaf extract of *Ficus benjamina* zero-valent Ag nanoparticles are made which have the capacity to remove cadmium as reported by [20]. Al-Qahtani [21] discussed that when the concentration of nanoparticles increases the removal efficiency also increases directly. Malini *et al*. [22] reported that nanoparticles can be encapsulated by additional materials. Green synthesis-based nanoparticles using leaf extract of *Prosopis chilensis* are encapsulated using chitosan as this shows 81% removal of copper ions.

2.1.2. Gold based nanomaterial

Functionalized AuNPs have a unique sensor property due to its high stability, morphology, and selectivity. Due to this sensor property, it is considered to be good material for sensing heavy metals and their remediation. Nitti [23] discussed in his studies that due to the high selectivity property of gold it is considered excellent removal material. Also, Lin *et al*. [24] reported that 97% removal of mercury species is done by gold-based nanomaterial such as AuNP-Al_3O_3 which is cost-effective and stable method.

2.1.3. Iron based nanomaterial

The absorption capacity of iron is high and due to this property Parvin *et al*. [25] stated that as compared to others, iron nanomaterials show higher absorption affinity which is 46 times higher than any other nanomaterial. Members of iron family such as maghemite, magnetite, ochre, rust, and sesquioxide also exhibit different sorption-affinity. Tahar *et al*. [26] reported in his studies that if compared between maghemite and magnetite, maghemite nanoparticles have the higher remediation ability of chromium in an aqueous solution than derivatives of magnetite nanoparticles. The co-precipitation method of iron ions adds a unique property to nanoparticles, this method made metals separate from water easily. Zhang *et al*. [27] justified this statement by reporting in his studies that nanoparticles showed high absorption capacity when prepared with the co-precipitation method.

2.1.4. Copper based nanomaterial

Copper has the potential of high porosity and high surface area. The main property of copper is its high surface area as discussed by [28] in his studies that due to the high absorbency and surface area of nano-flowers of copper, high removal percentage of lead was observed in contaminated water. A combination of different materials with copper is also used for remediation purposes. A study reported by Bazana *et al*. [29] discussed that Fe and Pb were removed by a combination of copper oxides with silver oxides and have high removal efficiency also this study proves the high surface area property of copper as it was seen that removal capacity of copper increases with the increase in the number of copper oxide nanoparticles.

2.1.5. Cerium based nanomaterial

CeO_2 based nanoparticles are usually used to remediate Pb (II), Cd (II), Cr (VI), and As (V) ions from contaminated water. A study reported that cerium-based nanomaterial can be used more readily when compared with iron oxide and titanium oxide-based nanomaterial because CeO_2 has a high level of Pb (II) elimination capacity but also show phytotoxicity which is its drawback whereas other iron and titanium have no toxicity toward anything [30]. Cerium-based nanomaterials can also be affected by pH ranges. Contreras *et al*. [31] reported that when cerium-based nanoparticles were used in an aqueous solution then its adsorption capacity was affected for the treatment of chromium and cadmium whereas unaffected in the case of lead treatment.

2.1.6. Aluminum based nanomaterial

Aluminum based nanoparticles have high electronegativity and also show hydrolysis properties due to this [32] suggested that alumina-based nanoparticles can be used for lead removal, and many forms of aluminum-based nanomaterial are used as adsorbent for heavy metal removal. Hossein *et al*. [33] reported that alumina nanotube when wrapped with polyol-y-sineresorcinol showed high cadmium removal capacity as compared with non-wrapped alumina nanoparticles. A study reported that gamma-Al_2O_3 nanoparticles were reusable for 3 times when treated for cadmium and lead [34].

2.1.7. Titanium based nanomaterial

Many industrial products use titanium oxide due to its stability and safety in nature. It is also used for heavy metal (lead, copper, and arsenic in the metastable form) remediation purposes. Titanium nanoparticles activity is also greatly affected by pH. An increase in pH increases the adsorption capacity of titanium oxide nanoparticles for the removal of lead and copper [35]. A combination of nanomaterials can be used such as ZnO and TiO_2 which increases the surface area for adsorption and also this micro-sized structure can be reusable for up to 3 times [36].

2.1.8. Manganese based nanomaterial

A combination of manganese-based nanomaterials is used and shows the removal efficiency of arsenic metal ions. The mixture of iron with manganese has the highest chromium adsorption capacity from wastewater. MnO also showed the elimination of arsenic from contaminated water [37]. Magnetic nanohybrids of Fe and Mn were also effectively used for the remediation of arsenic from water [38].

2.2. Application of nanomaterials

2.2.1. Imaging and biomedical imaging

Nanoparticles have the ability to modify its surface area according to preferences. It also gains popularity in the medical field due to refined optical capacities and as a carrier for medical delivery purposes which make them an acceptable choice for bioimaging [39]. ZnO nanoparticles show unique green luminescence properties while imaging as it is near the ultraviolet emission scale [40]. Characteristics features of silica nanoparticles such as its high hydrophilicity, solderability, and nano-composition make it possible to use it for biomedical imaging [41].

2.2.2. Drug and gene delivery methods

Due to the small size and easy preparation of nanoparticles, they are used for specific target drug delivery and gene therapy. Many biomedical applications now use nanoparticles to achieve different purposes. Silver nanoparticles, zinc oxide, iron oxide, and different silica-based nanoparticles are used for drug delivery [42]. The method of specific targeted delivery shows less toxicity, cost, and dosage which is a significant achievement of the pharmaceutical industry. Pathogenic treatment can also be done by using nanoparticles such as malarial pathogenic infection can be treated by silver nanoparticles [43]. Due to the vast application of silver nanoparticles, it is now used for dental processing to avoid microbial infections in teeth and gums [44]. Gene therapy is a crucial process and minor errors can cause the degradation of deoxyribonucleic acid (DNA) and thus failure of gene delivery. Among others, ZnO nanoparticles show reliable gene delivery applications which can protect the DNA from degradation and provide a better environment for the uptake of DNA [40].

2.2.3. Antimicrobial properties of nanoparticles
Increased resistance of microbial population hinders many biological functions. Resistance is now overcome by the use of nanoparticles as they incorporate into bacterial cells and damage the cell by converting into silver ions [45]. Nanoparticles are efficient tools as antiviral, antibacterial, and antifungal. A coliform bacterium such as *Escherichia coli* is highly exposed to silver nanoparticles [46]. Like silver nanoparticles, zinc oxide nanoparticles also show a photocatalytic effect which is used for antimicrobial purposes. Due to nano-sized nature of nanomaterials they can easily penetrate the microbial cell and damages the internal structure of cells. ZnO nanomaterials also regarded as bio-safe [47].

2.2.4. Industrial impetus
Industrial products are now fabricated by using nanoparticles to increase the yield of products. Due to the antibacterial properties of nanoparticles, they can be used in textile industries in which nanoparticles are fabricated on fabric to avoid bacterial infection and the texture of the product. Silver nanoparticles show antibacterial activity against *Escherichia coli* so it is used in the textile industry on different products [46], [48]. Other nanoparticles such as titanium and zinc oxide are also used in the textile industry because of several properties such as self-cleaning, ultraviolet (UV) absorber, and antibacterial. These are used for clean and good-quality fabrication but to some extent because due to nano-sized nature of NPs they can agglomerate and hinders their properties [49].

Industries also developing eco-friendly methods for treating contaminated water as it is the main issue of concern due to the increased contamination of water [50]. Due to many drawbacks of traditional methods complete removal of contaminants is impossible [51]. Now this problem can be resolved by the use of nanoparticles because they have the potential to remediate contaminants from water [52].

Food additive is a main accomplishment in the food industry to increase food storage, quality, and taste. NPs play a significant role in the food industry as it provides a good kind of whiteness to food, slipperiness, and have a better refractive index when used with food. TiO_2 NPs mainly used in sweets, candies, and gums due to its good anti-bacterial nature [53]. UV radiation affects the quality of food so using nanoparticles that protect food from UV could be a great source in the food industry such as ZnO nanoparticles.

2.2.5. Waste water treatment
Concerning the main issues in the environment, contamination of water becomes an issue due to a lack of traditional methods of decontamination toxicity of contaminants increases day by day. Traditional methods require time and involved chemical interactions it is a limited way to reduce water contamination [50]. A green synthesis method of producing nanoparticles approaches a new advancement in science. Due to the cost-effective and easy-to-use nature of green synthesis nanoparticles could be a great source for decontamination. Extracts from green tea and eucalyptus tree are used for the preparation of nanoparticles which shows activity against many dyes such as malachite green, orange II and even reducing hexavalent chromium which dissolved in water and make contaminated [54]. Das *et al*. [52] used *Madhuca longifolia* plant and prepared CuO nanoparticles and used them for treating water containing methylene blue dye. Nanoparticles act as an alternative and eco-friendly method to utilize at water contamination sites.

3. GREEN SYNTHESIS APPROACH OF NANOMATERIALS
Besides chemical processes, nanoparticles can be synthesized from different biological materials such as microorganisms and plants. Both are environmentally friendly and biodegradable in nature. Positive results were achieved by using biological components with nanomaterials for bioremediation processes and also their synthesis.

3.1. Use of microorganisms
Microorganisms have bioaccumulation and bio-adsorption ability for different toxic heavy metals. They use different procedures to remove contamination from the environment. Microorganisms usually use the reductase mediate process for the reduction of many heavy metals [55]. Environmental conditions affect the formation of nanoparticles and their activity as different microorganisms possess different growth conditions.

3.1.1. Bacteria
Rafique *et al*. [56] studied *Escherichia coli* for the synthesis of a silver nanomaterial as it has the ability to reduce silver ions within minutes. Gold nanoparticles were first synthesized from *Bacillus subtilis* 168 strain and held success in the gold nanoparticles field [57]. *Pseudomonas stutzeri*, *Staphylococcus aureus* and *Bacillus licheniformis* were used for the well-defined composition and shaped silver nanoparticles [58]–[60]. Chidambaram *et al*. [61] studied on the preparation of Pd nanoparticles using *Clostridium pasteurianum*

along with Pd (II) ions which were positively used for the bioremediation of hexavalent chromium into insoluble chromium (III) and showed 70% promising result. Removal of Cadmium was also possible by green synthesized cadmium sulfide nanoparticles from *Pseudomonas aeruginosa* [62].

3.1.2. Algae

Algae also participated in the production of green synthesized nanoparticles and according to the study reported by [63], a comparison analysis was done between iron nanoparticles from *Chlorococcum* algal species and bulk iron from chemical synthesis. They stated that 92% Cr removal potential was observed by green synthesized iron nanoparticles as compared to others which showed just a 20% Cr removal rate. ZnO nanoparticles were also prepared from green algae as they have a strong potential as a reducing and capping agent and are used in bioremediation strategies [64].

3.1.3. Fungi

Fungi show more tolerance towards heavy metals as compared to bacteria which makes them a better choice for the synthesis of nanoparticles. Fungi grow in bulk form thus a good choice for producing an economical quantity level of nanoparticles [65]. The presence of intracellular enzymes shows a greater amount of bioaccumulation and tolerance and proteins in fungi and due to this the down streaming process is simple and has the ability to produce a bulk number of nanoparticles [66]. Gold nanoparticles were also synthesized by using soil fungus *Aspergillus japonicus* AJP01 as it has the ability to produce eco-friendly and immobilized AuNPs [67].

3.2. Use of plants components

Plants have ability to grow in metal stress conditions and also accumulate different metal ions in different parts of plants such as roots, shoots, leaves, and stems. Nanoparticles prepared from plants are more stable due to the presence of different secondary metabolites and biochemicals in different parts of plants. Nanoparticles prepared from plant extracts are extracellular nanoparticles, intracellular nanoparticles, and phytochemical nanoparticles [45]. Many nanoparticles are synthesized by leaves of plants such as *Camellia sinensis* (green tea) leaves, extracts of *Citrus limon*, and extracts of *Mangifera indica* (mango) [68]. Hossain *et al.* [69] studied zinc oxide nanoparticles and prepared them from the extracts of *Citrus limon*. Al-Senani and Al-Kadhi [70] studied the preparation of green synthesized AgNPs from *Convolvulus arvensis* leaves extract and used for the adsorption of copper which showed 98.99% removal of copper ions. Plants show high adsorption capacity and due to this property, [71] prepared silver nanoparticles from five different plant extracts such as *Camellia sinensis*, *Quercus virginiana*, *Punica granatum*, *Eucalyptus globulus*, and green tea leaves which showed remarkable removal efficacy against arsenic.

4. BENEFITS AND DETRIMENTS OF NANOTECHNOLOGY IN THE BIOREMEDIATION FIELD

The green synthesis approach as discussed earlier is easy to apply, cost-effective, less laborious, environmentally friendly, shows high efficacy, and has a potential interaction with metal ions. Due to these properties, green synthesized nanoparticles are a more favorable choice in different applications of environmental science [72]. Nanotechnology aids improvement in of existing technologies and enhances the efficacy of bioremediation process. Already resistant biological species when used for synthesizing nanoparticles the chances of pollution rate decreases. By using bacteria, fungi, algae, and plants in nanotechnology, direct detoxification of toxic metals occurs but also no secondary waste products which make this technology more feasible in the field of science [73]. Microbe-based nanomaterials provide a better down-streaming process also microbial growth is controllable and highly achievable in less time so a green synthesis approach is a better way to get a clean, safe and biodegradable environment in no time [74].

Besides the beneficial nature of nanotechnology, many drawbacks are observed in real-time. One of the biggest drawbacks of nanotechnology is the commercialization of nanomaterials at a larger scale. Scale-up production of green synthesized nanotechnology requires the long-term stability of capping and reducing agents which are biological materials[75] and reusability of nanomaterials which after a few trials shows less efficacy [76]. Synthesis of nanomaterials also requires other chemical reagents and some green synthesis process require high temperature for the production of nanomaterials, lack of guidance in the production of nanomaterials as compared with chemical and physical synthesis, different extracts produce different size of nanoparticles which make them not suitable at large scale-up production also the low yield rate of green synthesized nanoparticles cause hinders in field of nanobiotechnology [77].

5. CHALLENGES AND FUTURE OUTCOMES

For more stable and pollution free environment, industries should more focus on greener and eco-friendly methods of remediation. By controlling the size and maintain monodisperse form of green synthesized nanomaterials and increase in flexibility opens doors for new nanotechnologies. By knowing the right mechanism behind the biogenic synthesis of nanomaterials and controlling the predictable synthesis the advancement increases. An understanding of the functionality of capping and reducing agents will help in gaining a high level of achievement in nano-field. Maintaining the growth rate and incubation time will speed up the actions of nanomaterials. It is highly recommended that if we study from production to disposal level, it is possible that nanotechnology will gain more potential in development. All of this requires nurturance from researchers and scientists to empower cost-effective and eco-friendly nanotechnology.

6. CONCLUSION

Nanotechnology is a rapid growth area due to the production and use of nanomaterials in different fields of environmental and biomedical science. This paper reviewed various nanomaterial, their synthesis from different biological materials, and their uses in different areas especially bioremediation of heavy metals. Ag, Au, TiO_2, ZnO, and iron nanoparticles have a great scope in the bioremediation of heavy metals. Microbes and plants are more applicable and cost-effective as they offer environmental advantages and high efficacy for the degradation of contaminants. By improving the stability, reusability, and rate of production of nanoparticles, nanotechnology will gain real-time large-scale commercialization at a bigger level.

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