Nature Environment and Pollution Technology
An International Quarterly Scientific Journal
p-ISSN: 0972-6268 (Print copies up to 2016)
e-ISSN: 2395-3454
Vol. 21 No. 1 pp. 321-330 2022
Original Research Paper
https://doi.org/10.46488/NEPT.2022.v21i01.039
Open Access Journal

Evolution of Technologies for Cadmium Remediation and Detoxification

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Nat. Env. & Poll. Tech.
Website: www.neptjournal.com

Received: 18-06-2021
Revised: 10-08-2021
Accepted: 20-08-2021

Key Words:
Cadmium remediation
Vitrification
Phytoremediation
Nanocomposites
Iron nanoparticles

ABSTRACT

Heavy metal pollution is one of the most serious global environmental concerns. As a result, the current research includes an overview of technologies that are being developed for remediating or eliminating such contaminants from the environment, such as physical and chemical approaches, as well as their ineffectiveness. A wide range of minute species was discovered for their potential to tolerate, resist, accumulate and absorb heavy metals. But they all are naturally occurring species and need optimal conditions as well as a longer duration to grow. Thus, there is a need for more reliable, efficient, and productive techniques to address the issue. The use of nanoparticles for remediation has paved the way for more research in this subject and the development of useful technology to cope with problems. The evolution of technologies for heavy metal remediation, particularly cadmium, is discussed in this article because it is one of the most hazardous heavy metals that necessitates immediate attention.

INTRODUCTION

The expeditious urbanization and industrialization undoubtedly led to exceptional growth but also has directly affected the environment. As a result, the contamination and degradation of the whole ecosystem have become a major problem and a threat to all life forms especially to human beings (EPA 1990). The research in this field has also gained its speed, particularly in the area of risk associated with heavy metal dispersal in the food chain and their remedies. Heavy metals can be derived from multiple sources, including industrial effluents and mining sectors. As a result, researchers have developed a keen interest in recovering heavy metals from effluents and soil. Cadmium is used in many industries. Uses of cadmium are shown in Fig. 1.

Many clean-up techniques were proposed and practiced by many researchers which mainly include reverse osmosis and active sludge treatments. But these techniques also have many disadvantages like reduction in biodiversity and inhibitory effects on plant growth (Chaudhury et al.1999). Researchers have a lot of options when it comes to using microorganisms to change or convert heavy metal toxins into a less harmful state. Bioremediation has a few drawbacks, the most significant of which is the presence of toxic microbial metabolites that might interfere with the entire remediation process, resulting in heavy metal non-degradation. However, as compared to alternative physical and chemical processes, this technology is a better option in terms of environmental friendliness and economic effectiveness.

HEAVY METALS SOURCES IN THE ENVIRONMENT

Natural and Anthropogenic Sources

Heavy metals have their origins in rocks. Heavy metals were absorbed into rocks as a result of physical damage caused by bad weather. They are found in soils as metal sulfides in combination with copper and iron sulfides. Similarly, cadmium is mostly created as a by-product of the zinc refining process, as it is with the zinc ore sphalerite. As a result, cadmium is found in water, soil, and the air. Heavy metals make their way into sewage and reservoirs from the surface of the soil.

There are many human-induced sources of heavy metals which mainly include: sewage sludge, paper industries, pesticides, batteries, tanneries, fertilizer industries, wastewater irrigation. Cadmium is mainly present in phosphate fertilizers. The use of phosphate fertilizers in fields led to contamination of water and soil with cadmium which ultimately takes Cd into the food chain. The sources of cadmium are shown in Fig. 2.

Human Cadmium Exposure and Its Toxicity

Prolonged exposure to cadmium through water, food, and soil is very toxic which leads to cancer. Humans are exposed to cadmium mainly through ingestion or inhalation. High cadmium levels can be measured in different body parts like hair, nails, urine, saliva, and blood. The major sources of cadmium toxicity are contaminated food, beverages,

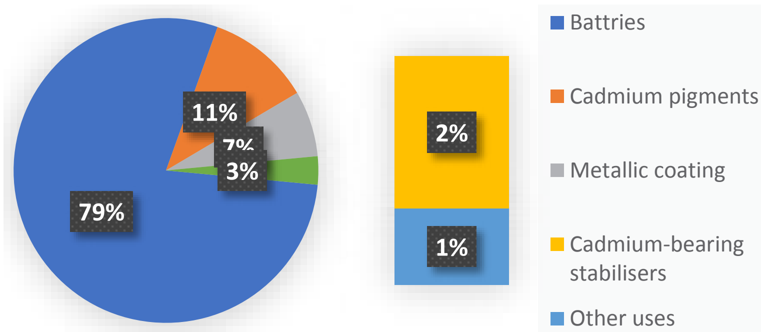

Fig. 1: Cadmium uses in industries (Sharma et al. 2015).

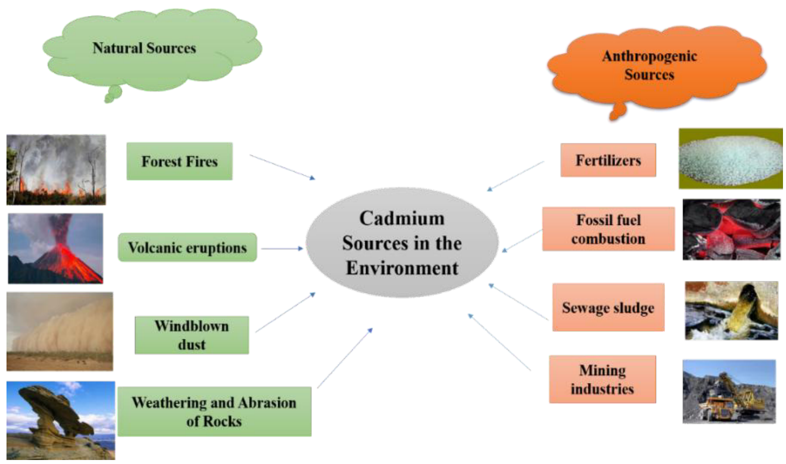

Fig. 2: Natural and anthropogenic sources of cadmium (Thornton 1992).

cigarette smoking (Friberg 1983), and welding purposes. The cadmium can be detoxified with the help of EDTA and other chelates. Antidotes with nanoparticles are required for patients with cadmium toxicity. Cadmium exposure to humans is depicted in Fig. 3.

The toxic nature of cadmium imposes harmful effects on human health. It mainly targets the kidney, bones, and reproduction system. Toxicity and health effects are depicted in Fig.4.

Technologies for Remediation and Detoxification

Physical Methods of Heavy Metal Remediation

This technique of remediation mainly involves the following methods:

Soil isolation: The onsite remediation of heavy metals is accomplished by soil isolation. It essentially entails the separation of polluted and uncontaminated soil. However, this technique is not comprehensive; several other engineering processes are needed. This technique restricts the other contaminates in a specified area. This method is used only when other methods are not economically feasible. Subsurface barriers are used in this technique to limit surface water and groundwater flow and to extract contaminated water from the soil. These barriers are used to keep uncontaminated water from flowing into contaminated water (Dawson 1996). Examples of subsurface barriers are grout curtains, sheet piles, and slurry walls. The barriers can be employed surrounding the contaminated site, downstream or upstream. To prevent the infiltration of contaminated water, these barriers are used in combination with the capping system to maintain the continuity of isolation of contaminated soil. Clay can be used underneath the contaminated soil.

Soil replacement: Before 1984, the soil replacement off-site removal method was commonly used for the removal of various contaminants from soil. In this method, the contaminated soil is replaced from the uncontaminated soil. This technique enhances soil efficiency by diluting the concentration of heavy metals. It can be done in two ways- 1. Soil spading 2. Importing new soil to the site. In spading technique, the soil is dug thoroughly and heavy metals are spread there. But in the 2nd method, the new clean soil is imported to the

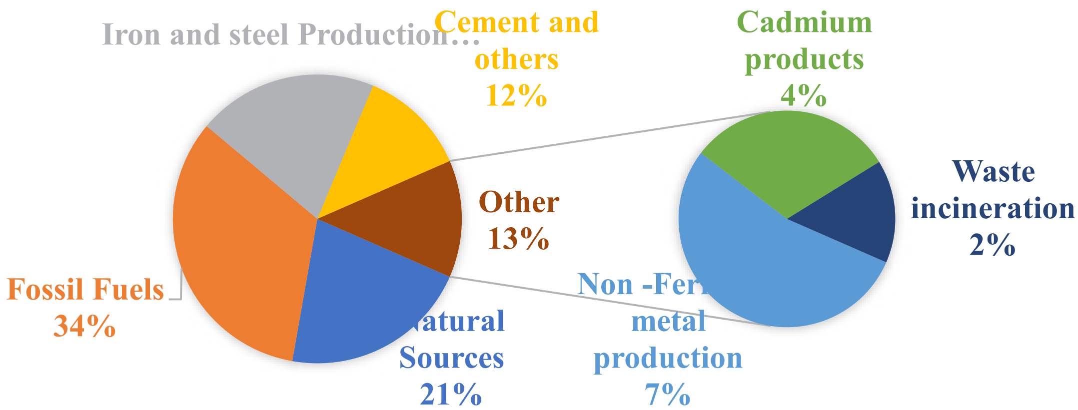

Fig. 3: Human cadmium exposure (Jaishankar et al. 2014).

contaminated site. The purpose of both methods was simply to dilute the concentration of heavy metal at the contaminated site. The main disadvantage of the soil replacement method is high labor work. But it is appropriate and efficient for removing contaminants from the soil in a small area.

Electrokinetic remediation: This remediation method involves the pouring of contaminated soil into the electrolytic tank and then applying an electric field gradient of suitable intensity. The principle behind this process is electro-migration and electrophoresis. A suitable electrolyte is required to efficiently remove the contaminated soil. The efficiency also depends upon the metal to be remediated. Ethylenediamine disuccinate (EDDS) was used to decontaminate cadmium and lead (Suzuki et al. 2014). This method is very easy to install and perform which makes it economical in nature. Also, it does not change the basic nature of the soil. The maintenance of soil pH is the limiting factor as pH cannot be easily maintained.

Vitrification: To restrict the mobility of heavy metals, a vitreous material can be produced inside the soil by applying high temperatures (Mallampati et al. 2015). This technique is easy to apply than other classical and physical remediation techniques. Both organic and inorganic contaminants can be remediated with the help of this technique. Vitrification is conducted in two different means, in-situ and ex-situ. In the in-situ technique, the electric current is provided by using an array of electrodes inside the soil. Also, in-situ is more favorable as compared to ex-situ due to its less energy requirements and low cost. Ex-situ technique involves many stages like excavation, mixing, pretreatment, melting, feeding, and casting of the melted product (Dellisanti et al. 2009). The efficiency of the technique can be enhanced by mixing additives. This technique is mainly suitable for pollution of large scale.

Chemical Methods of Heavy Metal Remediation

This technique of remediation uses reagents and chemicals to remediate heavy metals. It mainly involves the following methods:

Soil washing: In this technique, the heavy metals are leached out of the soil by using chemicals and reagents. Chelators

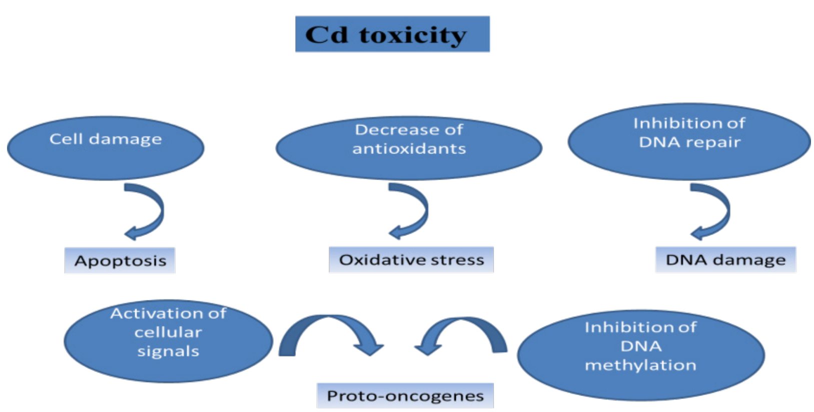

Fig. 4: Cadmium toxicity in human beings (Rafati et al.2017).

like EDTA are added to immobilize toxic elements in a less bioavailable form. During this technique, the contaminated part of the soil is dug out and then treated with the appropriate extractants. The choice of extractant depends upon the soil and the heavy metal to be extracted out. Both soil and extractant are then mixed thoroughly. Furthermore, the soil is transferred to the liquid phase through techniques like precipitation, absorption or chelation, etc. Soil washing is a widely used method of remediation of heavy metals due to its quality of completely removing the toxins and speedy performance. A huge number of reagents and chemicals are used such as EDTA, organic acids, cyclodextrins, and surfactants. Coal ash and EDTA have become popular materials for the extraction of heavy metals. The soil wash method was developed for cadmium-contaminated paddy fields (Kimura et al. 2007).

Immobilization techniques: It involves confining heavy metals in soil by using immobilizing agents. Many methods have been employed for the immobilization process such as adsorption, precipitation, and complexation. Organic and inorganic agents can be used for immobilizing heavy metals in contaminated soil. It mainly includes clay, zeolites, minerals, cement, etc. Heavy metals can be immobilized on solid particles to reduce their availability in soil. Organic agents and organic amendments are widely used which mainly includes biosolids and animal manures. Negative effects of biosolid application in the soil were reported (Cele et al. 2016). However, there are positive effects too as it is the best adsorbent for heavy metal stabilization in the soil (Venegas et al. 2015). Remediation methods are shown in Fig. 5.

Biological Methods of Heavy Metal Remediation

Biological methods mainly include approaches like bioremediation which uses living microorganisms as well as plants to remove pollutants heavy metals from the environment. Bioremediation itself has two approaches namely in-situ and ex-situ. In-situ is the removal of heavy metals on the contaminated site and ex-situ is treating them somewhere else. Both the approaches are being summarized in Table 1.

Table 1: Approaches of bioremediation.

Technology	Benefits	Limitations
In-situ	Cost-effective Treats both water and soil Can easily deal with dissolved and sorbed pollutants Minimal site disruption	Long treatment time Difficulty in the monitoring process Toxicity
Ex-situ	Low cost Can be done on-site pH can be controlled	Space requirements Limited groundwater depth (90-300 cm) Mass transfer problems

Many microorganisms and plants are reported to date for effective removal of cadmium from contaminated sites. A few recent studies have been mentioned below in Table 2.

Nanomaterials for Heavy Metal Remediation

Out of these methods, the biological method of wastewater treatment is widely accepted as they use efficient microorganisms but they are often slow and expensive. As a result, there is a pressing need for practical methods for long-term water management to achieve water security. Nanotechnology's progress has been noted recently, and its potential for removing pollutants from wastewater has been demonstrated. Nanotechnology uses nanoparticles with a size of a few nanometers to treat groundwater, surface water, drinking water, and industrial effluents, providing a new way to eliminate toxins from wastewater and lessen its impact on people. For the removal of heavy metals, various nanomaterials have been developed to date. A few of their types are presented in Fig. 6.

The attractive features of nanomaterials are high heavy metal removal rates, also they are cost-effective and easy to regenerate. With this context, a few newer nanotechnologies are discussed below

Carbon-based nano adsorbents: The main process by which the removal of heavy metals is achieved is adsorption. Carbon nanotubes are the most popular carbonaceous nanomaterials because of their high adsorption rates, mechanical strength, and chemical resistivity (Lee et al. 2012). The hydrophobicity of CNT graphite surfaces causes them to aggregate. The presence of grooves and interstitial spaces in the aggregates capture the organic molecules of wastewater through covalent bonding, hydrogen bonding, and electrostatic interactions (Rao et al. 2007). Due to electrostatic interactions and chemical bonding between surface functional groups, large metals or organic molecules adsorb onto the

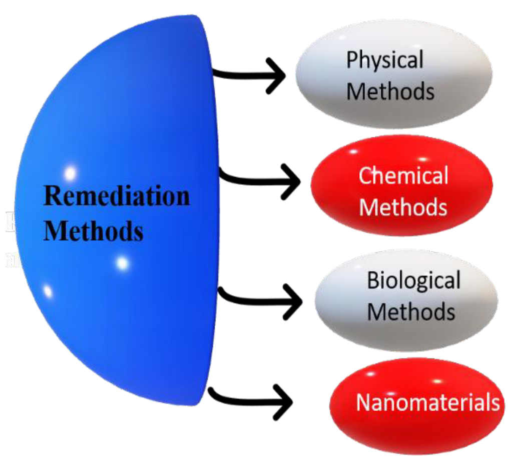

Fig. 5: Remediation methods

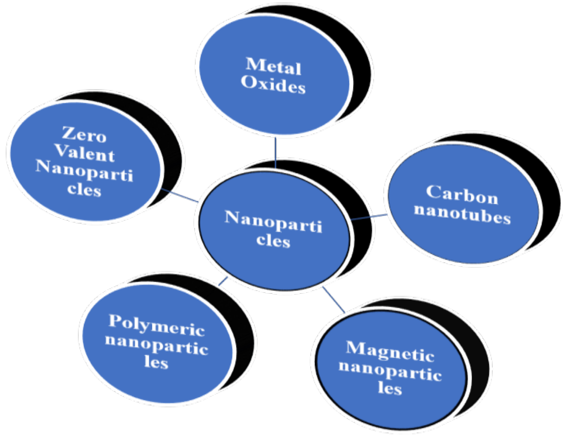

Fig. 6: Types of nanoparticles.

pores and become stuck on CNTs (Zhang et al. 2010). CNTs are of two types as depicted in Fig. 7 given below:

a. Single-walled carbon nanotubes (SWCNT): SWCNTs are constructed using a hollow tube having one atom thick wall made of a single layer of graphene, called one-dimensional carbon nanomaterials. 1-D SWCNT has a unique structure with porosity and high surface area. Many successful studies are being conducted by using SWCNTs for wastewater treatment. Adsorption of cadmium, copper, lead, and mercury was investigated using SWCNTs-COOH, SWCNTs-OH, and SWCNTs-NH_2 and it was observed that SWCNTs-COOH was having high adsorption capacity (150-230%) as that of others stated earlier (Anitha et al. 2015). SWCNTs nanocomposite designed with polysulfone showed 94% removal of extremely toxic heavy metals i.e., lead (Gupta et al. 2015). Thus, the above studies revealed that carbon nanotubes are promising tools for the removal of heavy metals from contaminated wastewater.

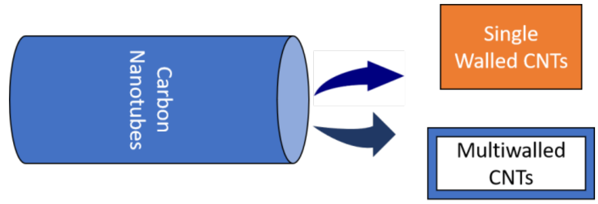

Fig: 7: Types of carbon nanotubes.

b. Multiwall carbon nanotubes (MWCNTs): MWCNTs have multiple layers of graphene. Numerous nanocomposites of MWCNTs were designed to remove heavy metals. Heavy metals such as As, Cr, Pb, Ni, and Cu were successfully conducted by using MWCNTs-Fe_2O_3, MWCNTs-MnO_2-Fe_2O_3, MWCNTs-Al_2O_3, MWCNTs-ZrO_2, and MWCNTs-Fe_3O_4 nanocomposites (Yang et al. 2009). MWCNTs which were oxidized with acid (Chemically) were studied for sorption of lead, cadmium, and chromium (Moosa et al. 2015). Plasma oxidized MWCNTs are known for having more sorption efficiency than chemically oxidized as they have more oxygenated functional groups (Li et al. 2009). Additionally, plasma-oxidized nanotubes are recyclable and reusable.

Metal oxide-based nano adsorbents: Metal oxide adsorbents are capable and efficient enough to remove heavy metals from wastewater. The nanometal oxides (NMOs)

Table 2: Recently studied microorganisms and plants for remediation.

Bacterial Strains	Targeted heavy metals	**References**
Cupriavidus necator	Cadmium, Copper, and Zinc	Vicentin et al. (2018)
Pseudomonas sp. Al-Dhabi-126	Cadmium	Al-Dhabi et al. (2019).
B. cereus A2 and *P. aeruginosa PS*	Cadmium and lead	Makki et al. (2019)
Bacillus sp.	Cadmium and lead	Heidari and Panico (2020)
Cupriavidus sp.	Cadmium	Minari et al. (2020)
Bacillus sp. TZ5	Cadmium	Ma et al. (2020)
Lactobacillus plantarum MF042018	Cadmium and Lead	Ameen et al. (2020)
Plants	**Targeted heavy metals**	**References**
Acacia nilotica	Cadmium	Shabir et al. (2018)
Atriplex lentiformis	Cadmium	Eissa and Abeed (2018)
Boehmeria nivea	Cadmium	Pan et al. (2019)
Noccaea caerulescens	Cadmium and Zinc	Kozhevnikova et al. (2020)
Sedum alfredii	Cadmium	Wu et al. (2020)
Youngia japonica	Cadmium	Yu et al. (2020)

which are commonly used are iron oxides, aluminum oxides, cerium oxides, titanium oxides, etc. as depicted in Fig. 8. Their shape and size play an important role in making them suitable nano adsorbents for heavy metals. Many methods are being employed to control their shape and make them highly stable (Cushing et al. 2004). The high adsorption ability of NMOs is due to the reaction of oxygen molecules of metal oxides with heavy metals.

There are numerous findings narrating the importance and extreme use of metal oxides for removing heavy metals. Few are depicted in Table 3.

Magnetic nanomaterials: Magnetic nanoparticles offer a faster and more economical approach for heavy metal removal from wastewater. They mainly comprise two components namely magnetic material (Iron, Cobalt) and a chemical component with the ability to remove metals. Super magnetic particles are magnetic particles with a smaller size that are highly effective for wastewater treatment. The nanoparticles' magnetic property allows them to easily separate the absorbents and reuse them in the system. Structural components are depicted in Fig. 9.

Among all magnetic nanoparticles studied to date, Fe_3O_4 magnetic nanoparticles were widely used and modified for cadmium removal from wastewater. Few findings are mentioned in Table 4.

Polymer-based nanocomposites: Polymer-based composites offer a reliable approach for the removal of heavy metals because of internal environment-friendly and decomposable features. They present superior physical, chemical, mechanical properties, pore size distribution, rigidity, compatibility as well as regeneration (Mahmoodi et al.2013). Polyaniline-based nano adsorbents and their derivatives have recently acquired popularity. As shown in Fig. 10, polymeric adsorbents can be divided into carbohydrate and synthetic polymers. Carbohydrate polymers include starch, cellulose, dextran, alginate, and other carbohydrates that are used to remove heavy metals from wastewater.

These polymers have functional groups like hydroxyl, amine, amide, and carboxyl which helps in removing heavy metals. Alginate offers the highest adsorption capacity due to carboxyl functional groups (Zare et al. 2018). However, the absorption ability of functional groups can be enhanced by using monomers such as carboxylic and amino groups which bind to polymeric matrices and help in targeting the contaminants (Mahmmodi et al. 2013). Numerous synthetic polymers have been used for wastewater treatment such as poly (styrene-alt-maleic anhydride) and poly (N-vinyl capro-

Table 3: Metal oxides for the removal of heavy metals.

Metal Oxide Nanoparticles	**Targeted heavy metals**	**References**
Aluminum Oxides	Cd (II) ions	Afkhami et al. (2010)
TiO_2 nanoparticles	Cd (II) ions	Engates and Shipley (2011)
Iron oxide	Cd (II) ions	Al-Saad et al. (2012)
Zinc oxide (ZnO) nanoparticles	Cd (II) ions	Sheela et al. (2012)
Manganese dioxide (MnO_2)	Cd (II) ions	Luo et al. (2013)
Copper oxide particles	Cd (II), Pb (II)	Taman et al. (2015)
Copper oxide nanoparticles	Cd (II), Ni ions	Hassan et al. (2017)

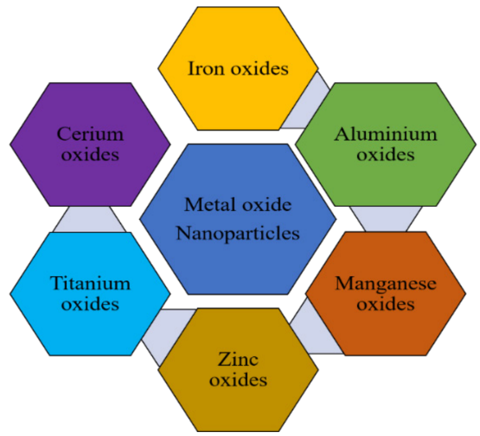

Fig. 8: Types of metal oxide nanoparticles.

Table 4: Recently synthesized magnetic nanoparticles for heavy metal removal.

Magnetic Nanoparticles	Targeted heavy metals	References
Aminopropyltriethoxy-silane Coated Fe_3O_4 nanoparticles (MNP)	Cd $^{(2+)}$, Pb $^{(2+)}$	Chen et al. (2016)
Superparamagnetic Iron Oxide	Cadmium	Goher et al. (2017)
Magnetite Fe_3O_4 nanoparticles	Cr (VI)	Rivera et al. (2019)
Fe_3O_4 magnetic nanoparticles	Zinc, Cadmium and Lead	El-Dib et al. (2020)
Silica coated Fe_3O_4.MNPs	Sr^{2+}	Salwa et al. (2020)
Magnetic zeolite nanocomposite	Cd (II), Zn (II)	Shubair et al. (2019)
Magnetic Fe_3O_4@ SiO_2-ethylenediamine tetra acetic acid	Pb (II), Cu (II)	Gong and Tang (2020)
Fe_3O_4@SiO_2 Nanoparticles	Cu^{2+} Ions	Irfai et al. (2020)

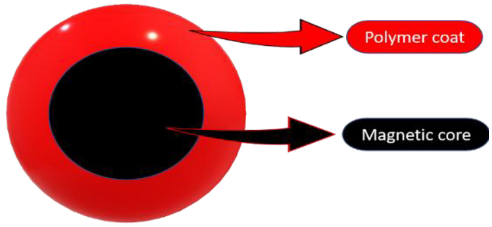

Fig. 9: Components of magnetic nanoparticles.

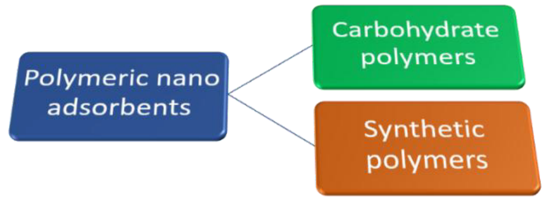

Fig. 10: Types of polymeric absorbents.

lactam-co-maleic acid) (Azarudeen et al. 2015, El-Aassar et al. 2016). Synthetic polymers have two main constituents namely polymer matrix and chelating groups. The PACA polymer is widely used as it has maximum adsorption capacity. The carbohydrate and synthetic polymers used to remove heavy metals are depicted in Table 5.

Zero valent nanoparticles: Zero valent ions are recently being used to remove heavy metals due to their enhanced reactivity and small size. The ordinate usage of zero-valent nanoparticles has increased owing to its high specific surface area (Galdames et al. 2020). The reaction of ZVI nanoparticles with heavy metals was investigated by looking at several parameters such as pH, dose, and the heavy metal's initial concentration (Chen et al. 2008). The potential adsorption of $Pb^{(2+)}$, $Cu^{(2+)}$, and $Zn^{(2+)}$ ions was examined using nanoscale zerovalent iron beads made from polyvinyl alcohol and guar gum (Zhang et al. 2019). Zero-Valent iron nanoparticles are extensively used for the decontamination of wastewater. It is reported to have a core-shell in the structure that contains oxidized iron (Phenrat et al. 2015)

Nano zero-valent iron nanoparticles were proved to be effective in lead removal (Ahmed et al. 2017). Nano dispersed powders of zero-valent iron were synthesized and effectively used for the removal of Cu (II), Cd (II), Co (II), Zn (II), Cr (VI) (Kholodko et al. 2021). The removal of cesium was also achieved by using nanoparticles-zeolite composites (Eljamal et al. 2019). Zero-valent iron nanoparticles were also reported to have the potential to remove Cd (II0 and Pb (II) ions (Danila et al. 2018). Zero-Valent iron nano adsorbents are employed to remove and remediate a wide variety of heavy metals as depicted in Fig. 11

Besides iron nanoparticles, zero-valent silver nano adsorbents were synthesized and studied for their application in the decontamination of wastewater by removing Hg^{2+} ions (Sundarajan et al. 2013). Effective removal of cadmium was achieved by zero-valent silver nanoparticles which were synthesized using *Benjamina* leaves extract (Al-Qahtani 2017) and observed that the rate of removal increased with the dosage of bio adsorbent.

Table 5: Carbohydrate and synthetic polymers used for removal of heavy metals.

Carbohydrate based polymers	Targeted metals	References
Alginate	Fe, Ag	Lu et al. (2015)
Starch	As, Pb	Cheng and Ou (2016)
Cellulose	Cd, Pb	Wang et al. (2013)
Synthetic polymers	**Targeted metals**	**References**
Polyethylene terephthalate, polyethylene, and polystyrene	Pb	Alsewailem and Al-Dzhlil (2016)
β-cyclodextrin polymers	Pb (II), Ni (II)	Yu et al. (2018)
Metal ion-imprinted polymers	$Ni^{(2+)}$ or $Co^{(2+)}$	Işıkver and Baylav (2018)
PVA with multiwalled carbon nanotubes	Pb	Zulfiqar et al. (2020)

CONCLUSION

The escalated urbanization and industrialization led to the massive generation of effluents with toxic pollutants such as cadmium in them. Thus, it is at utmost priority to remove contaminants and achieve water security too. In this regard, the use of nanomaterials is highly recommended owing to its numerous benefits not only to detoxify the wastewater but it is eco-friendly and cost-effective also. Many researchers are

Fig. 11: Applications of zero-valent iron nano adsorbents.

keen to develop creative, effective, and innovative approaches to deal with the problem and attain sustainability, such as the application of the green synthesis concept. The unique features of nano adsorbents have added a new dimension to wastewater treatment, however further industrial and pilot-scale research is needed to evaluate the total efficiency of nanomaterials for cadmium removal.

ACKNOWLEDGMENT

The authors are grateful to the University Institute of Biotechnology, Chandigarh University, Gharuan for providing a platform for study and valuable guidance.

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