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TOXIC HEAVY METAL IONS REMOVAL TECHNIQUES FROM WASTEWATER IN SAUDI ARABIA: A COMPREHENSIVE REVIEW

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ABSTRACT

Industrial wastewater has become a major problem due to high concentration of numerous harmful and hazardous pollutants, particularly heavy metals (HMs). Heavy metal pollution and its consequences for human health and the environment have increased interest in developing low-cost, long-term remediation technology. To remove heavy metals, various conventional physicochemical and green biological methods are used. This review article examines both conventional and biotechnological approaches for removing HMs from wastewater and rates their efficiency. The most appropriate technique for removing heavy ions from wastewater is determined by a number of factors, including operation cost, initial metal ion concentration, environmental impact, pH values, chemicals added, removal efficiency, and economic feasibility. These methods include adsorption treatments using various adsorbents, membrane treatments, chemical treatments, electric treatments *i.e.* electrochemical treatments. In general, chemical, adsorption, and membrane methods are the most practical method addressed in the literature. It has been noticed that there is a clear knowledge gap in the performance of treatment methods for the removal of heavy metal ions from real wastewater because most studies used synthetic wastewater in which one or few metal types are present. Accordingly, additional research should be conducted using real wastewater for treating different contaminants. More research on introducing cost-effective materials and methods for heavy metal removal from wastewater should be carried out. The best techniques for recovering metals with minimal environmental impact and low cost are still being developed and should be considered in future research.

INTRODUCTION

Water is exigent for agriculture and industrial activities as well as daily human life. Conspicuous population growth, economic development, and water consumption patterns diversity caused, the global water demand to increase by 600% in the past 100 years. In addition, urbanization also contributes distinctly to increase water consumption **(Chen *et al.*, 2017)**. It is estimated that 52% of global population live now under water scarcity **(Mekonnen and Hoekstra, 2016)**. According to the United Nation's Water Report, at least 3.6 billion people will face water scarcity in 2050. Human health, social and economic excellency, and the success of the ecosystem globaly depend on a reliable supply of high-quality water. In point of that water of great moment for all living beings, rapid urbanization and development have increased the demand for water. In addition, water is used in a variety of residential and industrial processes, including petroleum refineries as well as agricultural, medicinal, and pharmaceutical procedures. These activities introduce

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many toxic lesion pollutants and waste substances into water. Some of the industrial activities generate hazardous wastes.

Industrialization has hastily developed over the last century, that was accompanied with increase in the random usage of global resources and exacerbating environmental pollution crisis **(Briffa *et al.*, 2020)**. Some pollutants such as petroleum hydrocarbons, municipal waste and trace metals can accumulate to hurtful dangerous rates without being detected. Heavy metals are metallic elements with a higher density than that of water. Heavy metals also include metalloids like arsenic, which can cause toxicity even at low concentration, based on the idea that heaviness and toxicity are linked. Recently, environmental contamination by such metals has become a growing issue for the environment and global healthcare.

Large amounts of wastewater (WW) generated due to overpopulation and industrialization, are among the major wastes that pose a harsh threat to global environmental sustainability **(Eze *et al.*, 2018)**. Haste industrialization and urbanization are alpha and omega to blame for the rise in heavy metal contamination, which significantly accelerated rates of mobilization and movement in the environment. Petroleum refining industry, for example, generates wastewater comprise organic wastes such as benzene and hydrocarbons as well as inorganic wastes containing heavy metals. All industrial wastes, particularly heavy metals in wastewater, are highly toxic and have had disastrous impacts on the environment's balance.

In addition to the pollution resulted from fumes or volatile products of industrial reactions, hazardous industrial wastes are discharged into water in significant quantities, depending on the industrial activities. Increasing pollution hastily has prompter action to ensure a clean, pollution-free environment to support the growth of living beings. Accordingly, many stringent environmental laws and regulations governing wastewater systems have been enacted recently, recognizing that everybody around the world deserves to live in a clean and safe environment (**Razzak *et al.*, 2017**). The biological removal of heavy metals and other methods of nutrient removal from wastewater have recently received reality increased attention from the research community (**Razzak *et al.*, 2022**). Ensuing studies have resulted in technologies for removal of heavy metals from industrial wastewater for remediation of pollution caused by toxicants and pollutants. This review provides a description of various heavy metals, their effects due to environmental contamination, and different methods for the assessment of heavy metal pollution, in addition to common bioremediation techniques and factors limiting the bio-removal approaches. First, conventional remediation methods are presented, followed by a detailed analysis of the bioremediation processes.

Heavy metals are particularly problematic because they are non-biodegradable and thus persist. Heavy metals have been found in industrial wastewater. Cadmium is a non-essential metal that poses a health hurt even in trace rates in water. While cobalt can be used to treat anemia, having too much of it in the body can demolish the hematological systems and cause skin reactions (**Wang *et al.*, 2018**). To alleviate these potentially harmful health effects, environmental rules limiting heavy metal ion concentrations in water are becoming more imperative.

As a result, the focus of this investigation is on Toxic Heavy Metal Ions Removal Techniques from Wastewater in Saudi Arabia. The treatments are classified as adsorption, membrane, chemical, bio sorbents and photocatalytic. Each method is evaluated separately. Additional details about the operating conditions, removal efficiency, and important remarks of each method are listed for the reported studies in the literature. The available operating and performance parameters for each method are used to select the literature research.

METHODOLOGY

According to **Adeleye *et al.* (2016)** data acquisition. To collect data for this study, a systematic snowball approach was used. Primary and review articles were discovered using Web of Knowledge, Web of Science, Google Scholar, and additional articles were found using an iterative process for each relevant article. Evaluating the level of contamination in wastewater is important to determine the appropriate removal method according to the concentration of the pollutants. Several standard methods are available to analyze water and wastewater for contaminations. In the following sections conventional methods are described first and then biological treatment processes are presented in detail (**Razzak *et al.*, 2022).**

The tables provide data on sources of heavy metals, permitted concentrations, toxic effects, removal efficiency over a specified time period, removal capacity (generally in mg pollutant/g treatment technology), advantages and disadvantages of each method. In general, this allows for an objective comparison of these methods and, on that basis, determining the prospects for future studies **(Alimbaev *et al.*, 2021).**

Table 1 depicts the sources and toxic effects of heavy metals in industrial wastewater.

Table 2 depicts typical heavy metals found in wastewater and their sources, in addition to the health risks associated with excessive amounts and the permitted amounts in drinking water based on the world health organization (**WHO, 2014**) recommendations.

Conventional Methods for Removal of Heavy Metals

Prior to discharge into a water body, industrial effluents are subjected to some treatment stages involving a variety of physical, chemical, and biological techniques. Conventional treatment methods include membrane separation, chemical precipitation, oxidation/reduction, and biological treatment. For many years, investigators have used a variety of approaches based on electrochemical treatment, physiochemical treatment, and adsorption to remove heavy metals from industrial effluents. Fig. 1 depicts the majority of the conventional chemical and physical methods used for heavy metals removal based on these three approaches.

Electrochemical Processes

An electrochemical process is a chemical reaction resultant from the movement of an electrical current through an aqueous metal bearing solution caused by the application of a voltage between a cathode plate and an insoluble anode. During the process, heavy metals precipitate as hydroxides in a mildly acidic or neutralized electrolyte **(Tran *et al.*, 2017)**. The quality of the treated wastewaters would be determined by the amount of generated ions or charge loading, as well as the product of current and time **(Chen, 2004)**. Copper sulfate and nickel sulfate, for example are used as electrolyte in the process.

$CuSO_4 \rightarrow Cu^2 + SO_4^{2-}$and $NiSO_4 \rightarrow Ni^2 + SO_4^{2-}$

Electroflotation and Electrocoagulation

Electroflotation is a simple process that uses tiny hydrogen and oxygen bubbles formed on the cathode and anode of a flotation cell to float ions or solid particles suspended or dissolved in a liquid phase upward. The collector-to-metal-ion ratio is a critical determinant of a chemical species'

Table 1. Sources of heavy metals in industrial wastewater and their toxic effects

Heavy Metal	Major Source	Maximum Concentration Limit (mg/l)	Toxic Effect
Arsenic (As)	Smelting, mining, rock sedimentation, pesticides	0.01	Skin manifestations, visceral cancers, vascular disease
Lead (Pb)	Electroplating, pigments, mining, paint, burning of coal, manufacturing of batteries	0.015	Brain damage, anemia, malaise, loss of appetite, anorexia, kidney, liver, mental retardation in children, gastrointestinal damage
Copper (Cu)	Copper polishing, plating, printing operations, paint	1.30	Acute toxicity and neurotoxicity, diarrhea, dizziness, Wilson Disease, Insomnia
Mercury (Hg)	Mining, paint industries, paper industry, batteries	0.002	Kidney damage, corrosive to skin, eyes, damage to nervous system, muscles, protoplasm poisoning, dermatitis
Chromium (Cr)		0.01	Liver Damage, Headache, diarrhea, nausea, vomiting, carcinogenic
Zinc (Zn)	Plumping, refineries, brass manufacturing, mining	5.00	Causes depression, lethargy, neurological signs and increased thirst, short term metal-fume fever
Nickel (Ni)	Electroplating, porcelain enameling, paint formulation, non-ferrous metal	0.01	Lung cancer, chronic bronchitis, reduced lung function
Cadmium (Cd)	Refining, mining, welding, plastic, pesticide, fertilizer	0.005	Weight loss, renal disorder, kidney damage, bronchitis, gastrointestinal disorder, human carcinogen, Itai–Itai disease, hypertension, bone marrow, lung insufficiency, cancer

Source: Razzak *et al.* (2022)

Table 2. Shows typical heavy metals found in wastewater and their sources, as well as the health issues caused by excessive amounts and the permitted amounts in drinking water based on the world health organization (WHO, 2014) recommendations

Common heavy metal	Main source	Main organ and system affected	Permitted amount (µg)
Lead (Pb)	Lead-based batteries, solder, alloys, cable sheathing pigments, rust inhibitors, ammunition, glazes, and plastic stabilizers.	Bones, liver, kidneys, brain, lungs, spleen, immunological system, hematological system, cardiovascular system, and reproductive system.	10
Arsenic (As)	Electronics and glass production.	Skin, lungs, brain, kidneys, metabolic system, cardiovascular system, immunological system, and endocrine.	10
Copper (Cu)	Corroded plumbing systems, electronic and cables industry.	Liver, brain, kidneys, cornea, gastrointestinal system, lungs, immunological system, and hematological system.	2000
Zinc (Zn)	Brass coating, rubber products, some cosmetics, and aerosol deodorants.	Stomach cramps, skin irritations, vomiting, nausea, and anemia, and convulsions.	3000
Chromium (Cr)	Steel and pulp mills and tanneries.	Skin, lungs, kidneys, liver, brain, pancreas, tastes, gastrointestinal system, and reproductive system	50
Cadmium (Cd)	Batteries, paints, steel industry, plastic industries, metal refineries, and corroded galvanized pipes.	Bones, liver, kidneys, lungs, testes, brain, immunological system, and cardiovascular system.	3
Mercury (Hg)	Electrolytic production of chlorine and caustic soda, runoff from landfills and agriculture, electrical appliances, Industrial and control instruments, laboratory apparatus, and refineries.	Brain, lungs, kidneys, liver, immunological system, cardiovascular system, endocrine, and reproductive system.	6
Nickel (Ni)	Stainless steel and nickel alloy production.	Lung, kidney, gastrointestinal distress, pulmonary fibrosis, and skin.	70

Source: Naef *et al.* (2021)

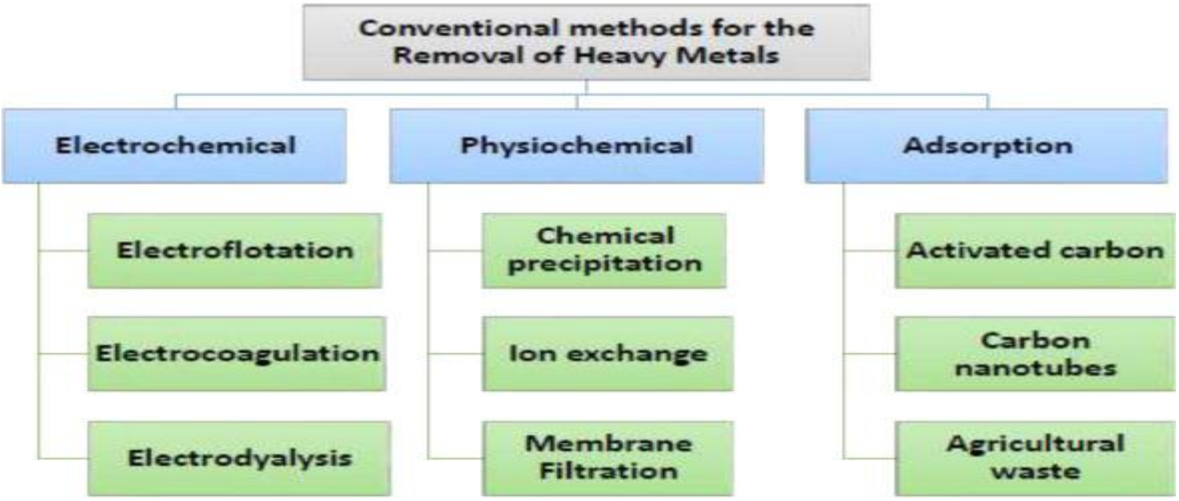


Fig. 1. Conventional methods used in removal of heavy metals from wastewater

Source: Razzak *et al.* (2022)

floatability. Accordingly, the collector used in ion flotation must be at least stoichiometric and have foamy characteristics (**De Oliveira Da Mota *et al.*, 2015**).

Electrocoagulation has been used in water and wastewater treatment to remove heavy metals, organics, bacteria, hardness, turbidity, and other pollutants **(Zhu *et al.*, 2005)**. Electrocoagulation is an electrochemical method that uses an electrical current to remove metals from solution. An electrocoagulation device can remove suspended particles, dissolved metals, tannins, and dyes. The electrical charge in wastewater keeps the pollutants in solution. These ions and other charged particles become destabilized and precipitate when they are neutralized by ions of opposite electrical charge produced by the electrocoagulation system.

Electrodialysis

Water is required in large quantities in some industrial processes. Thus, reducing, recycling, recovering materials, and reusing wastewater provide significant economic benefits. Electrodialysis is a good method for treating industrial wastewater. Electrodialysis is a membrane process that uses charged ion exchange membranes to separate charged metal ions from a fluid solution **(Cifuentes *et al.*, 2009)**. Because electrodialysis is a simple and efficient process, some studies have been conducted to determine its ability to remove contaminants from wastewater. This process only works with ionized substances in solution, such as sodium chloride, which forms sodium and chloride ions in solution. When terminals connected to the appropriate coordinate current supply are soaked in salt solution, the current stream is carried by ions. Cations are positively charged particles that are attracted to the negative cathode. Anions with negative charges flow towards the positive anode. Layers or a channel of specific anions and cations are placed between cathodes in electrodialysis. Cation channels allow a stream of anions but act as an obstruction for cations, whereas anion channels obstruct anions but allow cations to pass through **(Samiey *et al.*, 2014)**.

Physiochemical Processes

Precipitation

Chemical precipitation is a method that is widely used to remove inorganic metal ions from solution. Chemical precipitation occurs in an insoluble substance allowing it to precipitate. The method involves supply of a precipitation reagent to waste products resulting in chemical reactions producing insoluble substances. The insoluble substance forms particles, which are clumped by chemical coagulation and removed by filtration or sedimentation **(Akinterinwa, 2018)**. Heavy metals in wastewater can be removed using various treatment methods, including chemical precipitation. The mechanism is influenced by pH of the wastewater, the solubility product K_{sp}, and the metal content.

Ion exchange

Ion exchange reduces sludge volume while allowing for selective recovery and meeting stringent release requirements (**Zewail and Yousef, 2015)**. Allusion to using ion exchange method, researchers used various resins to remove heavy metals. (**Shaidan *et al.*, 2012)** used a robust acidic cation exchange resin in a fixed bed to remove nickel from wastewater. The resin has several benefits, including the ability to treat large volumes of effluent at once and to remove heavy metals from wastewater more efficiently. The researchers were able to remove 97% of nickel from wastewater.

Coagulation and flocculation

Coagulation is one of the most widely used wastewater treatment methods due to its simplicity and effectiveness **(Abu Tawila *et al.*, 2019)**. Coagulation simply refers to the neutralization of the particle charge. This method can filter out heavy metal solids from solution **(Verma *et al.*, 2012)**. This is a very practical method of dealing with heavy metal contamination whether you have water treatment tools or not. This mechanism destabilizes the particle charges by introducing highly charged small molecules into water. Furthermore, this technique is typically coupled with flocculation where the colloidal particles exit the suspension to flocculate and form sediments **(Wang *et al.*, 2005)**.

Membrane filtration

Membrane filtering techniques are classified according to the type of membrane used. Filtration has several advantages over other traditional techniques, including high separation efficiency, no phase changes, energy savings, ease of scaling up, and environmental friendliness **(Zhu *et al.*, 2014)**.

Ultrafiltration

At low pressures, ultrafiltration (UF) removes dissolved and colloidal particles. UF techniques include polymer-enhanced ultrafiltration (PEUF) and micellar-enhanced ultrafiltration (MEUF). MEUF is a method of treating wastewater with an anionic surfactant prior to physicochemical membrane separation. Anionic surfactant monomers combine to form micelles, and electrostatic forces trap heavy metals on the micelle's outer surface. Excess surfactant and heavy metals that are not trapped on the micelle's external surface pass through the UF membrane. MEUF has some advantages, such as high flux, high removal, and low energy costs, but it also has the disadvantage of increasing operating costs. **Landaburu-Aguirre *et al.* (2009)** extracted cadmium and zinc from wastewater using the MEUF method and sodium dodecyl sulphate as a surfactant. They were able to remove 98% of zinc and 99 % of cadmium.

Nanofiltration

This membrane rejected cadmium, chromium, and lead at rates 95%, 98%, and 93%, respectively, by this membrane. **Mehdipour *et al.* (2015)** used a polyamide nanofiltration membrane for lead removal and achieved 97.5% removal of lead by increasing the pressure and initial feed concentration.

Reverse osmosis

Reverse osmosis is a physical filtration procedure that uses a higher pressure, greater than the osmotic pressure, to force water through a semi-permeable membrane with specific properties to filter out unwanted molecules by forcing a solvent from a high concentration area to low concentration area **(Kanamarlapudi *et al.*, 2018).** Because it can only remove low levels of heavy metals from industrial wastewater **(Ranitha *et al.*, 2016).** Reverse osmosis was used to remove Cu (II), Ni (II), and Zn (II) using a polyamide thin-film composite membrane TW30-1812-50 **(Bakalar *et al.*, 2009)**.

Adsorption Process

Adsorption method is a low-cost method. Adsorption has many advantages comparably with traditional methods, like less biological and chemical sludge, lower costs, increased effectiveness, metal recovery, and adsorbent regeneration. Adsorbents for heavy metal inclusive of activated carbon, clinoptilolite carbon nanotubes, zeolites, manganese oxides, and agricultural waste products **(Sardella *et al.*, 2015)**.

Activated carbon

Activated carbon is produced from agricultural wastes such as coconut buttons, moso and bamboo, grape and olive stones and lignin. **(Renu *et al.*, 2017)**. **Anirudhan and Sreekumari (2011)**) created activated carbon from leftover coconut buttons to remove lead, mercury, and copper from wastewater. The utmost adsorption capacity for each of Cu(II) and Pb(II) was observed at pH 6.0, while the best adsorption capacity for Hg(II) was observed at pH 7.0. Activated carbon's adsorption capacity increases in the order of Cu(II) Hg(II) Pb (II). Lead removal is greater than 90%, and copper and mercury removal is greater than 95%.

Carbon nanotubes

Carbon nanotubes have sparked a lot of interest in the last decade ascribed to their specific area, stability and excellent electrical and mechanical properties. **Kosa *et al*. (2012)** used 8-hydroxyquinoline to customize multiwalled carbon nanotubes for the removal of cadmium, copper, lead, and zinc. Cadmium was found to have a removal efficiency of more than 80%.

Photocatalysis

Photocatalysis with non-toxic semiconductors allows rapid and efficient breakdown of environmental contaminants. Transfer, surface adsorption by the semiconductor, surface photocatalytic reactions, and eventual desorption and removal of contaminants in the interface area are all involved in this approach. A selenium-doped ZnO nanocomposite semiconductor was used to photocatalytically reduce and remove heavy metals in pharmaceutical waste, with removal capacities of 0.211 (Cr), 0.421 (Cu), 0.097 (Cd), and 0.147 (Pb) per 0.5g of ZnO/Se nanocomposite.

Overview of Conventional Methods

Heavy metal removal from wastewater is classified into three major process types, which are discussed. All methods are effectual and selective for high and low heavy metal concentrations in wastewater. As shown in Table 3, each method has several advantages and disadvantages. Eke, adsorption is a cost-effective technology that has several advantages over conventional methods, including the reduction of sludge, high effectiveness, minimal cost, the possibility of metal recovery, and adsorbent regeneration. Bio-sorption mechanism is used for adsorption. The efficiency of adsorbents for removal of heavy metals is determined by the type of adsorbent. Membrane filtering generates less solid waste, uses less chemicals, and is highly effectual. However, one of its drawbacks is the low flow rates. Even though the flocculation / coagulation technique has the benefit of achieving sludge dewatering and settling while high

Table 3. An overview of the conventional heavy metal removal process (Adapted from Al-Qodah and Al-Shannag, 2017 and Al-Qodah *et al*. 2017)

Process	**Methods**	**Advantages**	**Disadvantages**
Electrochemical	Electroflotation and electrocoagulation	Selective for metal ions, no chemical consumptions, most of the metal ions possible to remove	High current density requirement, high operational and capital cost
	Electrodialysis	Elevated selectivity	Elevated operation cost owing to energy consumption and membrane fouling
Physiochemical	Chemical precipitation	Low-cost, friendly operation,	Sludge disposal problem owing to huge quantity of sludge
	Ion exchange	Regeneration of materials, selective for metal ions	Accessible for less number of metal ions, expensive
	Coagulation and flocculation	Less sludge settling, dewatering	Costly and high consumption of chemicals
	Membrane filtration	High separation selectivity, low space requirement, low pressure	Maintenance and operation cost high
	Nano filtration	High separation efficiency, easy operation, reliability	When compared to ultra-filtration, it has a low anti-compacting ability
Adsorption	Activated carbon	High Efficiency	Performance depends on adsorbent, costly, no regeneration
	Carbon nanotubes	Highly selective and adsorption capacity	Very costly, no regeneration capacity
	Agricultural waste	Cheaper, relatively higher removal capacities and most of the metal removal possibilities, eco-friendly	High solid waste, Less research and application
	Photocatalysis	Simultaneous removal of metals and organic pollutants, less harmful bioproduct	Require longer time for separation, limited applications

Source: Razzak *et al.* (2022)

cost and high chemical consumption are its disadvantages. The ion exchange process has the advantage of not changing the pH of the effluent. Other benefits include safety due to chemicals not been used, stability, and high process reliability. However, the high membrane cost as well as the need for resin renewal due to fouling are its disadvantages. The method selected is determined according to the initial metal concentration, operating costs, and environmental impact.

The previous discussion indicates that conventional separation methods are promising, but the cost of investment and long-term operation costs are high for large-scale applications. At the same time, chemical treatment, the need for chemicals, and additional storage or solid handling costs are all factors to consider. Given all of the drawbacks associated with traditional heavy metal removal methods, there is a demand for unconventional and cost-effective alternatives. As a result, low-cost, efficient heavy metal removal methods that are also environmentally friendly are required. There is growing interest in using biological agents to remove heavy metals as an alternative to traditional methods. New technologies have given rise to the use of biological methods and means for the removal of metal ions from polluted industrial waters, with a high potential for higher production at lower costs than conventional heavy metal removal wastewater purification processes **(Hazarika *et al.*, 2015).** Taking into account all of the disadvantages accompanied with the use of conventional heavy metal removal methods, there is a demand for unconventional and cost-effective alternatives **(Barakat, 2011)**. Bio-HMR (heavy metal removal) systems use biomass to extract heavy metals from effluents and are frequently described as low-cost, environmentally viable, and simple to operate alternatives.

Biosensors for Detection of Heavy Metals

Evaluating the level of contamination in wastewater is critical for determining the best method for removing pollutants based on their concentration. There are numerous standard methods for testing water and wastewater contamination. The exposure of organisms, including plants, bacteria, and even in small mammals, to heavy metals can lead to metal accumulation, which may induce biological changes and disturbances to these organisms and affect their cell cycles **(Rogers and Lin, 1992)**. The response of immobile organisms to heavy metal toxicity reveals the extent of heavy metal pollution at the organism's location. As a result, plants and other microorganisms present at the site of contamination can be used as biosensors to biomonitor the environment for the presence of various pollutants **(Tovar-Sánchez *et al.*, 2019).** Because of their ability to absorb heavy metals, plants are more prone to DNA alteration or genotoxicity. Table 4 lists common plants that have DNA strand breaks as a result of heavy metal exposure.

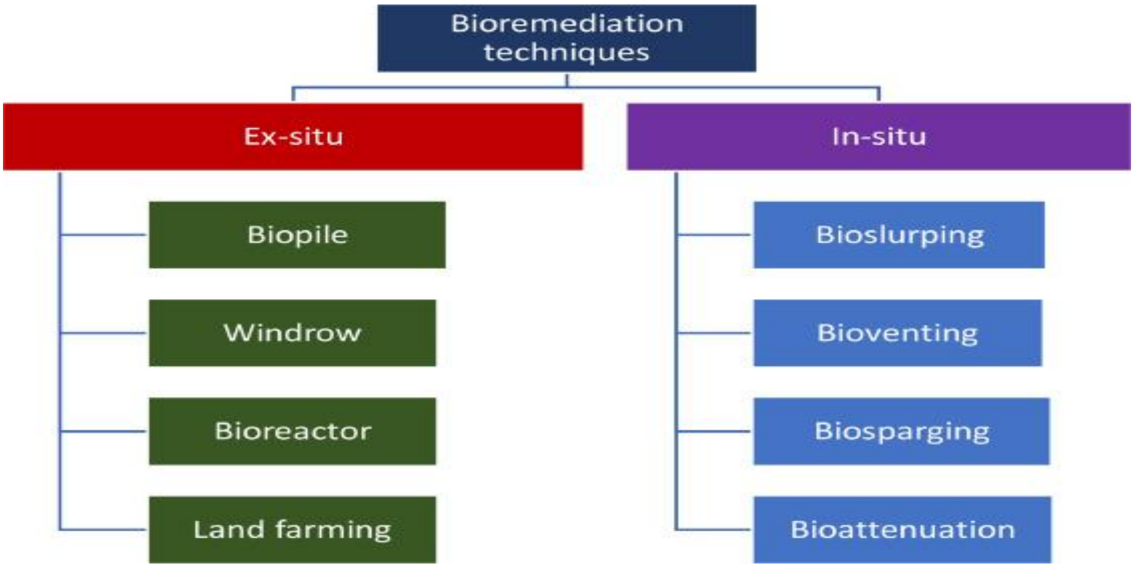
Bioremediation of Heavy Metals

Bioremediation is the process of removing harmful contaminants from a polluted environment such as heavy metals or transforming them into less hazardous substances. Bioremediation techniques are divided into two categories based on how they are used: in situ bioremediation and ex situ bioremediation (Fig. 2). The main distinction between the two is that in situ bioremediation is used at the site of contamination. Ex situ bioremediation, on the other hand, is used after the medium has been removed from the area to be treated or remediated. In situ biodegradation involves introducing oxygen and nutrients into polluted environments such as soil and wastewater in order to facilitate the oxidation of toxins by naturally occurring bacteria. In situ bioremediation is classified as either fundamental (intrinsic) or artificial (external) stimulation. The goal of intrinsic bioremediation is to activate the naturally occurring microbial community to increase metabolic activity by supplying nutrients and oxygen. Microorganisms, on the other hand, are introduced to the contamination.

Table 4. Plants that show biochemical effects upon their exposure to heavy metals

Plant	Scientific Name	The HM exposed to
Plantaginaceae	*Bacopa monnieri*	Cd
Asteraceae	*Nicotiana tabacum*	Cd and Cu
Fabaceae	*Pisum sativum*	Cr
Solanaceae	*Lycopersicum esculentum*	Cu
Wigandia	*Wigandia urens*	Pb, Zn, and Cu

Source: Razzak *et al.* (2022)



Source: Razzak *et al.* (2022)

Fig. 2. Classification of bioremediation techniques

site in engineered or artificial bioremediation **(Kulshreshtha *et al.*, 2014)**. Each bioremediation type has its own set of techniques. In situ bioremediation techniques may be less expensive than ex situ bioremediation approaches

Heavy metals in wastewater can be bioremediated in two ways: bioaccumulation and biosorption. Biosorption is a metabolically passive process carried out primarily with non-living microbial or biological materials (*e.g.*, agricultural waste), whereas bioaccumulation needs the presence of living organisms and occurs in the later stages of biosorption. Bioaccumulation and biosorption technologies have recently emerged as a new cost-effective and environmentally friendly approach to industrial wastewater treatment. Bioaccumulation is a metabolically active process in which microorganisms construct a translocation channel through the lipid bilayer to accept heavy metals into their intracellular space using imported complexes (*i.e.*, import system). Heavy metals can be isolated by proteins and peptide ligands after they enter the intracellular space. Adsorption of particles to a biological matrix by chemical or physical interaction, chelation, or complexation, on the other hand, is known as biosorption. At neutral pH, anionic moieties on microorganism extracellular surfaces serve as attachment sites for cationic heavy metals (**Michalak *et al.*, 2013).** The following two sections provide a detailed and critical review of biobased separation.

Bioaccumulation of Heavy Metals

The accumulation of pollutants such as heavy metals or pesticides in living organisms is known as bioaccumulation. Bioaccumulation is defined as a metabolically driven functional process in which metal ions are taken up intercellularly by the biosorbent and incorporated into living cells **(Wang, 2016).** This process is critical to the status of metals in the environment. Heavy metals are known to be taken up and collected by fluid plants. A team of scientists investigated the ability of aquatic vascular plants to absorb heavy metals. For the study, they collected wastewater samples containing heavy metals from the Donghe River in China. The bioaccumulation ability of a type of biomass for a target heavy metal is typically expressed as mg per g dry biomass weight Table 5 summarizes the values for cobalt (Co), cadmium (Cd), mercury (Hg), copper (Cu), uranium (U), and nickel (Ni). The sample used contained various spiked concentrations of the previous heavy metals. Another study looked at the use of Nasutrium officinale, an aquatic plant known for accumulating heavy metals like Ni, Cu, and Zn (**Kara, 2005).** The ability of two other plants, *Typha latifolia* and *Thelypteris palustria*, to accumulate Zn and Cu has also been studied **(Hejna *et al.*, 2020).**

Biosorption of Heavy Metals

Biosorption, like bioaccumulation, is a physicochemical pathway that allows certain biomass of biological origin to passively accumulate heavy metals by binding them to its cellular structure **(Fomina and Gadd, 2014)**. Biosorption, a type of adsorption that uses a biological matrix as the sorbent, is the fast and reversible binding of ions from aqueous solutions to functional groups on the surface of biomass. Biosorption employs a variety of microorganisms, plant-derived materials, wastes, biopolymers, and other substances in a reversible process. Accumulation is accomplished through a variety of interactions that allow ions to bind to functional groups on the bioabsorbent's external surface. This process is very efficient because it is a simple, energy-free operation that does not require any additional nutrients (**Wang and Chen, 2009**).

Bioadsorption is a promising treatment technology due to its increased efficacy with minimal chemical and/or biological waste sludge treatment, low cost and short time required, ability to reuse bioadsorbents, and versatility of bioadsorbents. Bioadsorption does not require additional nutrients and is also eco-friendly because the required material is locally available, selective bioadsorbents can be used for specific metals of interest, heavy metal ions can be removed regardless of their toxicity, does not produce secondary toxic compounds, is simple to implement, and low-cost bioproducts that can be used as bioadsorbents are available (**Al-Qodah *et al.*, 2017**). Biosorption has attracted a great deal of attention due to the availability of re-usable biosorbents and the low volume of sludge produced **(Maximous *et al.*, 2010**).

Aside from the benefits mentioned above, microbial cell metabolism and the location of metal removal have a significant impact on biosorption methods **(Papirio *et al.*, 2017).** Biosorption can be metabolically or non-metabolically dependent, depending on the activity of the biomass. Biosorption can occur via the following methods, depending on the nature of metal removal:

Table 5. A brief outline of bioaccumulation research for several heavy metal ions

Metal Accumulation (mg/g dry weight)	**Cd**	**Pb**	**Hg**	**Zn**	**Cu**	**Ni**
Species						
Nasturtium officinale				19	41	18
Hydrilla verticillata	3±0.5	8±1.5				
Typha latifolia				271.±17	47±4	
Thelypteris palustris				409± 17	105 ±4	
***Escherichia coli* BL21**	7.5				145	
***Escherichia coli* Rosetta**	2.24		0.82			
***Rhodopseudomonas Palustris* GIM1.167**			77.58			
***Ceratophyllum demersum* L.**		12.06			2.51	
Nasturtium officinale				17	88	14
***Ochrobactrum intermedium* BPS-20**		850 mg/L			2400 mg/L	

Source: Razzak *et al.* (2022)

(a) intracellular accumulation, (b) cell surface sorption/precipitation, and (c) extracellular accumulation/precipitation. Fig. 3 shows a flow diagram of the biosorption mechanism based on the presence or absence of metabolism

Complexation

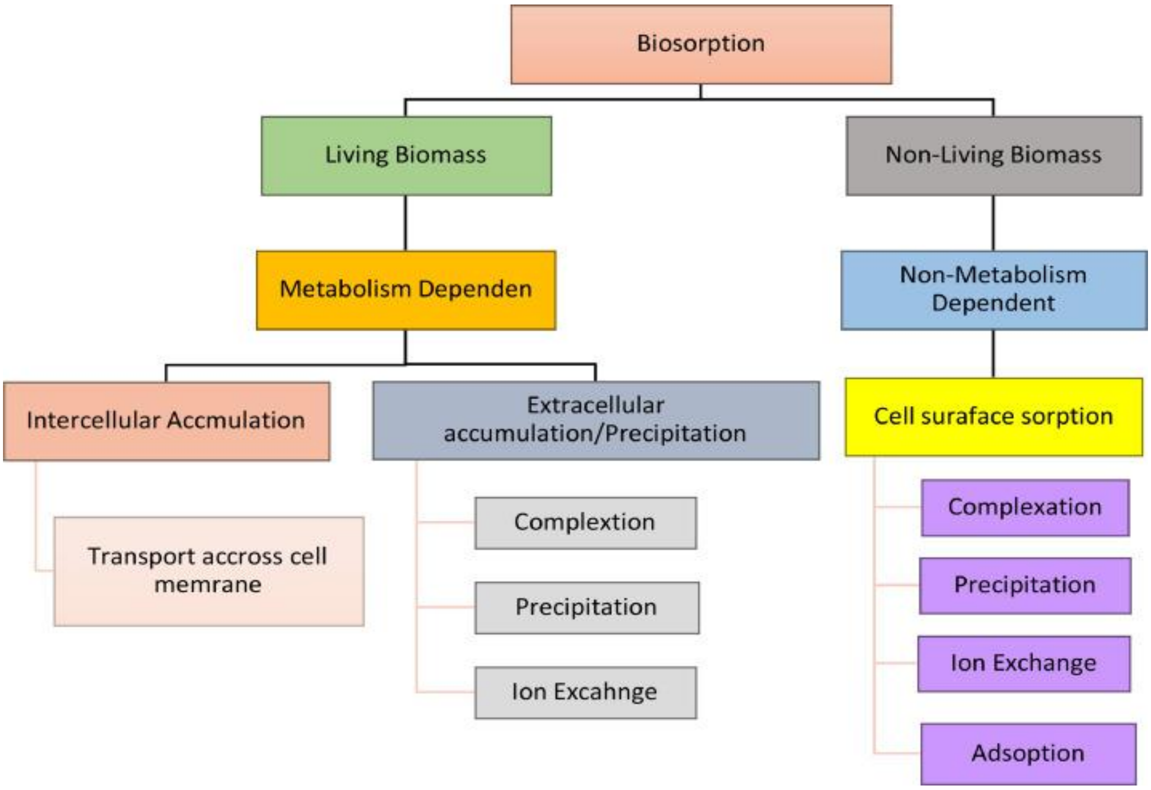
Complexation is a chemical reaction that occurs between a metal ion and a ligand that contains at least one atom and an unshared pair of electrons to form a metal complex **(Varma *et al*., 2004).** A metal ion and a ligand form a mononuclear complex, with the metal at the center. A polynuclear complex is formed by more than one metal atom in the center, and the metal's charge can be positive, negative, or neutral depending on the amount of binding ligand present (**Yang *et al*., 2017).**

Chelation

Chelation is a more sophisticated form of complexation. Chelation refers to the reversible attachment of molecules to metal ions. The formation of a metal complex known as the chelate results from the binding of the ligand, the chelant, to a metal ion. Because of its multiple bindings, the chelant can bind to metal ions in a variety of locations and form a stable encircling structure. As the number of binding sites increases, so does the chelate's stability **(Dautel-Morazin *et al*., 1991**).

Coordination

A coordination compound is a chemical structure in which a central metal atom is surrounded by nonmetal molecules, which are known as ligands or complexing agents in chemistry. A coordinating atom, which is part of the nonmetal molecule, and an acceptor, which is the metal ion, are both present in such an entity. Coordination compounds are compounds with such bonds when they are combined **(He and Chen, 2014).**



Source: Razzak *et al.* (2022)

Fig. 3. Correlation of biosorption mechanisms with metabolism and nature of metal removal

Ion Exchange

The ion exchange reaction is another biosorption reaction. Ion exchange is a chemical reaction that involves the exchange of at least one ionic component between two substances. This is one of the most useful concepts in biosorption, and it involves the exchange of metal ions with ions on the biosorbent. The majority of filtration procedures rely on ion exchange, which can be accomplished *via* anion or cation exchange. A cation exchanger can also be represented by the carboxyl group **(Kanamarlapudi *et al.*, 2018).**

Precipitation

Precipitation with a biosorbent is similar to precipitation with inorganic chemicals in that the metal forms a bond with a functional group on the surface of a microbial cell **(Kumar *et al.*, 2021).**

Reduction

Metals interact with functional groups and are reduced during the crystal growth process. Components such as gold and palladium are typically reduced. When metals bind to a biosorbent at specific locations, they are reduced. By reduction, harmful hexavalent can be converted to metallic chormium **(Kanamarlapudi *et al.*, 2018).**

Biosorbents for Biosorption

Biosorbents are living and non-living biomass cellular products that are used for effective adsorption. Many biological materials are suitable for use as biosorbents due to their superior efficacy in removing heavy metals from wastewater, economic effectiveness, and specific affinity for metals, metalloids, and other contaminants **(Gadd, 2009).** Many studies have been conducted to evaluate bacteria, archaea, algae, fungi, agricultural wastes, and yeasts for use as biosorbents. The following subsections describe the various types of biosorbents used for heavy metal removal.

Industrial By-Products

Waste treatment has made use of byproducts from various industries. Several industries, including the food industry, generate massive amounts of waste and byproducts. Because of the presence of metal-binding functional groups, these zero-cost waste can be used as effective biosorbents. Industrial waste from the aluminum, paper, fertilizer, and steel industries, among others, can be used as biosorbents **(Hegazi, 2013).** Waste beer yeast is a byproduct of the brewing industry with a high capacity for adsorption of heavy metal ions such as copper and lead. A group of researchers investigated the use of used beer yeast for copper and lead ion biosorption in wastewater. Their results showed that used beer yeast can be used as a biosorbent to remove copper and lead from wastewater **(Stanila *et al.*, 2016).** Another group investigated the use of rice husk, a byproduct of rice milling, for heavy metal biosorption as an alternative to costly treatment processes. The main components of rice husks are lignin, cellulose, and hemicellulose. They measured the ability of rise husk to remove Ni, Cd, Cu, Pb, and Zn from a solution containing the metals under ideal conditions **(Hegazi, 2013).** Table 6 shows the sorption capacity of various industrial by-products used as biosorbents.

Agricultural Waste Materials

The use of agricultural waste and byproducts as biosorbents is gaining popularity in the removal of contaminants from wastewater **(Ahluwalia and Goyal, 2007).** Due to their low cost and promising abilities, researchers have conducted numerous studies on the use of agricultural by-products as biosorbents for heavy metals. As previously stated, the process involves a sorbent and a solvent. The sorbent is the solid phase, while the solvent is the liquid phase, with dissolved components to be sorbed **(Nguyen *et al.*, 2013**; **Šoštarić *et al.*, 2015**). Table 7 shows the adsorbent capacities of agricultural waste-based materials used as adsorbents.

Microbial Biosorbents

Algae as biosorbents

The use of algae as a biosorbent is appealing due to its low nutrition requirements, wide availability, high surface area to volume ratio, high sorption capacity, and low sludge waste production. Algae are regarded as an environmentally friendly and cost-effective wastewater treatment solution. The cell wall components and characteristics of different types of algae differ. Brown algae cell walls contain cellulose, alginic acid, and sulfated polysaccharide, as well as an abundance of carboxyl groups, which aid in metal ion bio sorption. Because of the presence of sulphate polysaccharides composed of galactans, red algae is also of interest for bio sorption. Algae can biosorb 15.3-84.6 percent of pollutants, which is higher than the upper limit of other microorganism bio sorbents. Table 8 shows a summary of published results on bio sorption of heavy metals by different algae.

Bacteria as biosorbents

Biosorption relies heavily on cell surfaces. Peptidoglycan is the main component of bacterial cell walls. Bacteria are classified as gram positive (G^{+ve}) or gram negative (G^{-ve}) based on the structure of their cell walls **(Javanbakht *et al.*, 2014).** Gram positive bacteria are those that have a thick peptidoglycan bridged by amino acids **(Kiliç *et al.*, 2015).** Gram negative bacteria are bacterial organisms with less peptidoglycan on their cell walls **(Ahluwalia and Goyal, 2007).** Bacteria are excellent biosorbents due to their high surface area to volume ratio and abundance of potential active sites that can act as sorption sites. Table 9 summarizes bacteria-based microbial biosorbents used for heavy metal ion biosorption.

Table 6. Summary of industrial by-products used as biosorbents for various heavy metal ions

Source	**Material**	**Metal**	**Sorption capacity (mg/g)**
Juice and Jam industry	Peach and apricot stones	Cu (II)	4.45
	Banana peel	Cu (II)	4.75
		Zn	5.80
		Pb	7.97
		Ni	6.88
Paper mill	Sludge	Ni	13.7
		Cu	5.5
		Pb	14.1
		Cd	14.8
Cement industry	Fly ash	Pb	22
Rice mill	Rice husk	Ni	3.88
		Cd	16.18
		Cu	29
		Pb	4.23
		Zn	5.38

Source: Razzak *et al.* (2022)

Table 7. Summary of biosorbents based on agricultural waste materials for biosorption of heavy metal ions.

Material	**Metal**	**Sorption capacity (mg/g)**
Mango peel	Cd(II)	68.92
	Pb	99.05
Bael fruit	Cr(VI)	17.27
Chitosan	Cu(II), Mn(II), Fe(II), Ni(II)	99%
Coconut husk	Cu(II), Fe(II), Pb(II)	90%
Tea waste	Ni (II)	58%
Tomato waste	Pb(II)	152
Chestnut	Cd	34.77
Grape stalk	Ni	10.6

Source: Razzak *et al.* (2022)

Table 8. Summary of algae-based microbial biosorbents for various heavy metal ions

Algae Species	Metal	Initial concentration (mg/L)	Sorption capacity (mg/g)
Aspargopsin armata	Cd	135	32.2
	Ni	141	17.7
	Zn	182	21.6
	Cu	134.4	21.3
	Pb	124	63.7
Codium vermilara	Pb	83	63.3
	Zn	182	23.8
	Cu	182	23.8
	Ni	147	13.2
	Cd	135	21.8
Cystoseira barbata	Cd	117	37.6
	Ni	224	78.7
	Pb	414	196
P. boryanum cell	Cr(VI)	20-400	17.3
A. junii-coconut fiber	Cr(VI)	25-200	17.4
C. reinhardtii	U(VI)	50-1500	232.6

Source: Razzak *et al.* (2022)

Table 9. Summary of bacteria-based biosorbents used for biosorption of various heavy metal ions.

Bacteria	Metal	Initial concentration (mg/L)	Sorption capacity (mg/g)
Bacillus cereus	Hg	10	104
B. laterosporus	Cd	1000	159.5
	Cr	1000	72.6
B. lincheniformis	Cd	1000	142.7
	Cr	1000	62
Desulfovibrio desulfuricans	Cu	100	98.2
	Ni	100	90
	Cr	100	99.9
Enterobacter cloacae	Pb	7.2	2.3
Kocuria rhizophila	Cd	150	9.07
	Cr	150	14.4
Micrococcus luteus	Cu	80	408
	Pb	272	1965
Pseudomones aeruginosa	Co	58	8.92
	Ni	58	8.26
	Cr	52	6.42
P. jessenni	Ni	275	1.36
	Cu	300	10.22
	Zn	400	4.39
Pseudomonas sp.	Zn	275	3.66
	Ni	275	2.79
	Cu	300	5.73
Sulphate-reducing bacteria	As(III)	1	0.07
	As(V)	1	0.11
P. putida	Cu	10-40	22.23
Pesudomonas putida	U(VI)	100-1000	625

Source: Razzak *et al.* (2022)

Fungi as biosorbents

Fungi are economical and environmentally friendly bio sorbents due to their ease of modification, high yield, and ease of cultivation **(Acosta-Rodrguez *et al.*, 2018).** Fungi cell walls have good binding abilities due to unique characteristics such as lipids, chitin, proteins, and polyphosphates. Fungi cell walls are thought to be very rich in glycoproteins and polysaccharides with different types of groups that can bind metals such as amines, carboxyl, and hydroxyl **(C´ardenas *et al.*, 2019).** Several fermentation processes use fungal organisms for biosorption as well as industrial purposes Fungi's filamentous nature allows them to be separated using simple techniques such as filtration **(Herath *et al.*, 2014).** Yeasts are unicellular organisms that can bioabsorb a wide range of metals as well as a single metal ion. Bioaccumulation properties have also been reported for yeast. Table 10 summarizes fungi-based microbial biosorbents for heavy metal ion biosorption.

Table 10. Summary of fungi based biosorbents used for biosorption of various heavy metal ions

Fungi	Metal	Initial concentration (mg/L)	Sorption capacity (mg/g)
Aspergillus niger	Cu	100	15.6
	Pb	100	34.4
	Cr	50	6.6
Botrytic cinereal	Pb	350	107.1
Phanerochaete chrysosporium	Pb	100	88.16
	Cu	100	68.73
	Zn	100	39.62
Pleurtus platypus	Ag	200	46.7
Rhizopus oryzae	Cu	100	34
C. lipolytica	NI	25-460	123.43
Magnetic R. glutinis	U(VI)	10-320	226

Source: Razzak *et al.* (2022)

Conclusion

This review article gives an overview of recent results on the uses and efficiency of both conventional and biotechnological heavy metal removal techniques. Ion exchange, chemical precipitation, membrane separation, coagulation, flocculation, adsorption, electrochemical methods, and flotation are all described indicating their advantages and disadvantages. Various biotechnological techniques, such as bioaccumulation and biosorption are evaluated. Biosorption of heavy metals is a multi-step process comprising complexation, chelation, coordination, ion exchange, and precipitation. In general, readers will be able to learn some fundamental and recent knowledge that will be helpful in their planned studies.

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الملخص العربي

تقنية إزالة أيونات المعادن الثقيلة السامة من مياه الصرف الصحي في المملكة العربية السعودية: مراجعة شاملة

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أصبحت مياه الصرف الصناعي مشكلة كبيرة بسبب التركيزات العالية للعديد من الملوثات الضارة والخطرة، وخاصة المعادن الثقيلة (HMs). أدى التلوث بالمعادن الثقيلة وعواقبه على صحة الإنسان والبيئة إلى زيادة الاهتمام بتطوير تقنية معالجة منخفضة التكلفة وطويلة الأجل. لإزالة المعادن الثقيلة، يتم استخدام العديد من الأساليب الفيزيائية والكيميائية التقليدية والبيولوجية الخضراء. تتناول المقالة المرجعية هذه كل من الأساليب التقليدية والتقنية الحيوية لإزالة HMs من مياه الصرف الصحي وتقدير كفاءتها. يتم تحديد الأسلوب الأنسب لإزالة الأيونات الثقيلة من مياه الصرف الصحي من خلال عدد من العوامل، بما في ذلك تكلفة التشغيل، وتركيز أيون المعدن الأولي، والأثر البيئي، وقيم الأس الهيدروجيني، والمواد الكيميائية المضافة، وكفاءة الإزالة، والجدوى الاقتصادية. تشمل هذه الطرق معالجات الامتزاز باستخدام مواد ماصة مختلفة ومعالجات غشائية ومعالجات كيميائية ومعالجات كهربائية مثل المعالجات الكهروكيميائية. بشكل عام، تعد الطرق الكيميائية والامتزاز والغشاء هي الطريقة الأكثر عملية التي تم تناولها في الأدبيات. لقد لوحظ وجود فجوة معرفية واضحة في أداء طرق المعالجة لإزالة أيونات المعادن الثقيلة من مياه الصرف الصحي الحقيقية لأن معظم الدراسات استخدمت مياه الصرف الصناعي التي يوجد فيها نوع واحد أو عدد قليل من المعادن. وفقًا لذلك، يجب إجراء بحث إضافي باستخدام مياه الصرف الصحي الحقيقية لمعالجة الملوثات المختلفة. يجب إجراء المزيد من الأبحاث حول إدخال مواد فعالة من حيث التكلفة وطرق لإزالة المعادن الثقيلة من مياه الصرف الصحي. لا يزال يجري تطوير أفضل التقنيات لاستعادة المعادن مع الحد الأدنى من التأثير البيئي والتكلفة المنخفضة وينبغي النظر فيها في البحوث المستقبلية.

الكلمات الإسترشادية: المعادن الثقيلة، مياه الصرف الصحي، الامتصاص الحيوي، مصادر المياه، الامتزاز.

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