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A Comprehensive Review of the Development of Nano-bio Adsorbents for the Separation of Heavy Metals from Wastewater

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Abstract: **The need for water is growing, and this has made wastewater treatment necessary. The amount of wastewater has grown recently during the last 2-3 decades. Although it requires an expensive material, commercial adsorbent is widely utilized in the industry for the treatment of wastewater. The demand for a method that could successfully and safely remove heavy metal ions from contaminated water. It is imperative to evaluate all potential sources of natural adsorbents with a focus on high capacity of adsorption at a reasonable cost. Despite financial limitations, cost-effective and efficient methods must be provided to control the wastewater treatment process. Adsorption is generally used because of its adaptability in both design and operation, reversibility, and affordability. However, it seems that choosing the right and eco-friendly adsorbents for heavy metal removal is becoming ever more important. This review makes an effort to give a thorough overview of various modified adsorbents, and their efficiency. The nano-biomaterial has demonstrated remarkable attention for the reduction of these heavy metals from wastewater. The development of low-cost substitutes that were economically viable for the treatment of various types of wastewater has been the major goal of this review paper.**

***Keywords*: Wastewater, adsorption, heavy metal, adsorbent, nano-biomaterial.**

I. INTRODUCTION

Pollutants known as heavy metals are naturally occurring and also released by anthropogenic activities. Due to their abundance, stability, and toxicity in water-based solutions. It has been well known that these metal ions were extremely harmful to human life [1, 2]. Heavy metals were regarded as a severe danger to species and public health in the aquatic environment [3, 4]. There are a number of metallic elements that may be found in nature, including cobalt, vanadium, aluminium, arsenic, nickel, selenium, manganese, antimony, tantalum, zinc, tungsten, cadmium, tellurium, chromium, iron, indium, molybdenum, niobium, and selenium. The heavy metal concentrations in soil were found to be different such as for Selenium (0.0061 mg/kg), Antimony (0.11 mg/kg), Cadmium (0.25 mg/kg), Titanium (0.53 mg/kg), Mercury (0.53), Copper (0.76 mg/kg), Arsenic (1.1 mg/kg), Barium (1.9 mg/kg), Beryllium (2.6 mg/kg), Tin (3.5 mg/kg), lead (3.8 mg/kg), and Molybdenum (4.5 mg/kg), correspondingly [5-8]. According to World Health Organization guidelines, different metals had different concentrations of heavy metals in groundwater, such as iron (1.0 mg/l), copper (2.0 mg/l), arsenic (1.0 mg/l), barium (0.01 mg/l), antimony (0.07 mg/l), tin (0.05 mg/l), zinc (0.003 mg/l), lead (0.01 mg/l), nickel (0.05 mg/l), chromium (0.005 mg/l), mercury (0.01mg/l), cadmium (0.4 mg/l) and manganese (0.4 mg/l), respectively [9-16]. Due to these metals, the limited water solubility, surface area, and poor recovery, their removal is still significantly difficult when employing pure natural biopolymers. Biopolymers are very effective, environmentally acceptable surfactant adsorption compounds. Suitable composites may be synthesized quickly and readily to increase efficiency. It has a maximum adsorption capacity compared with conventional adsorbents. Biocompatible and non-toxic, as the majority of them are employed in the food sector. They can also be used to remove heavy metals and refractory organic contaminants after surfactant adsorption. However, for large-scale use, more study is necessary [17]. Since biopolymeric materials have a large porosity in the structure and surface area, strong reactivity, and weak solubility in water, there has been interest in producing composite materials using biopolymers and other components [18-22]. The heavy metals were retained and accumulated in tissues, they cause lung cancer, nausea, renal failure, neurological problems, skin conditions liver failure, and other consequences [23, 24]. As a result, it is imperative that they can be removed and separated to the lowest possible level [25-32]. In the agriculture sector, heavy metal contamination was creating an environmental problem in many countries. The major sources of soil contamination due to heavy metals include pesticides, organic compounds, paints, waste generation by industrial (including small and medium enterprises) fig (1), and mining activities. The restoration of heavy metal-contaminated soils was critical, provided that they may cause direct and indirect harm to living organisms and the environment [33-44]. The literature shows that several treatment techniques can be used to remove heavy metals from wastewater prior to discharging wastewater in rivers, and seas or for use in agriculture purposes. Adsorption is one of these techniques that are most frequently employed to remove residues of heavy metals because it is easy to apply, adaptable, and reversible. This method may create hazardous chemical sludge, were commonly expensive and inefficient, and an effective at concentrations below 100 mg/L [45-46]. Bio-sorption has gained popularity in recent years for the removal of heavy metals due to its accessibility, efficacy, and environmental sensitivity. Bacterial cells were one type of microorganism used to reduce material from water-based solutions. Because of their physical cellular interface, they have a large surface area and high metal ion bio-sorption capability.

One of the bio-adsorbents that cleans water of toxins like heavy metals and organic compounds is algae. About 4 million tonnes of algae were generated in 1990, of which 1.25, 2.5, and 0.15 million tonnes were produced as red, brown, and green algae respectively. Currently, human consumption of green and red algae in Asia is 0.5 & 66.5% respectively and 33% of marine culture produces around 94% of the world's edible algae. Algae were suitable for the removal of contaminants and may be identified and produced economically and in abundant quantity in seawater [43-44]. Nanoparticles have a high surface-to-volume ratio, making them particularly relevant as a platform for biotechnological applications. Bio-nano interactions aim to improve the capability of nanoparticles to target complex systems [45-46]. Brown algal cell walls contain 10- 40% molecules of alginic acid.

However, because of its low cost, high effectiveness, and simplicity of use, the adsorption techniques were believed to be the best method for removing even small quantities of heavy metal ions from effluents [47-51]. Heavy metals may be removed from wastewater using conventional sorbents, but their limited sorption efficiencies and capacities prevent their use in concentrated solutions. To effectively remove heavy metals from wastewater that has been polluted with metals, several effective adsorbents have been produced [52-58].

Around 1 billion people, according to the WHO, lack access to clean drinking water, and that figure is rising daily. As a result of interaction with harmful metals, millions of individuals were suffering from various diseases [59-62]. To ensure our sustainable growth on earth, we must create a straightforward process for wastewater reclamation employing inexpensive adsorbent materials.

The main aim of this review is to study the effects of adsorption and opportunities to remove heavy metals from nano-bio adsorbents. Metal oxide found in wastewater has many unique characteristics, including a high removal capacity and selectivity for heavy metals. Hence they have a significant deal of potential as effective heavy metal adsorbents. Flotation, adsorption, ion exchange, and electrochemical deposition are just a few of the traditional methods used to remove heavy metals from wastewater. The most popular method for removing heavy metals from inorganic wastewater is chemical precipitation [54-58].

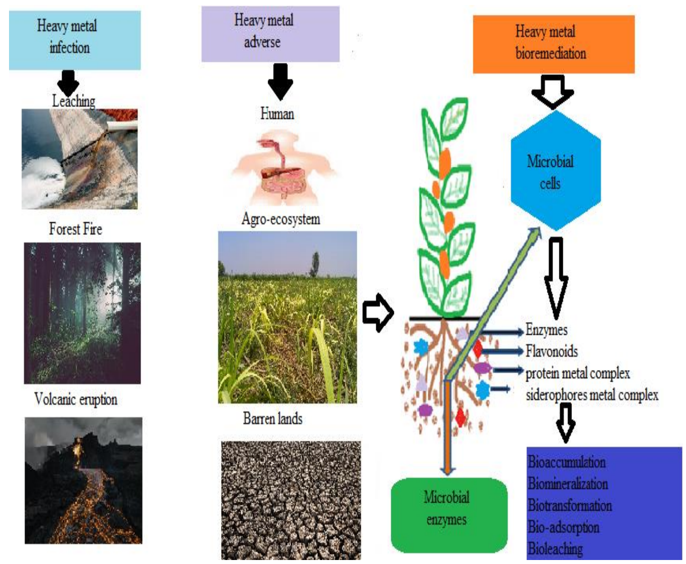

Fig 1: Sources and effects of heavy metals [55]

Current Wastewater Treatment Technologies

Since a vital natural resource is water, it is very much essential to avoid pollution. It is crucial to avoid pollution by organic and inorganic contaminants as it is significantly affected by the local flora and wildlife. However, many of the technologies utilized for this purpose discharge by-products or secondary toxins that deteriorate the environment's pollution. Therefore, there is a need for technologies that can effectively and economically treat wastewater [61-64]. In the last few decades, a researcher has been more interested in water reuse through industrial wastewater recycling. Due to the abundance of nutrients found in recycled wastewater, farming activities can greatly benefit from it [65-68]. Therefore, it is important to take great care when treating wastewater and applying it to agriculture to guarantee that it is cost-effective, ecologically sustainable, and boosts agricultural output [67-70]. Heavy and metalloids with densities above 4 g cm^{-3} fall into the category of trace elements and were called potentially toxic elements or PTEs. Lead (Pb), Silver (Ag), Iron (Fe), Cobalt (Co), Arsenic (As), Manganese (Mn), and Aluminium (Al) are some of them. [67]. These PTEs are present in wastewater which is not appropriately treated before being released into freshwater resources, which has a variety of negative consequences on environmental and health issues. Additionally, the crops occupy these PTEs and other plant species, and aquatic organisms before entering the human food chain and spoiling people's health [71-78]. In addition to natural and anthropogenic sources, there are two main categories of wastewater contaminants: organic and inorganic pollutants. Examples of organic contaminants were phenols, herbicides, pesticides, oils, dyes, proteins, starches, and pharmaceuticals, while chemical fertilizers, phenyltriethoxysilane, and excess nutrients were examples of inorganic contaminants. They lead to a decline in water quality and significant environmental issues [79-80]. Various techniques have been investigated to reduce the environmental and health risks posed by wastewater.

TABLE 1
Processes for Treating Wastewater and Potential Drawbacks

Treatment	Disadvantage	Remark
Ion switching	Partial removal of certain ions at a significant cost	On the exchange sites, ions held by electrostatic forces interchange with metal ions from diluted solutions.
Adsorption	Exhaust air disposal, plant fouling, adsorbent corrosion, and chemical regeneration requirements	When a gas or liquid solution gathers on the surface of an adsorbent, a molecular or atomic film is produced.
Inorganic precipitation	Additional operational costs associated with sludge formation	When coagulants like alum, lime, iron salts, and other organic polymer products are introduced, metal ions precipitate.
Reverse osmosis	High energy consumption resulting from membrane maintenance and pumping pressure	A semi-permeable barrier separates metal ions at a pressure greater than the internal osmotic pressure caused by the dissolved metals.
Electrodialysis	High operating costs brought on by membrane fouling and energy use	Semi-permeable ion-selective membranes are used to segregate metal ions from other ions. Cells of concentrated and diluted salts are created as a result of the separation of cations and anions caused by an electrical potential between the two electrodes.
Coagulation/ flocculation	High operating costs are brought on by the use of chemicals and the production of more sludge.	Coagulant causes colloidal particles to unite into tiny aggregates known as "flocks." These flocks draw suspended particles to themselves
Flotation	High startup costs, high operating and maintenance costs	To separate solids or scattered liquid phases, use a bubble attachment.

Pollutants in wastewater were removed using a variety of wastewater treatment techniques. Primary, secondary, and tertiary treatment were a few of these techniques. Preliminary treatment includes filtration and sedimentation of heavier particles that include waste from domestic, governmental, and industrial sources. Wastewater is treated aerobically and anaerobically in lagoons or ponds as part of secondary treatment. For pollutants that may be handled, this process uses microbial activity, degradability, and biotransformation chemical treatment methods used in tertiary treatment including membrane filtration, ion exchange, flocculation, and coagulation [79-82]. Due to its cheap cost and straightforward operation, adsorption continues to be the most efficient technology for removing contaminants from wastewater and water among the various current water treatment processes [83-86]. (Table 1).

Adsorbents Mechanism

Heavy metals were persistently accumulating in the environment because they were stable and non-biodegradable [87-88]. All of the interactions between the particles were evenly balanced inside the adsorbent, on the surface, where there aren't as many identical atoms or molecules, the forces are imbalanced or still present. The physical adsorption of heavy metals by biochar or activated carbon is often mediated by one of four mechanisms like cation exchange, complexation of surface functional groups, surface precipitation, or pore structure-dominated physical adsorption. [89-90]. The interactions between the adsorbent surface and adsorbate molecules under physisorption and chemisorption were illustrated in fig (2) (a) and (b) respectively. Chemical bonds between organic compounds and carbon nanotubes have occasionally been described as a mode of interaction. Nevertheless, it is feasible to do so by modifying them physically, chemically, or by administering additional medications. The adherence of atoms, ions, or molecules from a gas, liquid, or dissolved soil to the surface of the adsorbent is known as adsorption.

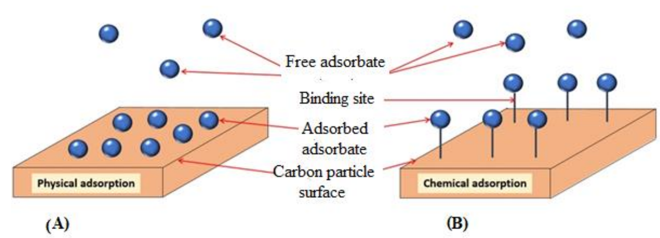

Fig 2: Adsorbents Interaction Mechanism [91]

Table 2 shows the adsorption capacity of some heavy metals in industrial waste. Typical by-products such as fly ash, blast furnace sludge, lignin, pulp waste, red mud, and sludge have historically been used as effective adsorbents because of their higher adsorption capacity [91, 92].

Nano-Bio materials and Nano-Bio composites

Nanomaterials that were synthesized using biomolecules or that employ biomolecules to encapsulate or immobilize another form of nanomaterial were referred to as bionanomaterials. Biomolecules from bacteria, plants, agricultural waste, insects, marine life, and some animals were used to synthesize bionanomaterials. Nano-biomaterials were an effective tool for medical applications due to their compatibility and novel effects brought on by the nanoscale. Innovative solutions in several sectors have made it possible to include biomaterials in the growth of nanostructures [123-128]. The manufacture of multipurpose nano indications based on biomaterials has garnered considerable interest. Due to their simplicity, fine tenability, and continued compliance, many nano-bio materials were excellent choices for biomedical applications in the current world.

TABLE 2
Adsorption Capacity of some Heavy Metals in Industrial Waste

Metals	Adsorbent	Adsorption Capacity (mg/g)	References
Zn(II)	Used-up powered sludge Olive-related industrial solid waste	168 5.40	[87-88] [89-90]
Ni(II)	Red earth Slag from a blast furnace Olive stone fragments	160 2.13	[91-92]
Cu(II)	Areca waste (AW) Waste slurry	1.12 640	[93-94] [95-97]
Cr(VI)	Iron(III) hydroxide	0.47	[98-100]
Cr(III)	Lignin	17.97	[101-102]
Hg(II)	Garbage sludge Garbage sludge	560 15.73	[103-104] [105-106]
Cd(II)	Orgabosolv lignin Lignin	1.10 1865	[107-108] [109-110]
Pb(II)	Tea industry waste	2	[111-112]
Fe(III)	Tea industry waste	24	[113-114]
As(III)	Zr (IV)-loaded orange waste	130	[115-116] [117-118]
As(V)	Leather industry waste Zr (IV)-loaded orange waste	88.0 26	[119-120]
V(V)	Metal waste sludge	24.8	[121-122]

More research on composite materials, particularly bio and nano-composites, is required in light of recent developments in the field of materials science because these materials were much better able to meet current needs for facilitating technological advancement than monolithic materials [129-132]. To enhance the use of composite adsorbent technology, current research by earlier investigators on the use of nano- and bio-composites was very much essential. The bio-composites were composite materials that include one or more biologically derived phases. These materials commonly include plant fibers like used paper or salvaged wood, cotton, hemp, and flax as well as leftovers from food crops. Recently, researchers have become more interested in bio-composites that can be strengthened with biopolymers or natural fibers [133-134]. Bio-nano composites were a special category of hybrid nanostructured materials with novel structural and functional characteristics that may be used in a variety of technological applications, such as those in the medical, food, industrial, ecological, and agricultural fields. The development of bio-nanocomposites with enhanced mechanical, thermal, and functional properties based on different nanoparticle fillers or matrix materials is of interest to many investigators [135-140]. Amorphous silica nanoparticles with diameters between 15 and 60 nm were produced by using a technique described by Jang et al [141]. The first description of the base-catalyzed Stöber hydrolysis and condensation of TEOS (Tetraethyl orthosilicate) for the production of ASNs was made by Jang et al. [141]. This technique, as well as its variations, has been used frequently to create monodisperse ASNs. In this technique they synthesized mesoporous silica nanoparticles with nanoscale, mesopores using a modified Stöber approach by employing triethanolamine and cetyltrimethylammonium chloride as surfactants to build sacrificial micelles that template pores inside ASNs.

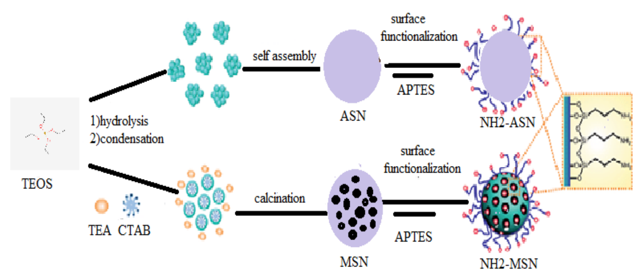

Fig 3: Schematic diagram of the synthesis of amorphous silica nanoparticle (ASN), mesoporous silica nanoparticle (MSN), and amino propyl-functionalized ASN and MSN [141].

By including a surfactant template in the reaction mixture and calcining the resultant particles, the one-step Stöber method may be modified to produce porous silica. Biopolymer-based technologies were frequently referred to as "green technology" due to their biocompatibility and biodegradability. These were particularly relevant to agricultural, medical, and food packaging technologies [141-145]. Table 3 shows the removal of heavy metal by biopolymer and nano-bio composites.

II. Conclusion

Nano-bio adsorbents were an increasingly important natural adsorbent with a high capacity for adsorption at the affordable cost of heavy metal removal from wastewater. To regulate wastewater treatment, cost-effective solutions must be offered because zinc is used in several industries. The removal of zinc from wastewater is a highly researched topic. It shows that advanced technology for wastewater treatment can be used which removes pollutants through physical, chemical, and biological processes as well as by using inexpensive adsorbents prepared from easily accessible natural resources. Nano biomaterials were utilized to substitute biological systems, and they may be used to produce durable materials. The low-cost adsorbents must undergo a systematic technical analysis to ascertain their overall process effectiveness and adsorption capacity before they can be used commercially. The demand for freshwater resources was rising worldwide due to the rapid growth in population, industrialization, and extensive agricultural methods. Several investigators have obtained and produced nanotechnology-based methods for the purification of water and wastewater from industrial effluents. The interaction between biomass components and the adsorption process, as well as other bio-sorption and removal processes, require more investigation. Access to biomass-derived adsorbents makes it simple and affordable to remove pollutants from the environment. Future research is anticipated to disclose improved and more targeted biosorbents and processes. However, it seems that finding the ideal and most environmentally friendly adsorbents for removing heavy metals is becoming more and more crucial. This paper attempts to provide a detailed overview of several modified adsorbents, their effectiveness, and nano biocomposites as alternative adsorbents.

TABLE 3

Removal of Heavy Metal by Biopolymer and Nano-bio Composites

Heavy Metal	Biopolymer	Nanoparticles	Metal removal mechanism	Adsorption Capacity(mg/g) Removal%	References
AS(III)	Chitosan	Iron oxyhydroxide	Adsorption	99.6 mg/g	[142]
Cu(II)	Chitosan Carboxymethyl	Fe^0	Adsorption	250.0 mg/g	[142, 143]
Zn(II)	Chitosan	(CNT)	Adsorption	96%	[142,144]
Cd(II)	Chitosan	Fe_3O_4	Adsorption	32.2mg/kg	[145]
Cr(VI)	Caroxymethyl cellulose	Fe^0	Reduction	94%	[146]
Pb(II)	Alginate	Magnetite	Adsorption	95.3%	[147]
Cr(III)	Poly(y-glutamic acid)	Fe_3O_4	Adsorption	24.60 mg/g	[145]
As(V)	Cellulose	Iron oxyhydroxide	Adsorption	33.2 mg/g	[146]
Hg(II)	Aqueous	Sillica	--	55.87 mg/g	[148]
Ni(II)	Pelletize	SiO_2	Adsorption	227.20	[149, 150]
Cr(IV)	Cellulose	-----	Adsorption	24.28	[151]

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