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Original Research

Removal of Heavy Metal: Activated Carbon, Metal-organic Frameworks Adsorbents Chemical Precipitation, Ultrasonic Removal of Heavy Metals and another Conventional Technology

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Abstract:

It is crucial to remove heavy metal ions from wastewater in order to maintain a clean environment and protect human health. There were a variety of approaches detailed for removing heavy metal ions from wastewater. Some possible categories for these approaches are those based on adsorption, membranes, chemicals, electricity, and photocatalysis. It has been noted that the majority of the recent research has been on adsorption methods. The capacity to simultaneously remove many ion types, a long retention duration, and cycling stability of adsorbents are the main challenges of the adsorption processes. While both chemical and membrane approaches have their uses, there are significant challenges with chemical procedures, including as the creation of enormous volumes of sludge and the necessity for post-treatment processes. A further improvement in membrane separation could be achieved by inhibiting fouling and scaling. Additional expenses, however, are associated with pre-treatment and frequent membrane cleaning. Although electrical-based approaches were also found to be efficient, addressing the problem of large-volume sludge formation and achieving separation on an industrial scale are also necessary. Currently, technologies that rely on electricity or photocatalysis are not as developed. Research on heavy metals removal should prioritise the use of actual wastewaters over synthetic ones. Sustainable, low-cost, and environmentally friendly materials and processes should be the centre of future study.

Keywords: Heavy Metal, Activated Carbon, Adsorbents, Chemical Precipitation

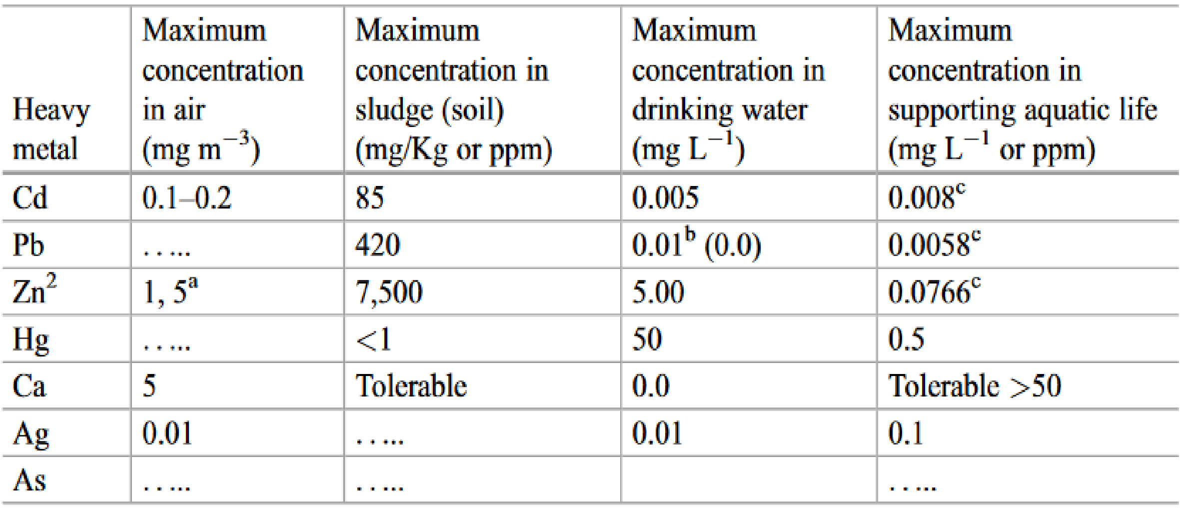
Introduction:

"Heavy metals" are any metals with a high density and a high potential for toxicity, even at trace amounts. The phrase "heavy metals" is used to describe a class of substances that includes metalloids and metals having an atomic density higher than 4 g cm 3 [1]. Despite long-established knowledge of heavy metals' negative health consequences, exposure to these pollutants persists and is even on the rise in many regions of the world. Lead (Pb), cadmium (Cd), zinc (Zn), mercury (Hg), arsenic (As), silver (Ag), chromium (Cr), copper (Cu), and iron (Fe) are all substances that are considered heavy metals. Heavy metals pollute the air (for example, during combustion, extraction, and processing), water (via runoff and releases from storage and transportation), and soil (through a variety of other processes and pathways). When thinking about potential threats to human health, atmospheric contamination is usually at the top of the list. The majority of lead emissions are from vehicles on the road and are thus dispersed evenly over the atmosphere [2, 3]. Anthropogenic mercury emissions are distributed geographically according to the amount of coal consumed in various areas, while cadmium emissions are mostly linked to fuel combustion and nonferrous metallurgy.

Heavy metal ion removal methods have recently been the subject of concentrated research. These methods include electrocoagulation (EC), adsorption with both synthetic and natural adsorbents, the utilisation of magnetic fields, enhanced oxidation processes, membranes, and more. These analyses weighed the pros and cons of a particular approach to wastewater treatment, one of which involved the removal of heavy metals [4-6]. We still don't have a full picture of how to remove heavy metals from wastewater resources.

So, to effectively remove heavy metal ions from wastewater, this evaluation covers all the existing technologies in great detail and provides critical feedback on each. Also, additional operating variables, beginning concentration, removal efficiency, chemicals added/adsorbents, and optimal treatment pH value should be considered while choosing the best relevant procedure. Treatments based on adsorption, membranes, chemicals, electricity, and photocatalysis are all included in this review. The methods are evaluated individually. The Supplementary Information file is a list of all the research that were reported in the literature. It includes details about the operating circumstances, removal efficiency, and noteworthy remarks for each approach. The accessibility of each method's operating and performance factors dictates the selection of literature study. In addition, the acute toxicity, nonbiodegradability, and accumulation of high quantities of metallic compounds into the world's aquatic ecosystems have been inevitable outcomes of the discharge of heavy metal wastes into wastewater over the last several decades [7, 8]. Lead (Pb $^{2+}$), cadmium (Cd $^{2+}$), copper (Cu $^{2+}$), and iron (Fe $^{2+}$) are present in high concentrations and can be toxic for a long time in industrial runoffs from a variety of sources, including printing and pigments, tanneries, oil refining, mining smelting, electroplating, paintings, and, more recently, e-wastes. Consequently, the effects of these heavy metals on human health have been the subject of substantial research, and organisations like the World Health Organisation (WHO) routinely assess this literature. The US Environmental Protection Agency (EPA) has set maximum allowable limits of 0.006, 0.01, 0.25, and 0.1 mg L 1 for these heavy metal ions (Pb $^{2+}$, Cd $^{2+}$, Cu $^{2+}$, and Fe $^{2+}$) in drinking water and inland surface water, respectively (Table 1) and Figure 1.

Table 1. Air, soil, and water heavy metal pollution limits set by the US Environmental Protection Agency (EPA).



Heavy metal	Maximum concentration in air (mg m^{-3})	Maximum concentration in sludge (soil) (mg/Kg or ppm)	Maximum concentration in drinking water (mg L^{-1})	Maximum concentration in supporting aquatic life (mg L^{-1} or ppm)
Cd	0.1–0.2	85	0.005	0.008^{c}
Pb	. . ...	420	0.01^{b} (0.0)	0.0058^{c}
Zn^{2}	1, 5^{a}	7,500	5.00	0.0766^{c}
Hg	. . ...	<1	50	0.5
Ca	5	Tolerable	0.0	Tolerable >50
Ag	0.01	. . ...	0.01	0.1
As	. . ...	. . ...		. . ...

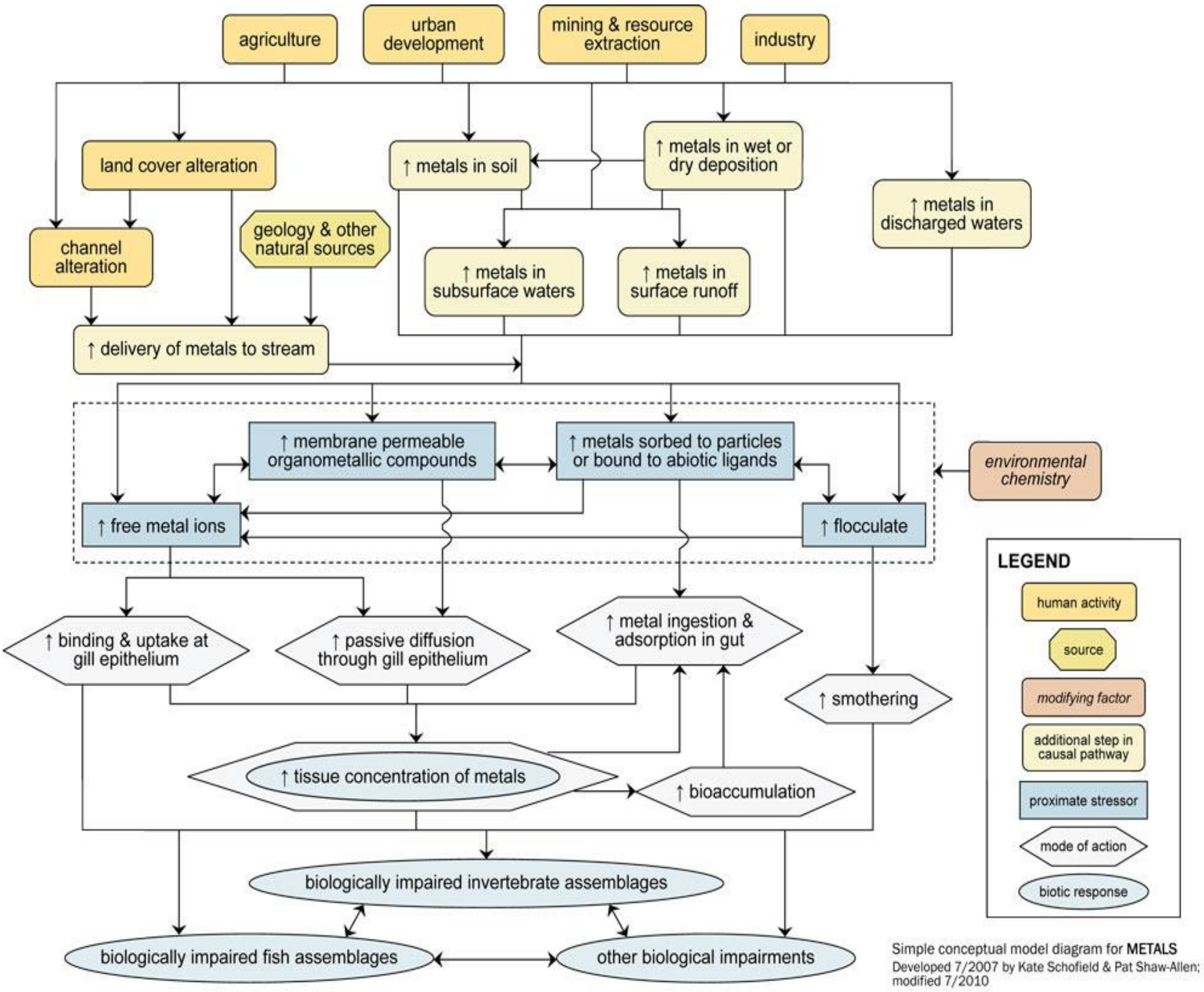


Figure 1. A basic schematic showing the relationships between metals and their effects, from their sources to the diseases they cause. To see a larger version of the diagram, click on it.

Particularly harmful to both humans and ecosystems are the byproducts of industrial processes, which in turn cause severe environmental damage [5]. Precious metals (Pd, Pt, Ag, Au, Ru, etc.), hazardous metals (Hg, Cr, Pb, Zn, Cu, Ni, Cd, As, Co, Sn, etc.), and radionuclides (U, Th, Ra, Am, etc.) are the three groups into which these heavy metals are supposedly classified for the sake of ease of analysis. Physical, chemical, and biological procedures are the mainstays of heavy metal ion removal from water. Some examples of conventional physiochemical procedures are filtration, ion exchange, electrochemical treatment, membrane technology, chemical precipitation, oxidation/reduction, filtering, and evaporation recovery. The majority of these methods are either not effective or too costly when the concentrations of metals are less than 100 mg L-1 [9-11]. The second category includes the more expensive physical processes, such as ion exchange, activated carbon, and membrane adsorption, which are particularly relevant when dealing with high volumes of wastewater [12, 13]. The use of ultrasonic technology for the management and removal of heavy metals has recently attracted some interest. In addition, the removal capability is enhanced when adsorbent materials are coupled with ultrasound. The concentration of heavy metal ions in solution can be reduced from parts per million to parts per billion using the adsorbent materials' intrinsic metal-sequestering properties. Ultrasound is a promising method for treating complex wastewaters with both huge volumes and low concentrations because of its ability to efficiently remove dissolved heavy metal ions from complex solutions, regardless of how dilute they may be.

Standard Methods for Heavy Metal Removal

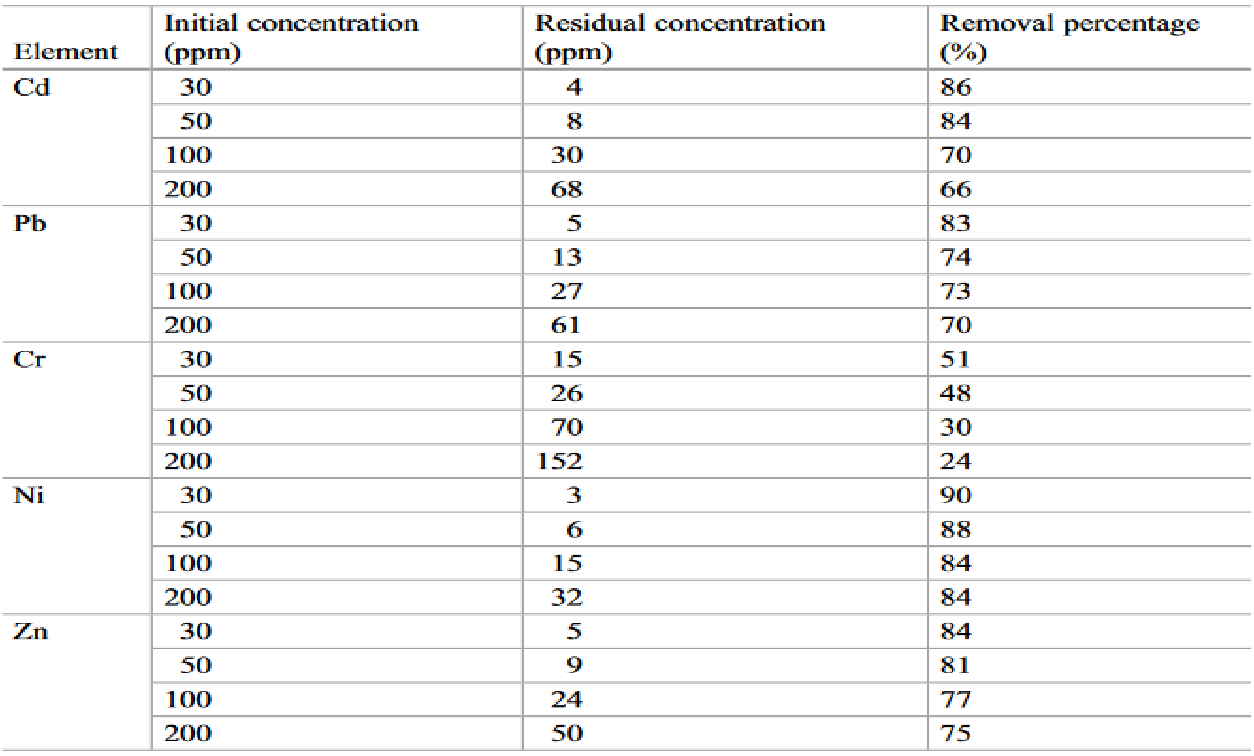
To remove heavy metals, conventional physicochemical approaches have been widely employed. These methods include activated carbon, precipitation, reverse osmosis, membrane separation, chemical oxidation, and ionic exchange. Initial metal concentrations in the range of 10-100 mg L 1 make most of those methods less effective, too costly, and environmentally harmful [14-16]. When the concentration of metal ions in an aqueous solution is between 1 and 100 mg L 1, for example, chemical precipitation and electrochemical treatment are ineffective and produce an enormous amount of sludge that is even more difficult to treat.

The following is an exhaustive list of the benefits and drawbacks of these traditional approaches:

Carbon Enzyme

This amorphous, or haphazard, kind of graphite is extremely porous and displays a wide variety of pore sizes in the form of apparent fissures, pits, and slits that are molecular in size. Heavy metals in industrial wastewater can be effectively removed by using activated carbon as an adsorbent due to its high specific surface area, microporous nature, and chemical surface nature. Despite its benefits, it has numerous drawbacks, such as a high preparation cost, carbon loss due to deactivation [17, 18], performance type depending on carbon, lack of selectivity, etc. The selectivity can be improved with modified or activated carbon, but the subsequent preparation cost is astronomical . The clearance % is highest at low heavy metal concentration and drops off sharply as concentrations get higher. The elimination percentages for Ni, Cd, Zn, Pb, and Cr at 30 ppm are 90%, 86%, 83.6%, and 51%, respectively. The clearance percentages of nickel, cadmium, zinc, lead, and chromium are highest at 30 ppm, although this is true across all concentrations. However, the clearance percentages of the heavy metals under study were 66%, 70%, 24%, 84%, and 75% for Cd, Pb, Cr, Ni, and Zn, respectively, at the highest heavy metal concentration of 200 ppm. While other metals show lower removal percentages at all concentrations, nickel displays the highest removal percentage across the board.

Table 2. The percentages of metals removed with activated carbon at varying beginning concentrations.



Element	Initial concentration (ppm)	Residual concentration (ppm)	Removal percentage (%)
Cd	30	4	86
	50	8	84
	100	30	70
	200	68	66
Pb	30	5	83
	50	13	74
	100	27	73
	200	61	70
Cr	30	15	51
	50	26	48
	100	70	30
	200	152	24
Ni	30	3	90
	50	6	88
	100	15	84
	200	32	84
Zn	30	5	84
	50	9	81
	100	24	77
	200	50	75

Adsorbents made of Chitosan

The presence of amino ($-NH_2$) and hydroxyl (-OH) groups gives the natural adsorptive polymer chitosan (CS) an affinity for contaminants in wastewaters. It has some interesting characteristics, however the regeneration is ineffective due to its weak mechanical strength and unstable properties. The limited porosity, high crystallinity, resistance to mass transfer, and low surface area of CS make it difficult to work with whether it is in powder or flake form. To get around these problems, people have suggested structural and chemical changes. By creating bridges between the polymer [19-21] chains and the functional groups, cross-linking chemical modification increases the strength of CS. But this method decreases uptake. Another chemical modification process called grafting increases the adsorption capacity of CS significantly by covalently connecting functional groups (such as amines and hydroxyls) to its backbone. The adsorption capacity, mechanical strength, and thermal stability of CS can be improved by combining it with other adsorbent materials, according to one proposal . To create adsorbents with a high selectivity for the 19 target metal ions, the ion-imprinting process was employed. Supplementary In order to remove heavy metal ions from wastewater. Possible causes of this behaviour include a highly stable link between the metal ions and the adsorbent surface, poor mechanical strength [23-28], incomplete desorption, a decline in the effective interaction between the adsorbate and the adsorbent, and a lack of available adsorption sites. To improve the CSs' reusability, new regeneration methods and adjustments should be suggested.

Electromagnetic sorbents

One type of material matrix that can hold iron particles, often in the form of magnetic nanoparticles like Fe_3O_4, is called a magnetic adsorbent. Potential building blocks include biomass, polymers, starch, carbon, or CS. Magnetic field, surface charge, and redox activity properties all have an impact on the adsorption process, as shown in Figure 2. They demonstrated an inexpensive, easily synthesised, exceptionally charged surface, and reusable material. Magnetite (Fe_3O_4), zero-valent iron nanoparticles (ZVI NPs), and iron oxides (hematite, maghemite, and spinel ferrites) are among the several magnetic adsorbents that have been suggested in studies. Several characteristics, including surface morphology and adsorbent magnetic behaviour, determine the sorption process's mechanism and kinetics. Additional experimental factors that impact them include pH, irradiation duration, adsorbent concentration, wastewater temperature [29, 30], and beginning pollutant dosage. Adsorbents containing iron particles are highly effective in removing metal ions from effluent. Heavy metal ion removal via coating Fe_3O_4 particles has been the subject of some research. The grafting procedure was deemed the best option because of its adaptability and simplicity. Nevertheless, the quantity of active functional groups and the active hydroxyl on the surface of Fe_3O_4 particles have a significant impact. One obstacle to commercializing this technology is that the adsorbents that were synthesized are not sufficiently cycle stable

The adsorbents made on metal-organic frameworks

In reticular synthesis, organic linkers are firmly linked to metal ions, resulting in metal-organic frameworks (MOFs). Scientists suggested a vast array of MOFs. The majority of the organic ligands utilised to construct numerous MOFs are both highly costly and highly hazardous, as has been noted. The hydrolytic-thermal stability and ease of incorporation of functional groups—such as amines, carboxylic acids, hydroxyl groups, and oxygen—make zirconium-MOFs (such UiO-66) a potentially useful nanostructure material for sorption applications. 36 or through the process of cross-linking. It is possible to increase the adsorption capacity of MOFs even more by using composite-based adsorbents. Table 7 provides a list of the heavy metal ions that various MOFs were able to absorb from wastewater [31-35]. Although MOFs have many desirable properties and are effective in removing heavy metal ions, some of the metals that are intended to be bound cannot pass through their micropores. The majority of them also don't hold up well when submerged. The majority of MOFs formed using Mn, Fe, and Cu had low chemical stability. Hence, in order to apply MOFs in industrial wastewater applications, additional study is required to fine-tune their structure and scale them up. In order to improve the stability and sorption kinetics of MOFs, it is necessary to suggest and implement various functionalization approaches.

Separation by Chemical Means

Heavy metal removal from inorganic wastewater is a common application of this method. Metal hydroxide is the usual form that precipitates out of solutions [39]. Due to the widespread availability of lime or calcium hydroxide, the most popular precipitant agent, this procedure can be applied globally. As an additional choice, lime precipitation is a reliable method for the easy treatment of inorganic wastewater containing metal concentrations greater than 1,000 mg L 1. Example: in a batch continuous system, it was useful for removing heavy metals like Zn(II), Cd(II), and Mn(II) cations with initial metal concentrations of 450, 150, and 1,085 mg L 1, respectively. Chemical precipitation has significant benefits, but it takes a lot of chemicals to get the metal concentration down to a safe level for disposal. Additionally, it is typically not applicable to wastewater with metal ion concentrations below 100 mg L 1. Its delayed metal precipitation [36, 37], rising sludge disposal costs, excessive sludge formation necessitating additional treatment, and long-term environmental effects of sludge disposal are additional downsides. Lime is the most popular base for hydroxide precipitation in industrial settings due to its low cost and simplicity of handling, however other hydroxides have also been employed to remove metals from wastewater.

Reducing and Oxidising Chemically

Incorporating oxidising or reducing chemicals into wastewater treatment processes converts harmful compounds into less harmful ones. According to certain reports, potassium ferrate (VI) can be used for oxidation and coprecipitation to successfully remove heavy metals such Mn2+, Cu2+, Pb2+, Cd2+, Cr3+, and Hg2+. Nevertheless, at now, powerful processes often employ chemical oxidation and reduction as a pretreatment for wastewater.

Sorting Through Membranes

Electrodialysis (ED), nano-filtration (NF), ultrafiltration (UF), and reverse osmosis (RO) are some of the membrane separation methods that have gained a lot of attention for treating inorganic effluent. These methods can remove inorganic contaminants like heavy metals, in addition to organic compounds and suspended solids. Despite its potential usefulness for situations involving low concentrations of metal ions [38], Tzanetakis et al. show that at starting metal concentrations of 0.84 mg L 1 and 11.72 mg L 1, respectively, the removal efficiencies of Co(II) were 90% and 69%. Inadequate selectivity, excessive energy consumption, and underdeveloped technology are the key drawbacks of these techniques that prevent them from being widely used

Heavy Metal Elimination via Ultrasound

Sonochemistry refers to the use of ultrasound in chemical processes and reactions. Because of its unique characteristics, sonochemical processes have been the centre of much research and development activity as of late. The process of acoustic cavitation is what makes sonochemical effects possible in liquids. Acoustic cavitation, a nonlinear phenomenon that concentrates the diffuse sound energy in liquids, is the source of ultrasound's chemical effects. Acoustic cavitation creates extremely high-energy circumstances, which in turn produce radicals and create this high-energy microenvironment. Bubbles in a liquid can form, expand, and eventually burst through a process called cavitation. Intense local heating, huge pressures, tremendous heating and cooling rates, and liquid jet streams travelling at approximately 400 km/h are all results of cavitation collapse.

The produced hotspots can achieve heating and cooling rates more than 10^10 K s^1, pressures greater than 1,000 atm, and temperatures greater than 5,000 K. This method is one-of-a-kind for a number of uses because the circumstances it produces are different from those of more traditional approaches, such as photochemistry, wet chemistry, hydrothermal synthesis, and flame pyrolysis [39, 40]. It is possible that these concentrated hotspots have an effective duration of just a few microseconds.

There are three areas where chemical reactions take place in aqueous sonochemistry:

(1) The bubble that forms during cavitation Within burst bubbles, where pressure and temperature are extremely high for short periods of time, chemical processes can place, producing gaseous byproducts characteristic of pyrolysis and burning.

(2) The second type of interface is the bubble/solution interface, which refers to the areas where cavitation bubbles and bulk solution meet at a high temperature below that of the bubble core. Accumulated nonvolatile solutes in this area have the potential to decompose thermally, triggering radical reactions. The thickness of this reaction zone at the interface is approximately 200 nm.

If the radicals generated inside the bubble and in the interfacial area make it through the migration to the bulk solution, they can react with the solutes there. When the transmembrane operating pressure (TMP) is low, ultrafiltration (UF) can be applied. It is possible to attach additives to metal ions in order to make them larger, since the pores of UF membranes may be bigger than the heavy metal ions themselves. The result is the suggestion of PEUF and MEUF, which stand for polymer enhanced ultrafiltration and micellar enhanced ultrafiltration, respectively. When UF and surfactant are bound together, MEUF is produced. MEUF requires less space, has a high removal efficiency, and uses less energy due to its high selectivity and flux. Wastewater with low amounts of heavy metals is ideal for MEUF. The MEUF process involves adding a surfactant to wastewater at a concentration greater than the CMC. The assembly and increased production of some micelles in the solution is facilitated by surfactant monomers beyond CMC. A hydrophilic head and a hydrophobic tail make up the surfactant. Due to electrostatic interactions, the surface of the micelles adsorbs counter metal ions, while the inner hydrophobic core solubilizes organic substances with low molecular weight. The greatest retentions are typically achieved by surfactants, whose electric charge is diametrically opposed to that of the metal ions. For efficient heavy metals extraction, materials such as polyelectrolytes (PE), cationic surfactants, and anionic surfactants like sodium dodecyl sulphate (SDS) are utilised. Figure 8 in the supplemental materials provides a synopsis of research on the MEUF procedure. Several factors affect MEUF performance. These include the solutes added, the surfactant type, the operating circumstances, and the membrane type. The combination of UF with binding polymers results in PEUF. Chelating or ionic bonds hold the functional groups of the bonding polymers together, which might be sulfonate, phosphonic, carboxylated, or amine. PEUFs go by a few different names: complexation enhanced ultra-filtrations, size improved, polymer-assisted, polymer-supported, and polymer-supported. The PEUF technique removes polymer-bonded metal ions while allowing water and simple components to pass through the membrane pores.

Electrodialysis

Ion separation by means of an electric potential difference is known as electrodialysis (ED). The ionic solutes are separated in ED by means of a sequence of anion exchange membranes (AEM) and cation exchange membranes (CEM), which are alternately stacked in parallel. As part of the ED process, cations go through CEM and anions through AEM. Half of the ED stack channels would be used to produce the treated stream (dilute), while the other half would be used to discharge the concentrated stream. ED is able to function across a broad pH range, has good water recovery, and does not undergo phase shift, reactions, or chemical involvement. But ED also has membrane fouling, expensive membranes, and electric potential demands. It has been achieved an extraction efficiency of 96.9% for Ni^{2+}, 99.9% for

Pb $^{2+}$, and 99.9% for K+ by using ED to separate these ions from a synthetic solution using a new ED heterogeneous CEM (made of 2-acrylamido-2-methyl propane sulfonic acid-based hydrogel and PVC), respectively. The recovery of Pb $^{2+}$ was achieved with a maximum separation efficiency of almost 100% using a batch ED method. Extractions of Cu $^{2+}$, Ni $^{2+}$, trace amounts of Cd $^{2+}$, Fe $^{3+}$, Cr $^{6+}$, and Zn $^{2+}$ were also achieved using a pilot-scale ED system, with a removal rate that surpassed 90%. With an efficiency of 91.38%, ED was able to extract As $^{3+}$ and As $^{5+}$ from metallurgical effluent.

Forming clumps and clots

The process of flocculation involves the aggregation of particles that have become unstable, whereas coagulation involves the neutralisation of forces that keep colloids separated. The neutralisation of ion charges is accomplished by employing traditional coagulants such as ferrous sulphate, ferric chloride, and aluminium. Thanks to flocculants like polyaluminum chloride (PAC), polyferric sulphate (PFS), polyacrylamide (PAM), and others, particles can be bonded together to form huge agglomerates. Even though PE flocculation was touted as one of the more feasible options, the resulting sludge could have harmful effects . Flocculants are often synthetic and do not break down in the environment. Figure 5 shows the process, including the sedimentation. The growing colour of the effluent, the inefficiency when using natural coagulants, the complexity of scaling up, the toxicity and health hazards of inorganic coagulants, and the vast volume of sludge are some of the drawbacks.. Heavy metals such as Cu$^{2+,}$ Pb$^{2+,}$ and Ni $^{2+}$ are commonly removed using this approach. Efficient removal of other metals including As $^{2+}$, Se $^{2+}$, Cr $^{2+}$, Sb $^{3+}$, Sb $^{5+}$, and Ag $^{2+}$ is also possible.

Conclusion:

Considerations such as operational cost, environmental impact, pH values, chemicals added, removal efficiency, economic feasibility, and beginning concentration of metal ions are crucial in determining the best method for heavy ion removal from wastewater. The methods can be categorised as follows: adsorption treatments (using various adsorbents such as carbon-based, carbon-composites, minerals, CS, magnetic, biosorbents, and MOFs), membrane treatments (such as UF, nanofiltration, microfiltration, reverse osmosis, forward osmosis, and electro-dialysis), chemical treatments (such as chemical precipitation, coagulation-flocculation, and flotation), electric treatments (such as electrochemical (reduction, EC, EF, and advanced oxidation) and ion exchange), and photocatalysis. Because of its low cost of reusability, high removal rate, strong application, and ease of operation, adsorption has been extensively studied as the most promising approach for removing heavy metal ions from wastewater. Nevertheless, this inclination is mostly reliant on the selection of inexpensive materials, significant absorption, and effective regeneration procedures. Though more developed and practical than adsorption, the membrane approach still faces challenges in minimising separation costs and fouling. There is a high level of technical maturity and practicality with the chemical-based approaches, particularly with chemical precipitation. Additionally, these procedures are thought of as being economical. In contrast to the electrochemical approach, which takes into account extra variables like electrodes, electrical energy, and fixed costs, these depend only on the chemical that is consumed. But they require sedimentation separation and produce a lot of sludge. Due to the passivation of electrodes and the high electrical energy consumption, the electrochemical process is a rather expensive technology. In addition, the electric methods are among the most immature technologies, with the exception of photocatalytic ones. The photocatalytic process is advantageous because it is environmentally benign since it uses no chemicals or very little chemicals and produces very little sludge. These days, scientists are devoting a lot of time and energy to finding new and better ways to filter water for heavy metals. Ultrasonic technique has demonstrated promise in the potential removal of heavy metals from water-based solutions and sludges. Furthermore, a cost-effective and environmentally benign method for heavy metal removal from polluted environments has been developed using a combination of ultrasonic waves and adsorbent materials. The elimination of heavy metal ions, such arsenic ions, is facilitated by OH radicals produced by acoustic cavitation. Heavy metal pollutants can be effectively adsorbed with the help of ultrasound, thanks to the physical effects of the ultrasound and the cavitation process. The observed improvement in heavy metal removal is due to increased mass transfer caused by the physical effects of cavitation. No industrial-scale methods for heavy metal removal using ultrasound have been established, despite the availability of lab-scale studies in the literature. If we want to see this technology implemented on a wide scale, we need to do more optimisation work and build bespoke ultrasonic equipment.

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