

Techniques for remediation of Mercury(II) from wastewater-A Review

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ABSTRACT

The toxicity of heavy metal ions released by industrial effluents is a huge concern to public health. Heavy metal ions are toxic even at trace level concentrations and can accumulate in living organisms, causing several disorders and diseases. One of the most potential among them is the toxic Hg(II). Mercury contamination in water is a widespread problem, not just locally but also globally. This article reviews the available literature on the remediation of Hg(II) from wastewater by various methods.

KEYWORDS: Heavy metal ions, Hg(II), adsorbents, wastewater

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I. INTRODUCTION

Though there has been plenty of water on earth, fresh water has not always been available when and where it is needed, nor is it always of suitable quality for all uses. Of recent origin the world has witnessed industrial spurge and industrialization has led to inferior quality of water sources. Contamination of heavy metal ions from industrial effluents makes water unsuitable for use. Hence there is an urgent need of elimination of the toxic heavy metal ions from wastewater.

Heavy metals are generally considered to be those whose density exceeds 5 g per cubic centimeter. A large number of elements fall into this category. Mercury(Hg) is a d-block element with an atomic number 80 and is in liquid form under standard conditions. Mercury is found in deposits of mercuric sulfide called cinnabar. Mercury pollution is caused by pharmaceutical industries, pulp and paper preservation, caustic

soda production industry, agriculture industry, etc. Mercury poisoning is called pink disease also known as acrodynia. Elevated exposure levels of mercury in any form can damage the kidneys, brain, developing fetus, etc. Mercury exposure can also cause lung damage, skin rashes, memory problems, and hair loss[1]. The permissible limit of Hg(II) in potable water is 0.002 mg/L as per USEPA (United States Environment Protection Agency) standard[2] and 0.001 mg/L as per Indian standard[3]. So it is highly imperative to decontaminate it from wastewater.

A number of studies on mercury(II) removal from wastewater have been reported. This paper presents a review of the selected literature dealing with the removal of mercury(II) using various techniques.

II. VARIOUS TECHNIQUES IN HEAVY METALS REMOVAL FROM WASTE WATER

The conventional processes for removing heavy metal ions from wastewater includes chemical precipitation, flotation, adsorption, membrane filtration, ion exchange etc. In chemical precipitation, adjustment of pH to the basic conditions (pH 9–11) is the major parameter that significantly improves heavy metal ions removal. Lime and limestone are the most commonly employed precipitant agents due to their availability and low-cost in most countries. Lime precipitation can be employed to effectively treat inorganic effluent with metal ions concentration of higher than 1000 mg/L. Other advantages of using lime precipitation include the simplicity of the process, inexpensive equipment requirement, and convenient and safe operations. However, chemical precipitation requires a large amount of chemicals to reduce metals to an acceptable level for discharge. Other drawbacks are its excessive sludge production that requires further treatment, slow metal precipitation, poor settling, the aggregation of metal precipitates, and the long-term environmental impacts of sludge disposal [4].

Ion exchange is another method used successfully in the industry for the removal of heavy metal ions from effluent. An ion exchanger is a solid capable of exchanging either cations or anions from the surrounding materials. Commonly used matrices for ion exchange are synthetic organic ion exchange resins. The disadvantage of this method is that it cannot handle concentrated metal ions solution as the matrix gets easily fouled by organics and other solids in the wastewater. Moreover ion exchange is nonselective and is highly sensitive to the pH of the solution. Electrolytic recovery or electro-winning is one of the many technologies used to remove metals from process water streams. This process uses electricity to pass a current through an aqueous metal-bearing solution containing a cathode plate and an insoluble anode. Positively charged metal ions cling to the negatively charged cathodes leaving behind a metal deposit that is strippable and recoverable. A noticeable disadvantage is that corrosion could become a significant limiting factor, where electrodes would frequently have to be replaced [5].

Electrocoagulation has been proposed as another effective method to treat various wastewaters such as landfill leachate, restaurant wastewater, tar sand and oil shale wastewater

urban wastewater, laundry wastewater and chemical mechanical polishing wastewater. Electrocoagulation produces by anodic dissolution followed by hydrolysis, aluminum or iron hydroxide flocs which destabilise and aggregate the suspended particles or precipitates and absorb dissolved contaminants. It has some significant advantages namely; simple equipment and easy operation and automation, a shorter retention time, high sedimentation velocities, more easily dewatered, reduced amount of sludge due to the lower water content [6].

Membrane filtration is a technique that can be used for the removal of contaminants from water. Membranes are synthetic materials with pores acting as selective barriers, which do not allow some constituents of the water to pass through. A driving force, such as pressure difference between the feed and the permeate sides, is needed to transport the water through the membrane. Generally, there are two categories of pressure-driven membrane filtrations: (i) low-pressure membrane processes, such as microfiltration and ultrafiltration and (ii) high-pressure membrane processes, such as reverse osmosis and nanofiltration [7].

Adsorption is the process where molecules are concentrated on the surface of the sorbent. The molecules go from the bulk phase to being adsorbed in the pores in a semiliquid state. The driving force for adsorption is the ratio of the concentration to the solubility of the compound. A variety of natural and synthetic materials has been used as sorbents, including activated carbons, biological materials, zeolites, chitosan, and industrial wastes. Adsorption offers significant advantages like low cost, availability, profitability, easy operation and efficiency, in comparison with other conventional methods especially from economical and environmental points of view [8].

Flotation is yet another separation process, based on the introduction of gas bubbles as the transport medium. Suspended particulate matter, being hydrophobic or conditioned to be so, is then attached to the bubbles and moves toward the water solution surface, contrary to the direction of gravity. Ion flotation involves the removal of surface-inactive ions from aqueous solutions by adding surfactants or collectors, usually with an ion having a charge opposite to that of the metal ion to be removed. Dilute aqueous solutions has been extensively investigated in the laboratory with batch-scale and continuous mode experiments by applying the sorptive flotation technique. This

method involves the preliminary abstraction or scavenging of metal ions using proper "sorbents", which exist at the fine or ultrafine particle-size range, followed by a subsequent flotation stage for the separation of metal-loaded sorbent particles from the treated solution[9]. In view of the above, several methods such as adsorption, chemical precipitation, reverse osmosis, ion-exchange, flotation etc have been reported for heavy metals removal. Table I presents the advantages and disadvantages of mercury removal techniques[10].

III. LITERATURE REVIEW

Satish I. Chaturvedi(2013) studied the removal of mercury from mercury containing waste water prepared synthetically by using sodium chloride as an electrolyte using sacrificial aluminum anode in a batch wise by electrocoagulation cell. The wastewater was obtained from a tank containing a mixture of exhaust dyeing solutions at a textile factory. The effect of applied potential, initial pH, initial concentration of solution, agitation, electrolyte concentration and energy consumption on percent removal of mercury have been investigated. The removal efficiency of mercury was 98.5% under optimum condition in which solution pH was 4.5, applied potential 9 V, initial solution concentration 50 ppm, electrolyte concentration 1.333 g/ lit. with a stirring speed 400 rpm[11].

Andrea. J. Santana et al.(2016) investigated powders obtained from the peel of the fruit of *Pachira aquatica* Aubl, in its in natura and/or acidified form, as an adsorbent for the removal of mercury ions in aqueous solution. The materials were characterized by Fourier Transform Infrared Spectroscopy and Thermogravimetric analysis. Batch experiments were done. The adsorption process was evaluated using Cold-Vapor Atomic Fluorescence Spectrometry and Cold-Vapor Atomic Absorption Spectrometry. The adsorption isotherm model, Langmuir-Freundlich, based on the performances best represented the adsorption process, and the maximum adsorption capacity was predicted at 0.71 and 0.58 mg g⁻¹ at 25°C in nature and acidified, respectively. The pseudo-second order model showed the best correlation to the experimental data. The presence of the metal ions interference Cd(II), Pb(II), Cu(II) in examined concentrations, on the adsorption of Hg in biomass, in its in natura and acidified form, generally was unaffected. For adsorption on real aqueous wastewater sample the % removal of Hg(II) was as 69.6 % for biomass acidified and 76.3 % for biomass in nature. [12]

Qing-Zhou Zhai(2014) prepared nanometer α -Al₂O₃ powders by solid state method which was used as the adsorbent and powder X-ray diffraction and scanning electron microscopy were used to characterize the prepared powders. Mercury stock standard solution was prepared from mercury nitrate. Mercury(II) content determination was accomplished on a Spectrophotometer Effects of pH, adsorption contact time, initial concentration of Hg(II), adsorbent dosage and effect of temperature was analysed. The optimal adsorption conditions for the initial Hg(II) concentration of 0.80 mg/ml, α -Al₂O₃ amount : Hg(II) amount (g/g) = 4.17, pH = 3.5, contact time 10 min, temperature 20°C. The effects of desorption of three type desorptive agents CH₃COOH, HNO₃, HCl, were probed. The optimal desorption agent was 0.10 mol/L HCl. When desorption was 10 min, the highest desorption rate was 97.92%. The maximum adsorptive amount of Hg(II) was 157.8 mg Hg(II) /g (α -Al₂O₃) [13].

Erhayem, M., Al-Tohami et al.(2015) dealt with the equilibrium adsorption of Hg(II) onto carbonized *Rosmarinus officinalis* leaves as adsorbent from aqueous solution. Samples were prepared by physical carbonization at 773 K for 1 h. Titration method was used to determine the concentration of Hg(II) before and after adsorption by ethylenediaminetetraacetic acid, EDTA, as chelating agent. Batch equilibrium studies were carried out under different experimental conditions such as Hg(II) concentration and temperature. The equilibrium sorption data were analyzed using Freundlich, Langmuir, Dubinin-Ra-dushkevich (DRK) and Temkin isotherms. The experimental results were found to fit the Langmuir isotherm model with a monolayer adsorption capacity of 588.2 mg/g at 318 K, while they were found to fit the Freundlich isotherm model at 298 K. The thermodynamic studies revealed that the process is spontaneous in nature and exothermic [14].

Jeriffa De Clercq(2012) analysed the performance of two adsorbents, a stable mesoporous adsorbent, i.e. a thiol containing ethene bridged periodic mesoporous organosilica SH-ePMO and a commercial ion exchange resin TP-214, for the removal of mercury from aqueous solutions. Batch adsorption studies were done. The operating variables studied were initial mercury concentration and contact time The adsorption isotherms were studied using Langmuir and Freundlich models. The Langmuir model yielded the best fit for the SH-ePMO, whereas the Freundlich model fitted best the adsorption on

TP-214. The maximum adsorption capacities were 66 and 456 mg/g for SH-ePMO and TP-214, respectively. The lower capacity of SH-ePMO was attributed to its small amount of thiol groups. The pseudo-first-order kinetic model showed a good description of the experimental data of both adsorbents. TP-214 was capable of purifying water to parts per trillion levels. [15].

E Vélez et al(2016), developed Fe_3O_4 and $\gamma\text{-Fe}_2\text{O}_3$ nanoparticles using iron salts and NaOH as precipitation agents, and Aloe Vera as stabilizing agent. Aloe Vera was used as stabilizing agent because of its low cost, and non-toxicity. Iron oxide nanoparticles are efficient adsorbents because they combine magnetic separation with ionic exchange capacity for heavy metals removal. The nanoparticles were characterized by three different measurements: using a Zetasizer Nano ZS for their size estimation, UV-visible spectroscopy which showed the existence of resonance of plasmon at $\lambda_{\text{max}} \sim 360$ nm, and by Scanning Electron Microscopy (SEM) to determine nanoparticles form. At any concentration value of iron oxide nanoparticles, the removal percentage of Hg(II) was above 60%, therefore it was not necessary to use high concentrations of nanoparticles for effective removal of mercury. Mercury removal of 70% approximately reported was confirmed by atomic absorption spectroscopy measurements [16].

Rahmanzadeh L et al(2016) assessed the adsorbent, magnetite-polyrhodanine core-shell nanoparticles for removing Hg(II) from aqueous solution. Different parameters like, effect of pH, initial Hg(II) concentration, initial adsorbent concentration and contact time on the efficiency of Hg(II) removal were investigated by batch experiments. The maximum adsorption capacity was obtained at pH=6.5 and 25°C with 10 g L⁻¹ nano adsorbent. The kinetic data of adsorption of Hg(II) ion on the synthesized adsorbent were best described by a pseudo-second-order equation, indicating their chemical adsorption. The Freundlich, Langmuir and Temkin isotherms were used for modeling and modeled best by the Freundlich isotherm in whole concentration range. The maximum adsorption capacity obtained was 29.14 mg g⁻¹ [17].

Shaojun Huang et al.(2016) applied Poly(1-amino-5-chloroanthraquinone) nanofibrils as nanoadsorbents for mercury removal from aqueous solutions. A series of batch adsorption experiments were conducted to study the effect of adsorbent dose, pH, contact time, and metal

concentration on Hg(II) uptake by nanofibrils. Kinetic data indicated that the adsorption process of nanofibrils for Hg(II) achieved equilibrium within 2 h following a pseudo-second-order rate equation. The adsorption mechanism was investigated by Fourier transform-infrared (FT-IR) spectra and X-ray photoelectron spectroscopy (XPS) analyses. Five consecutive adsorption-desorption cycles were done without considerable changes in the adsorption capacity to evaluate the practicability of the nanosorbent for purifying actual wastewater, a batch experiment was conducted on chloralkali wastewater. Nanoadsorbent exhibited a very high capability for adsorption of mercury ions (98.4%) from chloralkali wastewater. The adsorption isotherm of Hg(II) fitted well the Langmuir model, exhibiting adsorption capacity of 3.846 mmol of metal per gram of adsorbent [18].

Kamyar Yaghmaeian et al.(2015) revealed that carbon nanotubes which have tiny pores with uniform size and also wide specific surface have been used to remove pollutants. Multi-walled carbon nanotubes were used as sorbent to remove mercury from aqueous solution using batch technique. Cold vapour inductively coupled plasma optical emission spectrometry was used to determine the amount of mercury in solution. Effect of pH, contact time and initial concentration of mercury were studied. The best pH for adsorption was about 7. The rate of adsorption process initially was rapid but it was gradually reduced with increasing of contact time and reached the equilibrium after 120 min. More than 85 % of initial concentration of 0.1 mg/L was removed at 0.5 g/L concentration of sorbent and contact time of 120 min. Adsorption process followed the pseudo second order model and the adsorption isotherms was described by both the Freundlich and the Langmuir models [19].

George Z. Kyzas et al(2013) investigated two modified chitosan derivatives prepared in order to compare their adsorption properties for Hg(II) removal from aqueous solutions. One chitosan adsorbent is only cross-linked with glutaraldehyde, while the other, which is magnetic, is cross-linked with glutaraldehyde and functionalized with magnetic nanoparticles (Fe_3O_4). Many possible interactions between materials and Hg(II) were observed after adsorption and explained via characterization with various techniques (SEM/EDAX, FTIR, XRD, DTG, DTA, VSM, swelling tests). The adsorption evaluation was done studying various parameters as the effect of pH (optimum value 5 for adsorption and 2 for

desorption), contact time (fitting to pseudo-first, -second order and Elovich equations), temperature (isotherms at 25, 45, 65 °C). The maximum adsorption capacity (fitting with Langmuir and

Freundlich model) of at 25 °C was 145 and 152 mg/g, respectively[20].The summary of the above literature is given as Table 2.

Table 1. Advantages and disadvantages of mercury removal techniques[10]

Removal techniques	Advantages	Disadvantages
Ion exchange	Fast kinetics. High capacity of treatment. High removal efficiency	Resins synthetic is costly. Serious secondary pollution is caused by regeneration of the resins. Waste products are produced Selectivity is low.
Adsorption	Wide pH range. Low cost. Metal binding capacities are high. Easy operation conditions.	Waste products are produced. Selectivity is low.
Chemical precipitation	Operation is simple. Capital cost is low.	Generation of sludge. Sludge disposal needs extra operational costs
Membrane filtration	Separation selectivity is high. Requires small space and low pressure.	Membrane fouling is expensive. Process is complex. Permeate flux is low
Flotation	High metal selectivity and removal efficiency. More concentrated sludge is produced.	High initial capital cost, maintenance, and operation cost.

Table 2. The summary of the above literature

Authors name	Method of removal of Hg(II)	Optimum pH	Concentration range studied	%Removal/Adsorption capacity
Satish I. Chaturvedi(2013)	Electrocoagulation cell	4.5	30 - 100 ppm	98.5
Andrea. J. Santana <i>et al.</i> (2016)	Adsorption- Peel of fruit of Pachira aquatic Aubl in natura and acidified form	4	50 µg L ⁻¹	For real aqueous wastewater samples(dental waste)69.6% for biomass acidified ad 76.3% for biomass in nature
Qing-Zhou Zhai(2014)	Adsorption-nanometer α-Al ₂ O ₃ powders	3.5	0.1, 0.3, 0.5, 0.8, 1.0 mg/mL	Maximum adsorptive amount is 157.8 mg Hg(II) /g (α-Al ₂ O ₃)
Erhayem, M., Al-Tohami <i>et al.</i> (2015)	Adsorption-Rosmarinus officinalis leaves	2.4	100-400mg/L	588.2 mg/g
Jeriffa De Clercq(2012)	Adsorption and Ion exchange (mesoporous organosilica SH-ePMO and a commercial ion exchange resin) TP-214,	-	0-900 ppm	66 and 456 mg/g for SH-ePMO and TP-214
E Vélez <i>et</i>	Adsorption-Fe ₃ O ₄ and γ-Fe ₂ O ₃	-	4 to 50 mg/L	More than 87%(70%

al(2016).	nanoparticles			average)
Rahmanzadeh L et al(2016)	Adsorption- magnetite-polyrhodanine core- shell nanoparticles	6.5	50 to 300 mg/L	29.14 mg g-1
Shaojun Huang et al.(2016)	Adsorption-Poly(1-amino-5-chloroanthraquinone) (PACA) nanofibrils	6	10 mmol L-1	3.846 mmol of metal per gram of adsorbent
Kamyar Yaghmaeian et al.(2015)	Adsorption-Multiwalled carbon nano tube	7	0.1 to 10 mg/L	25.641 mg/g
George Z. Kyzas et al(2013)	Adsorption-chitosan adsorbent is only cross-linked with glutaraldehyde, while the other, which is magnetic, is cross-linked with glutaraldehyde and functionalized with magnetic nanoparticles (Fe ₃ O ₄).	5	100 mg/L	145 and 152 mg/g,

This review has established that the removal of the mercury(II) by the different methodologies are highly effective and promising results have been reported based on the adsorption capacity and % removal. It is evident from the literature survey that adsorption method is widely used over conventional methods and different types of adsorbents have been employed by the researchers for the effective removal of Hg(II).

IV. CONCLUSION

In this paper, an attempt has been made to review various techniques of mercury(II) removal from wastewater which established that adsorption has been used widely for the remediation of mercury(II) from wastewater. The future works can be added for the removal of mercury(II) from the onsite industrial effluents. These studies could be extended for analyzing the effects of other heavy metal ions present in the aqueous solution.

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