REMOVAL OF CHROMIUM FROM WASTEWATER USING LOW COST ACTIVATED CARBON BY PLANT BASED NANO PARTICLE DERIVED FROM NEEM LEAVES

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Abstract:

Chromium is a heavy metal that can be harmful to human health. It is often found in wastewater from industrial processes, such as tanning and electroplating. Neem leaves are a natural material that has been shown to be effective in removing chromium from wastewater. The present research focuses on exploring the effectiveness of neem leaves (Azadirachta indica) as a sustainable and low-cost adsorbent for the removal of Chromium from wastewater.

From many studies it was found that a low-cost activated carbon was prepared from neem leaves, characterized and utilized for the removal of chromium from wastewater. The results indicate that neem leaves exhibit substantial potential in adsorbing Chromium from wastewater. The optimum conditions for maximum Chromium removal were determined, and the adsorption mechanism was analyzed. In many studies it was seen that, the waste neem leaves (Azadirachta indica) gained visible attraction for neem leaf–activated carbon (NLAC) preparation in the thermal process by using ammonium carbonate as an activating agent. Using NLAC, a nanocomposite of NLAC/Fe_2O_3 was synthesized by hydrothermal techniques for Cr(VI) elimination from the aqueous solution. This study highlights the promising role of neem leaves as a natural, eco-friendly and cost-effective alternative for the mitigation of Chromium pollution in wastewater treatment. From many studies we have learnt that low cost nano adsorbent derived from neem leaves has a great effect on industrial wastewater management.

Key words: Activated carbon, Chromium, wastewater treatment, Neem.

Introduction

Industrial activities, particularly in metallurgy, electroplating, and tanning, contribute significantly to the release of heavy metals, such as chromium, into wastewater. Chromium contamination in water bodies can lead to serious health issues for both humans and aquatic life. The conventional methods for heavy metal removal, such as chemical precipitation and ion exchange, are often expensive and generate secondary pollutants. As a result, the search for cost-effective and environmentally friendly solutions for wastewater treatment remains essential. Heavy metal pollution imposes an alarming effect on environment due to their toxicity at traceable levels and adverse health impacts. Some of the heavy metals include chromium, nickel, copper, mercury, arsenic, lead and cadmium. Among all these heavy metals, chromium is considered to be highly carcinogenic. Chromium is also responsible for various respiratory and reproductive disorders As a result, the discharge standards of chromium are known to be stringent. The permissible chromium concentration of chromium in natural water bodies is 0.05 mg/L. The industrial sources of chromium include tannery, electroplating, battery, dyes, varnishes, welding, metallurgy, chromate and dichromate units Many conventional techniques like ion exchange, membrane-based treatments, chemical precipitation, electrodialysis, etc. exists, for their removal. However, the high cost and energy consumption of their techniques are considered as some of the disadvantages.

Adsorption is a suitable alternative to conventional chemical treatments in terms of simplicity, ease of operation and effectiveness However, most of the adsorbents need expensive chemical modifications to improve the sorption properties. Extensive studies were reported on bio based adsorbents to treat heavy metal pollutants. But the poor stability and decreased surface area resulted in limited sorption capacity.

Activated carbon has been widely recognized for its exceptional adsorption capacity for a variety of pollutants. However, the high production cost limits its application, especially in developing countries. Thus, there is a growing interest in exploring alternative sources of activated carbon from low-cost and locally available materials.

Neem leaves (Azadirachta indica), a common tree in many tropical and subtropical regions, have been reported to possess adsorption properties due to their porous structure and chemical composition.

These studies aims to investigate the potential of neem leaf-activated carbon (NLAC) as a low-cost adsorbent for chromium removal from wastewater. By using neem leaves, it shows our concern for the environment extends beyond the allocation of budgets. We believe that if we really intend to conserve natural resources, we need to start at the grassroots level by educating our own people about it. We can use lime, alum and carbon for the purification of wastewater. As we know alum is used for purification from ancient times. Lime adsorb the impurities from wastewater and this is the simplest way to purify water. Nowadays many new techniques are used for water purification with lime, alum and carbon.

Carbon adsorption is the method that has most often been used for the removal of refractory organic compounds in wastewater. Either granular or powdered activated carbon can be used. Our primary aim is to investigate the feasibility of carbon adsorption for the treatment of chromium-containing wastewater.

Hexavalent chromium can enter our body through water and show its poisonous impact on the body. At higher levels above the most contaminant level, hexavalent chromium causes skin and stomach disturbance, ulceration, dermatitis, harm to the liver, kidney dissemination, nerve tissue harm, and death as a result of huge dosages. Hexavalent chromium is more toxic than trivalent chromium for both strong and endless exposures. Acute ingestion of large amounts of hexavalent chromium causes gastrointestinal problems, vomiting, and hemorrhage, while less exposure brings about infection of the septum, which has dangerous consequences for the respiratory tract. Public health and animal studies recommend that hexavalent chromium mixtures, especially those that are water-insoluble, are responsible for different types of DNA harm and carcinogenicity. Consequently, urgent and efficient chromium removal is essential.

The aim of this study is to get a maximum reduction of organic load and removal of suspended particles especially in terms of chromium. Through this study, we learn to use different types of charcoal for chromium removal in wastewater which are easily available economically.

On the other hand, nanomaterials have unique properties like smaller size, increased surface area and improved mechanical stability. Specifically, magnetic nanoparticles are widely known for industrial wastewater treatment due to their unique magnetic properties and low toxicity specifically many researchers have reported the use of magnetic nanoparticles for dyes and heavy metal removal Magnetite nanoparticles now find good application in sorption processes due to their value-added properties. M.J. Jacinto et al. synthesize iron oxide nanoparticles from plant extracts in eco friendly manner. Very importantly, in recent years, tremendous effort has been put in synthesizing these nanoparticles using co-precipitation techniques .The advantages of this chemical based synthesis include the simplicity of the process and the ability to generate a large quantity of nanoparticles. A review by Anil Usmani et al. explains about new trends of using nanoparticles for wastewater. It was studied that Surya Pratap Goutam et al.in 2019 synthesized nanoparticles in green synthesized manner and studied their applications However, the use of organic chemical-based solvents is employed in most of the co-precipitation based iron nanoparticles synthesis. Very recently, plant-mediated synthesis of nanoparticles received wider attention in the field of nanotechnology. For instance, Chlorella vulgaris and Chlorella pyrenoidusa were utilized to synthesize gold nanoparticles. Most of the nanoparticles synthesized using plant extract possess major bio functionalities such as glycosides phenols flavonoids and carbohydrates. These functionalities ultimately enhance their desirable physical, chemical, and mechanical properties. Another review paper with such aim was about removal of caffeine and salicylic acid in aqueous solutions by using coconut husk as an magnetic bio char adsorbent by Bruno Salarini Poexoto et al. in 2023 which shows wastewater treatment by adsorption technique. In their study, coconut husk charcoal is employed as an adsorbent to remove pollutants. The ability of coconut coir to effectively remove pollutants from industrial effluent has been found. The current study is an attempt to utilize the functional groups of the neem extract and the iron nanoparticles in the adsorption of chromium from aqueous solution. In addition, it is significant to note that, in our earlier study magnetite nanoparticles were synthesized using L. camara extract as a capping agent and a high adsorption capacity of 227.20 mg/g is obtained for nickel ions. The study also aims in investigating the functional groups, thermal stability and the morphology of the nanoparticles using several characterization techniques. The use of magnetic nanoparticles facilitates easy separation of nanoparticles from water. For the sake of low cost and simplicity, co-precipitation technique has been employed in the synthesis of neem extract coated nanoparticles.

Methods for the treatment of toxic heavy metals

Several conventional methods have been examined in water and wastewaters purification. These methods include precipitation ultrafiltration coagulation/flotation electrochemical method reverse osmosis solvent extraction and ion-exchange methods Most of these methods produce productive outcomes, but they have limitations in terms of high energy requirements, incomplete disposal, high operating costs, complex operations and the formation of toxic sludge and are unable to treat low concentration levels of metal ions in water .The adsorption technique is the most recognized method that is considered as a promising alternative for environmental pollution control and most recommended as an operative technique and more economical for eliminating various contaminants from wastewaters. Besides, the adsorbent can undergo regeneration by a proper desorption procedure. Among all the several treatment techniques, adsorption is reliable and perceived as the well-known method and globally acknowledged for the removal of trace elements from wastewaters It is flexible in design and its operating processes produce potable water

Some of the strengths and weaknesses of some conventional methods for removing heavy metals from aqueous media as cited by Athar et al. and Ahmaruzzaman, are presented in Table1.

Table 1

Benefits and shortcomings of various techniques adopted in removing heavy metals

Purification method	**Benefits**	Shortcomings
Ion–exchange	• Selects metals • Intense regeneration of materials	• Expensive • Removes fewer metals
Membrane process and ultrafiltration	• Creation of reduced solid waste • Consumption few chemicals • Proficient	• High maintenance cost • Reduced flow rates • Reduced remote performance over time • Production of concentrated sludge
Chemical coagulation	• Sludge settlement • Dewatering	• Very expensive • Consumes a large number of chemicals

Purification method	**Benefits**	Shortcomings
Electrochemical methods	• Pick out metals • Do not consume chemicals • Pure metals extraction is possible	• High operational and startup cost
Chemical precipitation	• Cheap • Removal of most metals	• Mass production of sludge • Disposal problems
Oxidation	• A rapid removal of toxic pollutants	• High energy requirement • By-product formation
Biological Treatment	• Volume reduction • Active in removing individual metals	• Need sophisticated technology
Photochemical	• Do not produce sludge	• Formation of by-products

Elimination of Cr(VI) ions using low-priced biosorbents

Bayuo et al. investigated the optimization of Cr(VI) biosorption using response surface methodology on Arachis hypogea husk. The central composite design was applied to obtain optimal values of factors impacting the decontamination of Cr(VI) from synthetic water. More so, three-parameter isotherm and kinetic studies were examined. The ANOVA results suggested that the independent factors had a significant influence on the quantity of Cr(VI) adsorbed on the Arachis hypogea husk. The optimal amount of Cr(VI) adsorbed was 2.4 mg/g at 120.0 min. The Redlich-Peterson and pseudo-second-order were the best-fitted models.

Tsegaye Adane et al. carried out a study to maximized Cr(VI) elimination from an aquatic environment with Teff husk-derived activated carbon. The influence of interaction factors was optimized by the central composite design component of the Design-Expert software (version 7.0). At an equilibrium contact time (124.2 min), pH (1.9), biosorbent dosage (20.2 g/L) and initial Cr(VI) concentration (87.8 mg/L), the maximum percent removal of Cr(VI) by the Teff husk was 95.6%. The experimental data obtained on Cr(VI) by the Teff husk agreed well with the Langmuir and pseudo-second-order models.

Belachew and Hinsene, utilized kaolin modified with cetyl trimethyl ammonium bromide (CTAB) to eliminate Cr(VI) from aqueous media. The impact of some biosorption factors on Cr(VI) depollution was studied. At the following equilibrium conditions, 180.0 min removal time, 0.1 g CTAB–kaolin dosage and 10.0 mg/L Cr(VI), Ninety-nine percent (99.0%) of Cr(VI) was removed by CTAB–kaolin. The Langmuir and Freundlich isotherms were applied to explore the biosorption of Cr(VI) onto kaolin–CTAB composites. The isotherm data gave a good representation to the Langmuir and pseudo-first-order models suggesting monolayer and chemisorption, respectively.

Dawodu et al. examined the biosorption removal of Cr(VI) from polluted effluent onto Heinsia crinita seed coat (HCSC). The consequence of several operating factors encompassing biosorbent dose, temperature, contact time, pH and initial Cr(VI) concentration on the process was studied. The study uncovered that the HCSC is an appropriate biosorbent for Cr(VI) depollution from the effluent. The maximum amount of Cr(VI) adsorbed was attained at 0.3 g dosage, pH of 2.0 and 30.0 min contact time. The experimental data showed a better representation with the pseudo-second-order and Freundlich models. The thermodynamic studies suggested endothermic, physical and spontaneous processes.

Biosorbents-derived from bagasse was employed by Aruna, Nisha Bagotia Ashok Kumar et al. for adsorptive elimination of Cr(VI) from wastewater. The consequences of diverse variables such as contact time, pH and biosorbent dose on Cr(VI) decontamination were considered. At 25.0 °C, pH 4.0, the optimum removal efficiencies of 94.6% and 98.4% were obtained for synthetic bagasse and bagasse bio-polymeric gel beads respectively. However, at 25.0 °C and pH 6.0, the maximum percent removal for activated carbon was 64.8%.

Samaraweera et al. examined the biosorption of Cr(VI) and Cr(III) ions onto NaOH-modified peel obtained from Artocarpus nobilis fruit. A batch tryouts carried out at a pH range establishes that the optimal pH for Cr(VI) and Cr(III) biosorption removal was achieved at a pH 2.0 and 5.0 correspondingly. The maximum amount of Cr(VI) and Cr(III) adsorbed on the peel of Artocarpus nobilis fruit at pH 5.0 and 2.0 was 4.9 mg/g for both ions. The kinetic data followed the pseudo-first-order model at a room temperature of 27.5 °C.

The biosorptive decontamination of Cr(VI) from aqueous media was explored by Mondal et al. onto waste mosambi peel dust by applying response surface methodology as an optimization tool. The optimization operating conditions for the elimination of Cr(VI) were as follows: 5.0 mg/L initial Cr(VI) concentration, a pH of 2.0, 0.5 g dosage, 30.0 min contact time and 150.0 rpm agitation speed. The isotherm and kinetic data were best fitted to the D-R isotherm and pseudo-second-order model while the thermodynamic elements proved that Cr(VI) biosorption is spontaneous and endothermic.

The biosorption characteristics of activated carbons to eliminate Cr(VI) from wastewater was studied by P Lakuszewicz. Alicja Puszkarewicz shows different varieties of carbons obtained using WD-Ekstra (WDA), WD-Ekstra modified by salt acid WD (HCl) and nitrogen acid WD (HNO3) were applied. The study revealed that the biosorption removal of Cr(VI) was improved with an increase in solution temperature. The elimination of Cr(VI) by the activated carbons was found to be successful at pH 2.0 while the equilibrium time for WD (HCl) and WDA were 150.0 and 270.0 min, respectively. The Freundlich isotherm showed good fitness to the equilibrium data.

A study was conducted by Manzoor et al. To eradicate Cr(VI) and Cr(III) from wastewater using immobilized biomass from corn cob. The biosorption system factors (dosage, contact time, pH and initial ion concentration were maximized. The study indicated that the amounts of Cr(VI) and Cr(III) adsorbed on the corn cob biosorbent were 277.6 and 208.6 mg/g, respectively. The equilibrium data of both ions were found to be well represented with the Langmuir and pseudo-second-order kinetic models.

Junjun Wang Daneshvar et al,examined the decontamination of Cr(VI) from water by different microalga-based substances. Considering all the materials utilized, the microalgal biochar exhibited about 100.0% removal of Cr(VI). The experimental data displayed good agreement with the pseudo-second-order kinetic and Langmuir isotherm models.

Jonas Bayuo, Parlayici and Pehlivan et al. prepared green adsorbents from cranberry (Cornus mas) kernel shell (CKS), rosehip (Rosa canina) seed shell (RSS and banana (Musa cavendishii) peel (BP). The impacts of many factors comprising adsorbent dosage, pH, contact time, temperature and initial Cr(VI) concentration on the biosorption process were verified. The results from the batch mode suggested that Cr(VI) biosorption is dependent on these process factors. The biosorption kinetic data obeyed the pseudo-second-order while the Langmuir isotherm showed compliance to the equilibrium results with Cr(VI) maximum sorption capacities as 15.2, 10.4, and 6.8 mg/g for RSS, BP and CKS, respectively.

Table 2
Adsorption potential of some other agricultural adsorbents for the decontamination of Cr(VI) ions from aqueous media

Biomaterial	Removal efficiency (%)	Reference
Oat waste	> 80.0	Gardea et al.
Modified sawdust	62.0–86.0	Garg et al.
sawdust of beech	100.0	Acar and Malkoc,
Modified rice husk Bagasse	88.9	Bishnoi et al.
Wheat Bran	> 82.0	Farajzadeh and Monji.
Coconut Husk	> 80.0	Mohan et al.
Neem leaf powder	> 96.0	Venkateswarlu et al.
Rubberwood sawdust	60.0–70.0	Karthikeyan et al.
Raw rice bran	40.0–50.0	Montanher et al.
Fly ash of bagasse	96.0–98.0	Gupta and Ali

Factors affecting biosorption of heavy metals

Alfarra et al. conducted a review on the removal of heavy metals by natural adsorbent and summarized some of the factors affecting biosorption as presented in Table3.

Table 3
Factors affecting biosorption of heavy metals

Factor	Effects
pH of solution	It improves the biosorption of positively-charged metals but then decreases that of negatively-charged metals or acidic dyes.
Adsorbent particle size	Small adsorbent size increases biosorption in the batch mode because of the higher surface area of the biosorbent. However, not good for column adsorption owing to its low mechanical strength and column clogging.
Biosorbent dosage	It reduces the amount of absorbed pollutants per unit mass of biosorbent then upsurges its percent removal.
Agitation speed	It increases the rate of biosorption of pollutants by reducing its mass transfer resistance; however, could destroy the biosorbent physical structure.
Initial pollutant concentration	It upsurges the amount of absorbed pollutants per unit mass of the biosorbent but declines its percent removal.
Temperature	It increases the biosorption of pollutants by rising the surface activity and adsorbate kinetic energy; however, could destroy the biosorbent physical structure.
Ionic strength	It reduces the biosorption removal of pollutants by competing with the solute

Characterization results

The neem extract coated iron nanoparticles were subjected to SEM analysis. The nanoparticles are seen to be highly dispersed and uniformly distributed containing cubic shaped particles. Characterizing neem leaves nanoparticles involves analyzing their physical, chemical and biological properties. The different fragments of iron oxide nanoparticles seen before

Chromium removal methods

Chromium is a heavy metal that can be harmful to human health and the environment when present in wastewater. There are several methods for removing chromium from wastewater, and the choice of method depends on factors such as the concentration of chromium, the specific form of chromium (hexavalent or trivalent), and the characteristics of the wastewater. Here are some common methods for chromium removal:

Chemical Precipitation:

Hydroxide Precipitation: Adding chemicals like lime (calcium hydroxide) or sodium hydroxide to the wastewater can raise the pH, causing chromium to precipitate as insoluble chromium hydroxide. This is effective for removing trivalent chromium.

Sulfide Precipitation: Adding chemicals like sodium sulfide or hydrogen sulfide can convert hexavalent chromium into less soluble trivalent chromium sulfide, which can then be precipitated.

Ion Exchange: Cation exchange resins can be used to remove trivalent chromium ions by exchanging them with other cations present in the resin.

Membrane Processes:

Reverse Osmosis (RO): RO is a pressure-driven membrane process that can effectively remove chromium ions from wastewater.

Nanofiltration: Nanofiltration is a membrane process with smaller pore sizes than RO, capable of removing divalent ions like chromium.

Electrocoagulation: Electrocoagulation involves applying an electric current to electrodes in the wastewater to generate coagulating agents that aid in the removal of contaminants, including chromium.

Biological Treatment: Some microorganisms have the ability to reduce hexavalent chromium to trivalent chromium, which is less toxic and more easily removable through precipitation or adsorption.

Chemical Reduction: Chemical-reducing agents like sodium metabisulfite can be used to convert hexavalent chromium to trivalent chromium, which is less soluble and can be removed by precipitation.

Advanced Oxidation Processes (AOPs): Advanced Oxidation Processes (AOPs) involve the use of powerful oxidizing agents like ozone, hydrogen peroxide, or UV radiation to break down chromium compounds into less toxic forms.

Adsorption:

Activated Carbon: Activated carbon can adsorb both hexavalent and trivalent chromium ions from wastewater. It's commonly used due to its high adsorption capacity.

Other Adsorbents: Various other materials like zeolites, clay minerals, and certain industrial byproducts can also adsorb chromium ions.

It's important to note that the choice of method should be based on the specific circumstances of the wastewater and the desired level of chromium removal. Additionally, regulatory guidelines and environmental considerations should be taken into account when selecting and implementing a chromium removal method.

Different Adsorption Techniques:

This review is focused on the different adsorption techniques for the removal of chromium ions from wastewater. In the present scenario, the adsorption technique is found to be a very suitable and economical technique for the removal of chromium from wastewater. Due to high porosity, chromium ions adhere to the surface of the adsorbent that has a high surface area. The surface charge of the adsorbent also plays an important role in the adsorption technique. There are various adsorbents present including natural adsorbents, composites, bio-sorbent, metal oxides etc. It is noticed that natural coagulants are safe for wastewater treatment. Lu et al. (2016) successfully removed hexavalent chromium from an aqueous solution using an iron electrode via the EC process. Ali et al. (2016) used acrylonitrile-grafted banana peels (GBPs) for the removal of hexavalent chlorine from wastewater. Dehghani et al. (2016) used treated waste newspaper pulp (TWNP) to remove Cr6+ from an aqueous solution using batch experiments. Some research studies showed that clay minerals are used as adsorbents.

Ahmed et al. (2019) showed sodium chlorite-modified coir coconut (SCM-CC) was useful for the removal of hexavalent chlorine. A total of 99.92% of Cr removal was removed at pH 2. Freundlich isotherm fits better here than the Langmuir isotherm. Begum et al. (2020) utilized chitosan-coated banana and areca fiber for the removal of hexavalent chromium from wastewater. Aigbe and Osibote (2020) removed hexavalent chromium from aqueous solutions utilizing the sorption technique with nanomaterials. Chakraborty (2021) used sawdust for the removal of hexavalent chromium.

Adsorption using activated carbon derived from Neem Leaves

Sankeeth. K.V, Asha Rani. N.R, from Bangalore University, showed the use of neem leaves charcoal for wastewater treatment. We can use neem bark, stem, roots and leaves as a coagulant in wastewater treatment. Here, after studying many review papers we can easily understand that we can use charcoal made from neem leaves for the removal of hexavalent chromium from wastewater and it would be cost effective also.

The neem tree has been used for centuries in traditional medicine due to its potential health benefits. Some of its medicinal uses include antibacterial and antifungal Properties, anti-inflammatory effects, antioxidant properties, improves skin and dental care, immune system support, as well as insecticidal and repellent properties.

Neem trees (Azadirachta indica) have been shown to have the ability to absorb heavy metals from the soil. The leaves, bark, and seeds of neem trees all contain compounds that can bind to heavy metals, preventing them from being released into the environment. The ability of neem trees to absorb heavy metals is due to the presence of a variety of compounds in their leaves, bark, and seeds. These compounds include:

Azadirachtin: Azadirachtin is a natural insecticide that is also known to have chelating properties. This means that it can bind to heavy metals and form complexes that are less harmful to the environment.

Tannins: Tannins are natural compounds that can also bind to heavy metals.

Saponins: Saponins are natural detergents that can help to remove heavy metals from the soil.

The use of neem trees to absorb heavy metals is a promising way to clean up polluted water and soils.

Conclusion

As hexavalent chromium is very harmful to human health and may be carcinogenic and causes skin disorders and lung tumors, so it is necessary to remove it from wastewater. Charcoal derived from Neem leaves is the easiest and cheapest way to reduce hexavalent chromium in wastewater. This process takes place by adsorption of chromium particles over neem leaves the charcoal surface. As the neem tree is easily available in tropical and subtropical regions, so this technique is economically beneficial for further research in this area.

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