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Sustainable Approach to Remediation of Hexavalent Chromium using Modified Biochar: A Review

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Abstract

Industrialization and other anthropogenic activities have contributed immensely to development, but have engendered environmental pollution by toxic contaminants. Amongst these contaminants, heavy metals are one of the major classes of concern. Chromium is a renowned heavy metal that exists in various oxidation states and is produced through industrial processes such as electroplating, textile production, paper industry, printing amongst others. The most common forms of Cr are Cr (VI) and Cr (III), with Cr (VI) being of more concern due to its toxicity, carcinogenicity and teratogenic nature. In sequel to this, various methods have been employed for the removal of Cr (VI) from wastewater, which includes physical and chemical means. More recent methods also include membrane technologies and advanced oxidation processes. One of the methods that has been well explored is adsorption. Researchers have investigated the use of innovative adsorbents documenting their inherent limitations. Biochar has been remarkable for the removal of Cr (VI) due to its unique physicochemical properties, resulting in a relatively more efficient removal when compared to most other adsorbents. The application of biochar has, however, been constrained by challenges such as limited efficiency, high cost and secondary pollution. The advancement towards the modification of biochar using various modifiers has become more promising due to the enhanced porosity, improved surface area and enhanced surface functionality. This review work therefore delves into recent advances and future direction in the use of modified biochar in the removal of Cr (VI) from wastewater.

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Key words: adsorption efficiency, adsorbent regeneration, chemical modification, sustainable remediation, water treatment.

Introduction

1.0 General overview on hexavalent chromium: chemistry, sources and toxicity

Chromium is a heavy metal and well known for its toxicity and carcinogenicity in its Cr (VI) form based on evidence from animal and human studies. It is found naturally in minerals such as chromite ($FeCr_2O_4$). It is released into the environment through the activities of various industries such as in leather processing, dye production, metallurgy, printing industry, paper production, preservatives in woods, and in the electroplating industry [1,2]. As a transition metal, chromium has different oxidation states, with Cr (III) and Cr (VI) being the most common forms in aqueous media. Amongst the various oxidation states of this metal, Cr (VI) has been reported to be of serious concern. Also, compounds of Cr (VI) such as dichromate ($Cr_2O_7^{2-}$) and chromate ($HCrO_4^-$) are highly soluble, stable and easily transported into the environment due to their high solubility in this oxidation state. The remarkable stability and difficulty in the reduction of Cr (VI) further accounts for its persistency for relatively longer periods and ease of transmission along the food chain. Exposure to chromium (VI) for a long period of time can result in health challenges such as oxidative stress, damage to DNA, damage to the immune system and impairment of the respiratory system [3]. Various techniques have been explored for the removal of Cr (VI) from different environmental matrices, some of which include coagulation, solvent extraction, ion exchange, stabilization amongst others. Adsorption using adsorbent materials has also been studied as a more reliable alternative solution. In spite of some of the recorded successes, each of these techniques has its own inherent limitations, such as energy demand, operational cost, time-consumption. The use of biochar therefore becomes a promising alternative and aligns with the advocacy for sustainable development. Biochar is a carbonized solid material with high aromaticity and low density, produced from the pyrolysis of environmental biomass in the absence of oxygen. Raw materials for biochar production include municipal waste, agricultural waste and algal biomass. This material possess remarkable adsorption efficiency for chromium (VI) due to its unique physicochemical properties such as large surface area, pH buffering ability, numerous pores and presence of charged ions and

functional groups. The efficiency of metal removal using biochar is affected by factors such as the dose employed, nature of biochar and the initial concentration of metal in the media [4,5].

The use of biochar is, however, limited due to secondary contamination, irregularity in porosity, high cost and limited efficiency. Furthermore, the broader application of the adsorbent is affected by the difficulty in separating the powdered biochar from the wastewater after treatment [6]. On this note, recent studies have explored the use of modified biochar with improved physicochemical properties through surface functionalization and modification of biochar. This review therefore focuses on the sustainable removal of Cr (VI) using modified biochar. It highlights the various strategies for modification of biochar. It discusses the various mechanisms involved. As one of its kind, it delves into recent advances and future direction in this regard (figure 1).

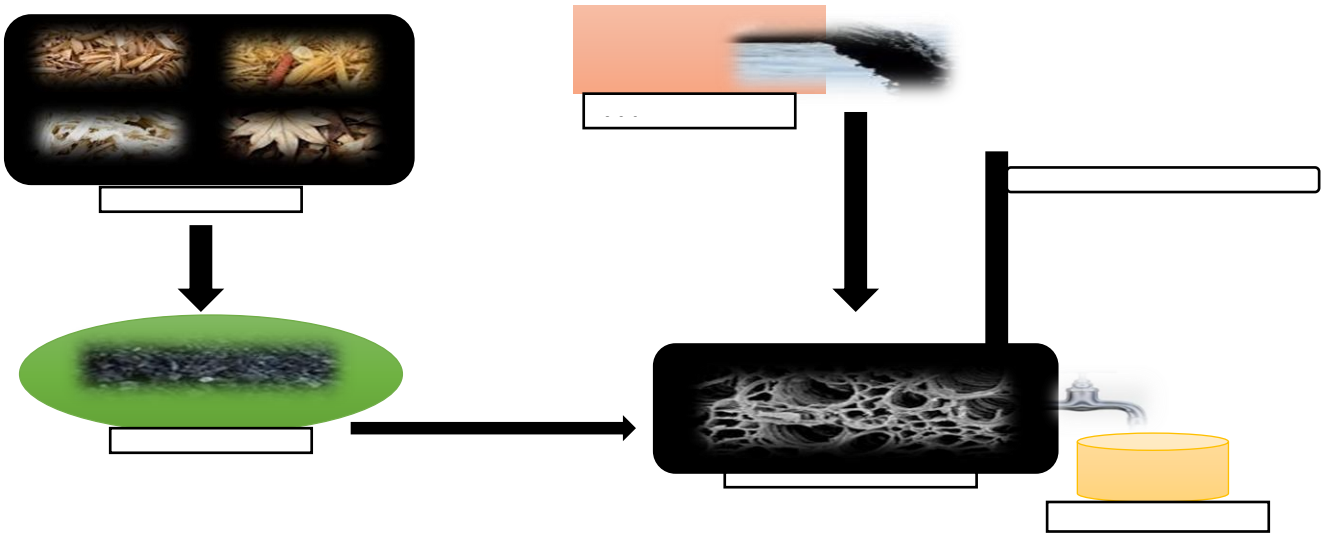


Figure 1: Schematic representation of the review work

2.0 Production of biochar

Biochar production starts with selection of the starting raw material, which can be biomass such as corn stalk, rice husk, maize straw, and other agricultural wastes. The materials are subjected to drying to reduce the moisture composition, which enhances the effectiveness of pyrolysis. Pyrolysis involves heating the biomass at temperature ranges of 300 to 700 ^{0}C in the absence of oxygen in a reactor. The material then undergoes thermal breakdown during which organic matter is

volatilized, giving rise to the solid mass rich in carbon known as biochar [7,8]. Production of biochar is affected by various process parameters. The temperature of pyrolysis affects the surface characteristics and chemical composition of the biochar produced. The presence of some functional groups such as amide, carbonyl, amines and hydroxyl groups play a remarkable role in the removal of Cr (VI) from the wastewater. At very elevated temperatures the oxygen-rich functional groups in the biochar are greatly reduced, bringing about the formation of an aromaticstructure with an improved level of carbonization. The high temperature can, however, bring about the breakdown of the morphology of the biochar and possibly block some of the pores, thereby affecting its adsorption. The rate of heating also influences the overall yield during pyrolysis. Studies have documented a decrease in the yield of biochar with higher temperature rates. Rapid rates of heating induce the breaking of the biopolymer bonds present in the biomass, which therefore leads to a decrease in the biochar yield

3.0 Strategies for modification of biochar

The modification of biochar brings about improved chemical functionality, enhanced surface area, increased site of adsorption, which promotes the interaction between the biochar and the Cr (VI). Various strategies are employed for the modification of biochar (table 1). The nature of modification also affects the mechanisms in the removal of the Cr (VI) as well as influencing the adsorption efficiency. It is therefore paramount to select the most suitable agent for the modification process. Some of the strategies for modification of biochar include the use of chemical agents such as alkaline treatment, carbondioxide modification, acid treatment, magnetic adjustment, and non-metal and metal modification. Physical methods for biochar modification include activation using carbon dioxide, ball-mining and steaming [9].

3.1 Acid treatment: The adsorption potential of biochar can be enhanced through acid treatment. This is achieved by treating the feedstock before and after pyrolysis, resulting in the production of the modified or activated biochar. In acid treatment, the biochar is immersed in acid solution, usually at a high temperature (>1200^{0}C). This process can be carried out for days during which the acid and biochar are kept in contact. The negative charge on the surface of the biochar is further enhanced when treated with acid. Furthermore, the acid modification increases the degree of disorderliness through the creation of more sites of defects in the activated biochar, which are paramount for the enhanced adsorption during the treatment of Cr (VI).

3.2 Alkaline treatment: Activation using alkali brings about enhancement in the porosity, surface area and overall surface alkalinity of the biochar. This treatment enlarges the microspores into

mesopores through the induction of carbon removal, thereby ensuring a route for adsorbent transport and boosting the adsorption efficiency. Alkaline treatment has been widely adopted at varying steps in the production of modified biochar. Alkaline treatment also reduces the hydrophilicity of the biochar, thereby making it possible for the removal of Cr (VI) through hydrophobic interaction. The post-alkaline treatment, when carried out, helps in the introduction of functional groups that are rich in oxygen, hence making the removal of Cr (VI) by the biochar more efficient. The adsorption mechanism is based on pi-electron interaction, hydrogen bonding and ion exchange. [10] carbonized cycad leaves at high temperature and subsequently treated the biochar produced using KOH.

3.3 Metal doping: The use of metal doping in the modification of biochar aids in improving the rate of adsorption of Cr (VI) and also helps in impacting magnetism. This modification is vital in the enhancement of multiple cycles, thereby boosting the removal of the Cr (VI) ions. Thus, magnetic modification makes it easier for the separation and increases the efficiency of adsorption. Some metals have the potential of using oxygen functionalities as active sites for the formation of metal centers with high stability. The introduction of the metal changes the surface functionality, dispersibility and electronegativity of the biochar. The formation of a metal-oxygen bond by the metal improves the potential for the adsorption of Cr (VI). [11] investigated the removal of Cr (VI) from wastewater using biochar from Ziziphus jujube seeds modified with iron. The study reported a 98% adsorption efficiency in the removal of the Cr (VI) from the aqueous medium. The regeneration of the adsorbent was achieved using 0.3M hydrochloric acid.

3.4 Doping with heteroatom: The doping of biochar with heteroatom improves the adsorption potential through the augmentation of the number of functional groups and surface area. Some studies have documented the use of melanine for the introduction of N atoms onto the surface of the biochar. This enhancement aids the efficient elimination of Cr (VI) and gives good recyclability. Introduction of additional nitrogen-based functional groups can be achieved through the use of raw materials that are rich in nitrogen, such as animal manure. The introduction of heteroatoms such as B, P, and N help in the surface modification of biochar composites. The groups are introduced on the surface or covalently enclosed in the carbon structure, thereby improving its performance. This modification paves way for the adjustment of volume as surface features of the carbon, bringing about alteration of chemical and physical properties and functionality. [12] modified biochar from rice husk using amide and evaluated its efficiency in removing Cr (VI) from wastewater. They

reported the optimum conditions for the removal of Cr (VI) to be pH 2, with an adsorption time of 1 hr, and adsorbent dosage of 2g/L. They reported a % removal of 97. Similarly, [3] investigated the potential of Cr (VI) removal using biochar modified using sulfur. They observed a high Cr (VI) of 92%, with the sulfur modified biochar showing significantly higher removal efficiency than the unmodified.

Table 2: A summary of various strategies for modification of biochar for Cr (VI) removal

Modification strategy	Underlying principle	Advantage	Limitation	Study using the strategy
Treatment using acid (e.g sulphuric and hydrochloric acid)	Introduction of acid group on the surface of the biochar and addition of proton	Boosts the ability of the biochar to bind to the Cr (VI)	The acid can lead to degredation of the biochar at higher concentration	
Introduction of amine group and heteroatoms	Amine groups boost the metal attracting ability of the biochar due to the enhanced affinity	A strong binding agent which increases the selectivity towards Cr (VI)	Amine containing groups are compounds of potential environmental concern	[3]
Treatment using alkaline solution (KOH and NaOH)	Enhancement of the basic groups present on the surface of the biochar	Improves the number of pores and active sites present on the biochar	May alter the surface morphology of the modified biochar	[10]
Impregnation using metals and metallic oxides	Metals such as Fe, Cu, Mg or their oxides are embedded within the biochar	Fe has the ability to introduce magnetism. Most metals increase the redox activity for the removal of Cr (VI)	There is a high tendency of the metals to leach from the biochar over time reuse.	[11]
Composite formation through blending with other materials	Integrating the biochar with other materials with natural affinity for Cr (VI) such as clay minerals	Increase the adsorption efficiency through a synergistic process	Synthesis of biochar composites could be a challenging process	[13]

Integration with polymeric materials	Presence of such groups increase the functionality on the biochar surface	The mechanical properties and selectivity of the biochar is enhanced	There is potential challenges due to stability of the modified biochar	[14]

4.0 Principle and mechanisms involved in removal of Cr (VI) using modified biochar

Modified biochar has various functional groups such as carbonyl, hydroxyl, epoxy and carboxylic groups, which makes it suitable for the removal of Cr (VI). The functional groups aid in the adsorption of the metal through complexation and electrostatic attraction. The enhancement of the adsorption efficiency is achieved through chemical functionalization by introduction of suitable ligands onto the surface of the biochar through oxidation, co-polymerization and alkali/acid treatment [15].

There are various mechanisms involved in the removal of Cr (VI) using biochar through which the adsorption and reduction is achieved. One of the mechanisms is adsorption, which is achieved through surface complexation and electrostatic attraction. The functional groups on the surface help in the formation of very stable complexes with the Cr (VI) ions. Also, the surface charges on the modified biochar are able to attract the metal of interest in its different forms (dichromate and chromate ions) from the aqueous media. Another route in the removal of Cr (VI) is the reductive transformation. Especially when integrated with suitable reducing agents such as sulfur, iron and other reducing species. This has the potential of catalyzing the conversion of Cr (VI) to Cr (III), which is less toxic through a reduction reaction [16]. During this process, electrons are transferred from the surface of the biochar or enclosed reductants to the Cr (VI). After the reduction to the less toxic Cr (III), the new chemical species is then precipitated into oxide or hydroxide that is not soluble, thereby removing the chromium further from the medium. Chelation is a vital process during which some of the introduced functional groups, such as sulfide, amino acid and thiol chelate Cr (III), thereby stabilizing on the surface of biochar, hence preventing the process of re-mobilization. The overall efficiency of removal of Cr (VI) using this adsorbent is affected by pH, which affects the surface charge and reduction process. Adsorption is typically favored by acidic conditions. Detailed information about the molecular level of the mechanisms of adsorption for the removal of Cr (VI) is provided from various instrumentations such as X-ray diffraction and X-ray photoelectron spectroscopy.

This gives comprehensive insight into the interaction between the modified biochar and Cr

(VI) [5].

5.0 Regeneration and reuseability of modified biochar

Regeneration is the process of getting the adsorbent ready for repeated usage. It involves the desorption of the Cr (VI) using suitable solvents and treatments. Numerous solvents have been employed in this regard with their merits and challenges (Table 2). When the Cr (VI) covers the active sites on the surface of the modified biochar, the adsorption potential decreases over time. This calls for the need for treatments in restoring the active sites on the surface of the modified biochar. A suitable modified biochar must be able to pass through repeated sorption and desorption cycles, making it possible for recyclability and reusability. The regeneration of the modified biochar is achieved through a two-step process. It involves the adsorbate desorption and the decomposition of the adsorbate. An adsorbate is considered to be efficient if it can be reused with repeated adsorption and desorption processes, paving the way for recycling. There are different methods for achieving the regeneration of an adsorbent, which include solvent reactivation, thermal regeneration, supercritical fluid reactivation and microwave regeneration. This regeneration process is influenced by some factors such as properties of the biochar, the dose applied, and the selected regeneration technique. There are different solvents that have been found relevant in the regeneration of the adsorbent for Cr (VI) removal. [14] carried out the desorption of the biochar using nitric acid, with the adsorbent giving 63% and 97% removal for Cr (VI) and Pb (II) respectively, even after three cycles.

Table 3: Solvents for the regeneration and desorption of Cr (VI) from modified biochar

Solvent	Merits	Demerit	Challenge
Nitric acid	An acid with strong oxidizing property	A corrosive acid hence can degrade the modified biochar at higher concentration	Disposal is an area of concern as well as safety issue during handling
Hydrochloric acid	A very common acid that is a relatively cheaper solvent and suitable for desorption of Cr (VI)	Capable of generating waste containing chloride and able to degrade the modified biochar	Environmental and handling concerns
Ethylenediaminetetraacetic acid (EDTA)	A solvent that does not pose harm to the environment with remarkable chealating property	Relatively more expensive than other solvents, and its mildness calls for longer period of contact	A complete desorption of the Cr (VI) is not assured due to the mild nature of this solvent

Citric acid	A mild solvent that does not constitute environmental concern	The desorption of efficiency for Cr (VI) will be relatively lower using this solvent	Due to its mild nature, there could be need for repeated cycles of treatment to get desired result.
Hydrogen peroxide	A good desorbing agent for Cr (VI) due to its high oxidizing property	May result to the degradation of the morphology of the modified biochar overtime	Regulation of reaction conditions may be difficult using this solvent due to its oxidizing property.

6.0 Challenges associated with the use of modified biochar for removal of Cr (VI)

Limited cycles of regeneration: The regeneration of modified biochar overtime usually results in a decrease in the adsorption efficiency due to a gradual decrease in the number of active sites present, blockage of the surface pores, reduction in porosity and fouling. This therefore constitutes a serious challenge for large applications [10,17].

Large scale practicability of pH conditions: Most studies have reported that the optimum pH for the highest adsorption efficiency for most modified biochar is acidic condition with values ranging from 2 to 6. Such conditions, though very easy on a laboratory scale, may be difficult to maintain due to their cost implications for large scale operation.

Disparity in synthesis conditions and source of raw materials: The experimental conditions for the pyrolysis and modification of the biochar significantly affect and influence the physicochemical properties of the biochar synthesized. The various biomass sources for the production process also differ, thereby resulting in inconsistencies in the efficiency of the modified biochar.

Cost perspective of modification step: Modification of synthesized biochar involves the use of alkali, acids, and other agents. Though the cost is minimal for laboratory trials and experimentation, this may not be sustainable for large scale removal of chromium (VI). The regeneration of the adsorbent for subsequent usage also has cost implications as it requires various types of solvents to achieve this process.

Experimental and real scenarios: Most laboratory experimental studies carried out using modified biochar have proven to be highly promising with significantly higher adsorption efficiency. In real case scenarios, however, this would constitute challenges due to the occurrence of co-contamination where various kinds of interfering groups such as nitrates, sulfates, chlorides, and other heavy metal ions are also present in the wastewater with a potential to interfere with the active sites and functional groups present on the surface of the modified biochar thereby reducing the adsorption.

7.0 Future trends in the utilization of modified biochar for removal of Cr (VI)

There is need for more studies focusing on unraveling the comprehensive reaction kinetics involved in the removal of Cr (VI) using modified biochar. Also, the existing models on adsorption are on the basis of some assumptions which may be difficult to obtain from previous experiments. Furthermore, the real-time efficacy of biochar should be studied in complex real-life scenarios using industrial wastewater, where various organic materials and other competing ions present in the water can affect the process of adsorption. Lifecycle evaluation studies will be vital in the determination of the sustainability of the production process, experimental modification and strategies for disposal, from sustainability perspectives. Scaling up investigations in the synthesis of biochar for large-scale applications call for the need for optimization studies. The integration of modified biochar with other innovative technologies could have synergistic effects, thereby boosting the efficiency of the modified biochar in the removal of Cr (VI).

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