REVIEW ARTICLE

Chromium (VI) adsorption from steel industry wastewater using biomass and agro-based adsorbents: A Review

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ABSTRACT

In recent years, increased steel production has resulted in an increased release of Cr (VI) effluent from electroplating processes. The current conventional technologies for Cr (VI) removal, for instance, chemical precipitation, ion exchange, membranes, and electrochemical technologies, have proven to be expensive and not eco-friendly. Adsorption using biomass or agro-based adsorbents has been demonstrated to be an eco-friendly method. In this study, the potential of several biomass and agro-based adsorbents for Cr (VI) removal from steel industry wastewater has been reviewed. The application of adsorption parameters, isotherms, and kinetic models has also been reviewed. The study review reveals that the maximum adsorption of Cr (VI) ions from wastewater was dependent on the pH, temperature, contact time, type of adsorbent material, and initial metal concentration. The literature review demonstrated that maximum Cr (VI) adsorption efficiency ranged from 90–100%, with optimum pH, contact time, and temperature ranging from 2–5, 30-180 min, and 25–30 °C, respectively. The equilibrium adsorption data was best described by the Langmuir and Freundlich isotherms and the pseudo-second-order kinetic model. The thermodynamic studies revealed that the adsorption process was spontaneous and endothermic. It has been found that biomass and agro-based adsorbents are potentially eco-friendly alternatives for Cr (VI) removal from industrial wastewater. However, several gaps have been identified to improve the Cr (VI) removal efficiency, spent adsorbent reuse and safe disposal, optimisation, and commercial application of agro-based adsorbents.

Keywords: chromium (VI), adsorption, adsorbent, conventional methods, eco-friendly

1.0 Introduction

Cr (VI) metal is one of the most toxic and persistent heavy metals (ACGIH, 2021). It is widely applied in electroplating and metal finishing processes in the steel industry for alloy production (APAC, 2021; Bhatt et al., 2015; Sunghwan et al., 2020). The release of electroplated wastewater laden with Cr (VI) prior to treatment poses a health risk to both aquatic ecosystems and human beings (Nganga et al., 2023; Shalenie et al., 2021). Cr (VI) is carcinogenic even at low concentrations and can lead to kidney and liver damage, dermatitis, and gastrointestinal ulcers. For this reason, new and stricter environmental regulations are being enacted globally in order to control the discharge limit of heavy metals such as Cr (VI) into the ecosystem (UN, 2021).

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Currently, there are several existing technologies being applied to the removal of Cr (VI) metal from industrial wastewater. This includes chemical precipitation, electrocoagulation, ion exchange, electrodialysis, membrane separation, membrane filtration, electroflotation, and electrooxidation (Bayuo et al., 2021; Harshika et al., 2021; Rakesh et al., 2021). These technologies have proven to be expensive and not eco-friendly due to the high operational costs, sludge problems that create secondary pollution, complex design systems, and high electricity demand (Bo et al., 2020; Nasir & Fasil, 2021; Tang et al., 2020). As a result, adsorption treatment using low-cost agricultural or biomass-based adsorbents has demonstrated to be an eco-friendly technology due to optimisation of adsorption parameters and regeneration of spent adsorbents through the desorption process (Amina et al., 2022; Gupta et al., 2021).

Some low-cost materials that have demonstrated high chromium (VI) removal rates and regenerative abilities include calcined wheat bran (Fumihiko et al., 2020), walnut shell (Evans et al., 2022), spent tea leaves (Nur-E-Alam et al., 2018), almond shell (Rai et al., 2018), wheat straw (Song et al., 2016), avocado kernel seed and papaya peels (Mekonnen et al., 2015), orange fruit peel (Amir et al., 2017), banana fruit (Payel & Sarke, 2018). The efficacy of these adsorbents in metal ion uptake is due to the hydroxyl functional group on the active site (Anushka et al., 2022; Gupta et al., 2021). This article reviews recent research efforts being made on the use of eco-friendly agricultural and biomass-based adsorbents in the potential removal of Cr (VI) ions from steel industry wastewater.

2.0 Chromium (VI) use and pollution from the steel industry

Chromium is widely applied in the manufacturing of alloys, particularly in stainless steel production, where chromium improves the anti-corrosive properties of iron, which is normally susceptible to rusting (Fahim et al., 2006; Hadi & Joan, 2019). The alloy formation process results in the production of shiny stainless steel, which is often applied in the manufacture of kitchen utensils, cutlery, and motor vehicle accessories (APAC, 2021). During the alloy process, chromium increases the hardness of the alloy, making it a reliable material in the production of strong ball bearings that can withstand more than 6.895 × 109 Pa of pressure (Dongwook et al., 2020; Testa & Jacobs, 2004).

A chrome plating bath involves the use of an electrolyte solution that consists of chromium (VI) as chromic acid together with catalysts or activators in which the material to be coated is dipped (Michael, 2005; USGS, 2022). In the final stages, the electroplated material is rapidly rinsed of the excess electrolyte on its surface (Ken et al., 2020; Shammas et al., 2008). The rapid rinsing process is the main source of industrial wastewater, which consists of Cr (VI) and Cr (III) ions (approximately 35%) (Saha & Orvig, 2010; Surasit et al., 2019).

Cr (VI) is a hazardous heavy metal and exists in varying structural and functional forms: dichromate ($Cr_2O_7^{2-}$), bichromate ($HCrO_4^-$), and chromate (CrO_4^{2-}) which are influenced by pH and chromium concentration in solution, as shown in Figure 1 (ACGIH, 2021; OSHA, 2022). At pH values below 1.0, chromium exists as H_2CrO_4, $HCrO_4^-$ at pH values between 1-6, and chromate at pH 6 (Avila et al., 2014; Hadi & Joan, 2019). $Cr_2O_7^{2-}$ occurs when chromium concentration is 1 g/L (Dongwook et al., 2020; Mohan et al., 2006). This means that at normal

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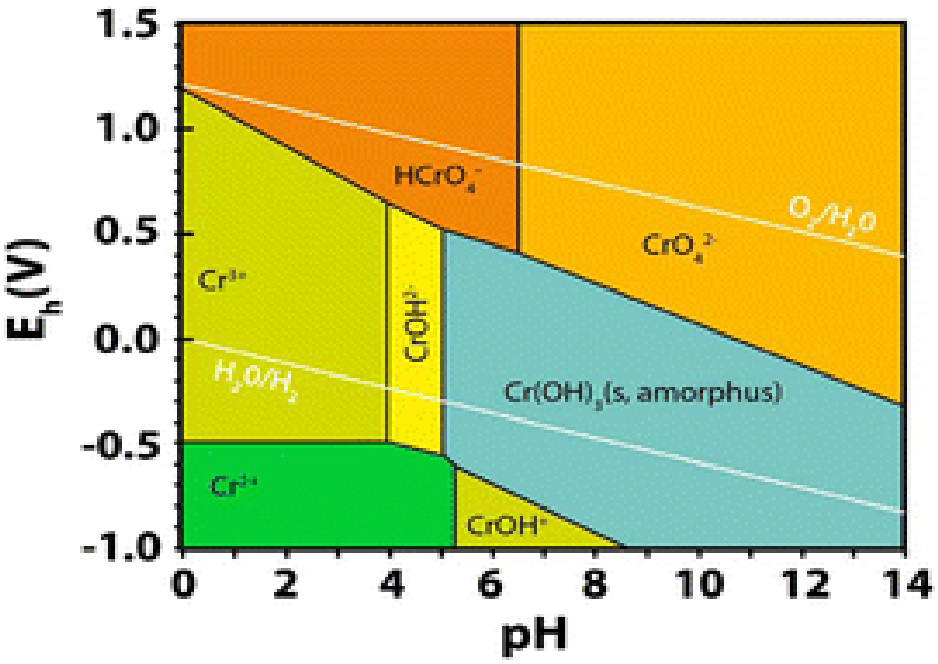
pH conditions, dichromate, bichromate, and chromate ions exist as Cr (VI) ions, which are mobile and relatively soluble in aquatic streams, thus making the treatment and management of the Cr (VI) effluent more challenging in comparison to other heavy metals (Avila et al., 2014; Ken et al., 2020).

The level of Cr (VI) concentration shown in Table 1 is from a typical steel industry wastewater effluent. The Cr (VI) effluent is not compliant with the discharge standard limit set by the World Health Organisation (WHO), the United States Environment Protection Agency (USEPA), the World Bank, the European Union, Kenya, and China, as illustrated in Table 1. The high level of Cr (VI) is attributed to the electroplating process in the steel industry and the subsequent lack of wastewater treatment at the discharge facility (Sunghwan et al., 2020). The presence of high levels of Cr (VI) is a threat to public health and surrounding ecosystems (Kinuthia et al., 2020). Therefore, there is a need to enforce the strict effluent standard by relevant authorities, and the steel industry should treat the wastewater effluent prior to discharge into the environment.



Source: (Hadi & Joan, 2019)

Figure 1: Eh-pH dependence for Cr species in water

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Table 1. Cr (VI) effluent from the steel industry and compliance to discharge standard limit (Kinuthia et al., 2020).

Parameter	Temp ℃	pH	Conductivity (µs/cm)	Cr (VI) (mg/l)
Direct effluent from Typical Steel industry	30	7.43	400	0.15-1.5
Recommended discharge standard limit for Cr (VI) effluent in ppm or mg/l				
World Health Organization	20-32	6.5-8.5	400-600	0.05
China	-	6.5-8.5	-	0.05 Max
Kenya, NEMA	-	6.5-8.5	-	0.05
USA EPA	-	6.5-8.5	-	0.1
World Bank	-	6.5-8.5	-	0.05
European Union (EU)	-	6.5-8.5	-	0.05

Note: Wastewater effluent is from direct discharge into surface water

3.0 Human health and environmental impact of chromium (VI) effluent

The toxic effects of orally ingesting hexavalent chromium on human health have been observed in the human population through ingestion of food or potable water (Carla et al., 2021; Coetzee et al., 2018). The reduction process of Cr (VI) to Cr (III) through glutathione reductase (GR) and other antioxidants such as Nicotinamide Adenine Dinucleotide Phosphate Oxidase (NADPH) leads to the formation of long-lived Cr (V) intermediates (Jianlin et al., 2021; Lee & Son, 2012). The reduction process also leads to the release of diverse reactive oxygen species (ROS), such as superoxide anions, hydroxyl radicals, and singlet oxygen species (Hong & Max, 2022; Karihtala & Soini, 2007). This leads to an imbalance (oxidative stress) between ROS formation and the capacity of cellular antioxidants (Guiping et al., 2022; Quinteros et al., 2008), as shown in Equation 1. This stress has the potential to lead to breakage in DNA single strands that can subsequently cause irreversible genetic change in human beings (James et al., 2019).

$$Cr^{6+} + GR/NADPH \rightarrow Cr^{3+} \rightarrow Cr^{5+} + ROS \quad (1)$$

Exposure to Cr (VI) can exacerbate concentration-dependent morphological changes in cells, for instance, lipid peroxidation, which can subsequently lead to cell membrane destruction as a result of Cr (VI)-induced ROS activity (Carla et al., 2021). The oxidative stress attributed to the presence of ROS is also responsible for the cell cycle arrest that consequently leads to the death of programmed cells (apoptosis) (Guiping et al., 2022). This phenomenon can manifest through damage to proteins and modified proteins (He et al., 2007; Meiduo et al., 2022). High exposure to hexavalent chromium can lead to two main types of dermatological toxicity: skin ulcers and allergic contact dermatitis. Several case studies that have been carried out on cancer and illness related to chromium waste are illustrated in Table 2 below:

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Table 2: Human health impacts associated with exposure to Cr (VI) metal ions.

Chromium (VI) related illness and diseases	References
Ulcers, chronic lung disease e.g asthma, rhinitis, pulmonary fibrisis and skin ulcers and allergic contact dermatitis in humans	
Lung cancer	(Deborah et al., 2021; Jin et al., 2017; Kamila et al., 2016; Tawhid et al., 2018)
Colorectal cancer	(Zecai et al., 2020)
Prostrate cancer	(Changwen et al., 2019)
Epigenetic changes causing human lung cancer tissues	(Zhishan et al., 2018)
Bladder cancer risk	(Stamatiou, 2012)
Colorectal cancer	(Fenghe et al., 2020)
Gastrointestinal tract cancer	(Haney, 2015; Nicole et al., 2010)
Tissue cancer	(Haney, 2015)
Lung, nose and nasal sinus, stomach and laryngeal cancer	(Shalenie et al., 2021)
Head and neck cancer	(Rim et al., 2013)
Genetic damage and immune inflammation	(Jianlin et al., 2021)
Small intestine cancer	(Roman & Scott, 2021)

4.0 Cr (VI) adsorption using biomass/agro-based adsorbents

The adsorption process is classified as physisorption, which involves Van der Waal's forces (Masatoshi & Takashi, 2013), and chemisorption, which involves covalent bonds (Chikazawa & Takei, 2006; Amina et al., 2022). In solid-liquid adsorption systems, the first process in adsorption involves the transfer of metal ions from the wastewater to the adsorbent surface (Gupta et al., 2021). The metal ions are then transferred onto the adsorbent pores, particularly the porous adsorbent. The final step involves the attraction of metal ions by adsorbent binding sites, which are the hydroxyl groups located inside the pore structure (Si et al., 2013; Juliana et al., 2021).

4.1 Factors influencing Cr (VI) adsorption and experiment design

Batch adsorption experiment design is often performed to examine the influence of physio-chemical parameters. This is because metal ions exist in various forms at varying pH conditions and temperature (Tinega & Warui, 2021). Significant parameters, i.e., the initial concentration of Cr (VI), adsorbent dose, contact time, and solution temperature, have a huge influence on the effectiveness of the adsorption process (Bhavsar & Patel, 2014; Anushka et al., 2022). Data on the impact or influence of contact time and initial concentration can be applied to determine parameters in kinetic and isotherm models, respectively (Nur-E-Alam et al., 2018). The factors influencing metal adsorption by biomass-based adsorbents are summarised and presented below in Table 3 (Tinega et al., 2023).

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Table 3: Factors influencing metal adsorption by biomass-based adsorbents

Factors	Effects
Adsorbent dose	Reduces the amount of adsorbed metal ions per unit mass of adsorbent and thereafter increases its removal percentage
Adsorbent particle size	The small adsorbent size increases metal adsorption in the batch study due to large surface area.
Initial metal concentration	Increases the amount of adsorbed metal ions per unit mass of adsorbent but reduces its removal percentage
pH	Improves adsorption of positively-charged metal ions
Temperature	Increases adsorption of metal ions by increasing the surface activity and metal ion kinetic energy; but higher temperature could destroy adsorbent physical structure
Agitation speed	Increases adsorption of metal ions by reducing its mass transfer resistance; but higher speeds could destroy adsorbent physical structure
Ionic strength	Reduces adsorption of metal ions by competing with the solute for vacant binding sites of the adsorbent
Other contaminant concentration	In case there are coexisting pollutants competing with the targeted metal ion for binding sites it will decrease the removal rate of targeted metal ion

4.2 Adsorption kinetic models

Kinetic models are used to determine the rate of reactions and how experimental conditions influence metal ion adsorption (Guerra et al., 2014; Evans et al., 2022). Kinetic studies are also able to reveal crucial information on the rate of adsorption and possible intermediate processes that occur during the formation of the adsorbate-adsorbent complex reaction (Cao et al., 2013; Daniel et al., 2021). These findings can be obtained through the application of several kinetic models, which are pseudo-first-order kinetic models and pseudo-second-order kinetic models. The pseudo-first-order kinetic model is commonly applied to depict kinetics in the liquid-solid phase adsorption process discovered by Lagergren in 1898 (Fumihiko et al., 2020). This model also reveals the adsorption rate in a reaction based on the adsorption capacity. The model is illustrated in equation 2.

$$\frac{dq_t}{dt} = \left(k_1(q_e - q_t)\right) \quad (2)$$

where k_1 (min-1) is the pseudo-first-order rate constant, q_e (mg/g), and q_t (mg/g) is the adsorption capacity at equilibrium and at time t, respectively. Integration of the above equation using boundary conditions as shown in equation 3, whereby t = 0, q_t = 0, and t = t, q_t = q_t (Sen & Bhattacharyya, 2011; Dim et al., 2021).

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$$\mathrm{In}(q_e - q_t) = \mathrm{In}q_e - k_1 t \quad (3)$$

A graph plot of *ln* $(q_e - q_t)$ vs *t* reveals a straight graph line, and the k_1 value is obtained from the gradient slope. The k_1 value rises with increasing initial metal concentration but varies depending on the adsorption process. This model is only suitable and fit to describe the initial phase of adsorption, which is the initial or first 20 to 30 min; therefore, it can't be suitable if the whole metal-adsorbent time of contact is taken into consideration (Mohammad et al., 2022).

The pseudo-second-order kinetics model is also widely applied to demonstrate the adsorption kinetics of metal adsorbents. This model is best suited to describe adsorption systems whereby the process of metal adsorption-desorption at the adsorbent surface influences the whole metal adsorption process (Plazinski et al., 2013; Selimin et al., 2021a), as illustrated in equation 4.

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (4)$$

k_2 is the rate constant for pseudo-second-order kinetics. The above equation can be integrated using the boundary conditions where t = 0, q_t = 0, and t = t, q_t = q_t, obtaining equation 5 (Robati, 2013; Fikiru et al., 2021);

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

k_2, which is the rate constant, depends on the time factor, which is influenced by the initial metal ion concentration. The rate constant declines with increased initial metal concentration, which means more time is required to attain equilibrium if the initial metal ion concentration increases (Sen & Bhattacharyya, 2011; Dim et al., 2021).

4.3 Adsorption isotherm model

The isotherm adsorption studies are also significant in investigating the adsorption mechanism (Guerra et al., 2014; Selimin et al., 2021b). The most commonly used isotherm models include the Langmuir and Freundlich models. The Langmuir isotherm is a widely applied model in the metal adsorption process. Theoretically, it assumes that metal adsorption occurs on the first monolayer (Hu et al., 2009; Sasireka et al., 2022), and the adsorbent surface is homogeneous, with all the active sites having the same energy and no interaction among the adsorbed ions (Boparai et al., 2011; Nader et al., 2020). The Langmuir model is illustrated in equation 6.

$$\frac{C_e}{q_e} = \frac{1}{q_{bm}} + \frac{C_e}{q_m} \quad (6)$$

C_e (mg/L) is metal concentration at equilibrium, and q_e (mg/g) is adsorption capacity at equilibrium, while q_m (mg/g) represents the maximum metal adsorption capacity. The Langmuir constant is b (L/mg), which relates to the adsorption energy (Alomá et al., 2014; Binu et al., 2022).

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The Freundlich isotherm is an adsorption model that depicts adsorption as multilayered and characterised by a heterogeneous layer (Jung et al., 2013; Khan et al., 2021). The model is depicted in equation 7.

$$q_e = k_f C_e^{\frac{1}{n}} \quad (7)$$

q_e (mg/g) is metal adsorption capacity at equilibrium, and C_e (mg/L) is metal concentration at equilibrium. The Freundlich constants K_f are related to adsorption capacity, and n is related to intensity (Desta, 2013; Nader et al., 2020). The linear equation for the Freundlich isotherm is illustrated in equation 8:

$$\mathrm{In}q_e = \mathrm{In}k_f + \frac{1}{n}\mathrm{In}C_e \quad (8)$$

4.4 Adsorption thermodynamic studies

Thermodynamic parameters include Gibbs free energy, entropy, and enthalpy. The equilibrium constant obtained from the Langmuir model at a given temperature can be applied to investigate the thermodynamic parameters (Amina et al., 2022). The Gibbs free energy change of adsorption can be expressed with respect to the Langmuir adsorption constant in equation 9.

$$\Delta G^{\circ} = -RT\mathrm{In}K_L \quad (9)$$

The Gibbs energy change is linked to the change in enthalpy and entropy under constant temperature, as illustrated in equation 10.

$$\Delta G^{\circ} = \Delta H^{\circ} - T\Delta S^{\circ} \quad (10)$$

Combining equations 13 and 14, we obtain equation 11:

$$\mathrm{In}K_L = \frac{\Delta S^{\circ}}{R} - \frac{\Delta H^{\circ}}{RT} \quad (11)$$

where R represents the ideal gas constant (8.314 J/mol.K), is the distribution coefficient for the adsorption (mL/g), and T represents the absolute temperature of the aqueous solution.

The values of and are obtained from the slope and intercept, respectively, by plotting against $\mathrm{In}K_L$ against$\frac{1}{T}$.

4.5 Cr (VI) removal studies by low-cost biomass and agro-based adsorbents

Numerous studies on the use of agricultural by-products or waste have demonstrated high potential for Cr (VI) removal due to high carbon content, surface functional group, pore size and surface area (Binu et al., 2022; Khan et al., 2021; Selimin et al., 2021~~b~~; Mohammad et al., 2022). Rye is a grain and precursor material for beer, vodka, whisky, and bread (Bledzki et al.,

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2010). During the production of rye flour, the rye husk, which is the outer part, is discarded. The rye husk waste revealed maximum Cr (VI) adsorption capacity of 22.61 mg/g at optimum pH 3. This was due to the carboxyl and hydroxyl surface functional groups (Altun et al., 2015). In this study, maximum Cr (VI) ion adsorption occurred due to electrostatic forces between $HCrO_4$ ions and the adsorbent surface (OH group).

Wheat bran has also been applied for Cr (VI) ion removal (Kaya et al., 2014). Modification of wheat bran by tartaric acid demonstrated high adsorption capacity using the Langmuir model at 4.53 mg/g and 5.28 mg/g. The wheat bran was modified to increase the amount of carboxyl groups thus increasing Cr (VI) ion binding sites (Nameni et al., 2008). The Cr (VI) ions would then bind to the carboxyl and hydroxyl groups at low pH, subsequently leading to a reduction of Cr (VI) to Cr (III). Other studies on the use of wheat by-products from mill plants have demonstrated high adsorption capacities ranging from 0.9 mg/g to 35 mg/g (Kanwal, 2012; Ahmad & Nisa, 2013). The highest removal efficiency was also observed at low pH, and this occurred as a result of electrostatic forces between Cr (VI) ions and the adsorbent surface.

Rice straw, a byproduct from the mill plant, has also demonstrated the ability to remove metal ions from waste water (Elmolla et al., 2015). A study on chemically modified rice straw revealed a high Cr (VI) ion adsorption capacity of 20 mg/g at low pH (Elmolla et al., 2015; Wu et al., 2016). Other studies have demonstrated the use of rice husk fly ash obtained through burning of the husks under air or nitrogen conditions in a fluidized-bed reactor (Georgieva et al., 2015). For instance, removal of Cr (VI) by this byproduct showed high adsorption capacity from 4 to 20 mg/g (Georgieva et al., 2015).

Studies on the application of by-products from maize plantations and milling plants have also demonstrated the ability to remove Cr (VI) metal ions (Singh & Rattan, 2014). Previous studies on the potential use of maize stover, maize cob ash, and maize husk revealed low adsorption capacity at below 5 mg/g using the Langmuir isotherm model (Wang et al., 2014; Singh & Rattan, 2014). The removal mechanism was due to electrostatic forces. A study by Kanwal on the use of polyanaline-corn bran composites demonstrated a high adsorption capacity of 22 mg/g at pH 4 (Kanwal, 2012). The adsorption of Cr (VI) ions was due to ion exchange between Cl^- from the composite and the $Cr_2O_7^{2-}$ ions.

Sugarcane bagasse (*Saccharum officinarum L.*) is a fibre-rich waste or byproduct produced after crushing the cane stalk and extracting juice (Ullah et al., 2013). Studies have attempted to investigate its potential ability to remove metal; however, they revealed a low adsorption rate for Cr (VI) ions. Aloma et al., using the Langmuir model, observed a high adsorption capacity of 1.8 mg/g at pH 2 and a temperature of 25°C (Alomá et al., 2014). This was due to electrostatic forces. In a different study, the use of sodium hydroxide to modify sugarcane bagasse demonstrated an improved adsorption capacity of 6 mg/g (Langmuir) at 25°C (Mahmood-ul-Hassan et al., 2015). The use of sodium hydroxide to modify sugar cane bagasse hydrolyzed the hemicellulose, which led to increased porosity of the adsorbent surface. This also led to the hydroxyl and carboxyl groups being exposed, thus increasing the binding sites for Cr (VI) ion adsorption.

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Mythili and Karthikeyan examined the biosorption potential of chromium ions by *Bacillus spp.* and *Staphylococcus spp.* microorganisms from industrial discharge with an initial metal concentration of 100 mg/l. The optimum chromium biosorption capacity was 94.5 mg/l at pH 7 and 92.9 mg/l at 37 °C for *Bacillus spp.,* while for *Staphylococcus spp.*, it was 72.3 mg/l at pH 8.0 and 72.6 mg/l at 37 °C (Mythili & Karthikeyan, 2011). Cr (VI) removal through a trickling filter is a new biological method for Cr reduction. Dermou et al. experimented using three different modes of trickling filter operations: batch, continuous, and SBR with recirculation by applying indigenous bacteria (*Acinetobacter sp.*) as inoculum for the filter. The results revealed a high Cr (VI) removal rate from industrial effluent (Dermou et al., 2005).

The algae species, for instance, *Spirullina sp., Chlorella sp., Scenedesmus sp., and Scenedesmus sp.,* have demonstrated the ability to adsorb heavy metals due to the presence of polysaccharides in their cells (Mahboubeh et al., 2022). The fungi have also demonstrated a potential ability to absorb metal ions through the hyphae, which aids in their metabolism or breakdown (Christopher et al., 2016; Lin et al., 2021). Srivastava and Thakur carried out a study on the biosorption of Cr (VI) from industrial discharge with the use of Aspergillus sp. fungi, with an 85% chromium removal rate at pH 6 in the bioreactor system (Srivastava & Thakur, 2006). Bio-adsorbents such as *Parawaldeckia*, *Borassusaethiopum* (Tadesse & Seyoum, 2015), Cyprus papyrus (Kassaye et al., 2017), *Hyptissuaveolens* (Sivakumar et al., 2016), *Chrysopogonzizanioides, and Cyperusrotundus* (Bekele, 2018) have also demonstrated the potential to remove Cr (VI) from industrial wastewater.

Several studies have demonstrated improved Cr (VI) adsorption performance due to pretreatment of the adsorbent surface. For instance, a study by Obakeng et al. (2020) examined the Cr (VI) removal efficiency of macadamia nutshells treated with sodium hydroxide, nitric acid, and fenton-like reagents. The study findings revealed that the base-treated adsorbent demonstrated maximum Cr (VI) adsorption performance at 98% compared to the acid and fenton-treated adsorbent.

A study by Qian et al. (2022) on the adsorption performance of pretreated peanut shell by deep eutectic solvents (DES) on Cr (VI) removal revealed that the rigid structure of the peanut shell was broken down. This was attributed to DES modification of the adsorbent surface, which had a slight acidic character, leading to a maximum Cr (VI) adsorption capacity of 5.36 mg/g. Seyyedeh et al. (2021) investigated the effects of pretreatment on Elm tree sawdust adsorbent by alkali and alkaline earth metals (NaCl, KCl, $CaCl_2$, $MgCl_2$, and Elm tree ash) and deashing solutions (water, HCl, HNO_3, and aqua regia) before carbonization. The results revealed that HCl leaching pretreatment of the biomass improved the activated carbon adsorption capacity of Cr (VI) from 114 to 190 mg/g. The high adsorption performance was attributed to the development of the micro- or mesoporous structure of activated carbon after pretreatment.

A study by Madhumita et al. (2014) on sulfuric acid-modified avocado seed adsorbents for Cr (VI) removal revealed the development of agglomerated and rodlike structure particles and mesoporous structure. Also, the presence of an oxo-functional group on the adsorbent surface resulted in a maximum adsorption capacity of 333.33 mg/g at pH 2 and 25 °C.

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Recent studies have demonstrated that agricultural and biomass-based adsorption could be optimised to improve adsorption efficiency. For instance, a review study by Bayuo (2021) revealed that maximum Cr (VI) removal efficiency at almost 100% was attained by several low-cost adsorbents at optimised conditions, which were pH 2–6, contact time 30-180 min, and 25–30 °C. A study by Yusuff (2018) on Cr (VI) adsorption using *Leucaena leucocephala* seed shell activated carbon achieved 96% removal efficiency due to optimised parameters, that is, pH 4.22, 0.57g adsorbent dose, and temperature at 26.2 °C.

In the study by Surafel et al. (2022), the optimised parameter values for maximum Cr (VI) removal rate at 95% by teff straw-based activated carbon were achieved at pH 2.2, an adsorbent dose of 2.7 g/100 ml, and a contact time of 109 min. A study by Seyed et al. (2022) using modified sesame hulls to remove Cr (VI) from an aqueous solution observed that maximum adsorption efficiency was attained at 100%. This was achieved at optimised values, that is, pH 3, 120 min contact time, and 4.9 g/l adsorbent dose. A study by Hammad et al. (2021) using carbonaceous materials obtained from domestic firework places attained a maximum Cr (VI) adsorption capacity of 1.37 mg/g at an optimum pH of 2, a temperature of 33.6 °C, and a contact time of 94.7 min.

The spent adsorbent disposal also poses an environmental challenge since it's laden with Cr (VI) ions (Baskar et al., 2022; Bayuo et al., 2020a). In order to optimise the adsorption process, the spent adsorbent is regenerated or reused, and Cr (VI) is recovered through the desorption process in several adsorption-desorption cycles (Chatterjee & Abraham, 2019; Grover et al., 2012). The optimisation process prolongs the life cycle of the adsorbent material while reducing its cost (Baskar et al., 2022). Adsorbent regeneration is significant in optimising the commercial, health, and environmental benefits of the adsorption process in both batch and continuous designs.

During desorption process, the Cr (VI) metal ions held onto the adsorbent surface are desorbed through proton exchange by acid and base eluants (Chatterjee & Abraham, 2019; Sun et al., 2020). A previous study by Rais et al. (2012) demonstrated that if the spent adsorbent is recycled or regenerated after the first adsorption cycle, then the process is not only commercially viable but also eco-friendly. Bayuo et al. (2020b) performed desorption of Cr (VI) on spent groundnut husk adsorbent using 0.1M hydrochloric and sulphuric acid as desorbing agents or eluants, respectively. A maximum desorption rate of 76% was attained by HCl, respectively, at an optimal contact time of 1 hour, 30 °C, and 120 rpm. The study results revealed that the spent adsorbent could be optimally reused or regenerated in three successive adsorption-desorption cycles without compromising the metal adsorption capacity of the adsorbent. The study also discovered that acidic agents desorb Cr (VI) faster than basic or neutral agents.

Several studies have proven the applicability of agricultural and biomass-based adsorption at the pilot scale. For instance, a study by Quintelas et al. (2009) evaluated the applicability of a biofilm in Cr (VI) removal in solution at a pilot scale. The batch adsorption studies were applied to develop a pilot bioreactor that was able to treat Cr (VI) in an aqueous solution. Data obtained

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in the pilot-scale reactor revealed an average removal percentage of 99.9% in the first 30 days for the initial concentration of 10 mg/l and an average removal percentage of 72% for the same period for the initial concentration of 100 mg/l. Shinde et al. (2018) carried out a study on the adsorption efficiency of Cr (VI) ions from electroplating effluent by powdered activated charcoal at bench and pilot scales. It was observed that the pilot-scale process was equally effective as the bench-scale process for the removal of Cr (VI) ions. At the pilot scale, a 97% Cr (VI) removal rate was achieved. Renata et al. (2020) performed Cr (VI) adsorption on a batch and continuous scale. The fixed bed column pilot test results revealed a breakthrough time of 135 min with 5g of granular activated carbon and 40 mg/l of Cr (VI) ions. The removal efficiency was achieved at 92%. The optimal values were pH 2, an adsorbent dose of 10 g/l, and 50 min of contact time. The above results on pilot-scale adsorption of Cr (VI) ions by agricultural or biomass-based adsorption demonstrate its applicability at the industrial scale. Further study on adsorption with the combined use of agro-based adsorbents and nanoparticles is also necessary prior to large-scale application in order to optimise the removal efficiency and commercial viability. The adsorption of Cr (VI) ions using other low-cost adsorbents is summarised in Table 4.

Table 4: Summary of low-cost biomass adsorbents and their Cr (VI) adsorption capacities

Adsorbent	pH	Temp. (°C)	Adsorption capacity mg/g	Removal rate %	Isotherm model	Kinetic model	Reference
Arachis hypogea husk	8.0		2.4		Redlich-Peterson	Pseudo-second order	(Bayuo et al., 2020a)
Activated teff husk carbon	1.9			95.6	Langmuir	Pseudo-second order	(Adane et al., 2020)
Kaolin modified with cetyl trimethyl ammonium bromide (CTAB)				99	Langmuir	Pseudo-first order	(Belachew & Hinsene, 2020)
Heinsia crinite seed coat	2.0				Freundlich	Pseudo-second order and endothermic reaction	(Dawodu et al., 2020)
Bagasse Biopolymer gel beads	4.0	25		98.4			(Kumar et al., 2020)
Artocarpus nobilis fruit modified by NaOH	2.0	27.5	4.9			Pseudo first-order kinetic	(Samaraweera et al., 2020)
Mosambi peel dust	2.0				Dubinin-Radushkevich	Pseudo second order and endothermic reaction	(Mondal et al., 2019)
Corn cob			277.6		Langmuir	Pseudo second order	(Manzoor et al., 2019)
Microalgal biochar				100	Langmuir	Pseudo second order	(Daneshvar et al., 2019)
Chemically activated chicken pea husks			59.6		Langmuir	Pseudo second order and endothermic reaction	(Özsin et al., 2019)

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Adsorbent	pH	Temp (°C)	Adsorption capacity (mg/g)	Removal rate %	Isotherm model	Kinetic model	Reference
Leucaena leucocephala seed shell	4.2	26.2		95.6	Freundlich	Pseudo second order	(Yusuff, 2018)
Spent tea leaves	10		10.6	95.4			(Nur-E-Alam et al., 2018)
Microporous activated almond shell carbon	2.0			100	Langmuir	Pseudo second order and endothermic reaction	(Rai et al., 2018)
Olive stones derived carbon	2.0		74.9		Langmuir	Pseudo second order and exothermic reaction	(Ba et al., 2018)
Activated paper mill sludge carbon	4.0	45	23.2		Langmuir	Pseudo second order and endothermic reaction	(Gorzin & Abadi, 2018)
Activated ziziphus jojuba cores carbon	2.0			27.2 – 62.1	Joth & Elovich model	Pseudo second order and endothermic reaction	(Labied et al., 2018)
Iron treated tea waste biomass	2.0			99.9			(Gupta & Balomajumder, 2017)
Macadamia nutshell biosorbent	2.0				Langmuir	Pseudo second order	(Pakade et al., 2017)
Almond green hull powder				99.0	Freundlich		(Nasseh et al., 2017)
Wheat straw & Eupatorium adenophorum	1.0	35	89.2	99.9	Langmuir	Pseudo second order and endothermic reaction	(Song et al., 2016)
Onopordom heteracanthom (weed) biochar	1.0				Langmuir	Pseudo second order	(Ghorbani & Behnajady, 2016)

Adsorbent	pH	Temp °C	Adsorption capacity (mg/g)	Removal rate %	Isotherm model	Kinetic model	Reference
Passiflora foetida plant seed					Freundlich model	Pseudo second order and exothermic reaction	(Velumani et al., 2016)
Phragmities stem modified by conc. sulphuric acid	1 & 2		200		Freundlich	Pseudo second order	(Regmi et al., 2015)
Mangrove leaf powder	2.0				Langmuir	Pseudo second order and endothermic reaction	(Sathish et al., 2015)
Avocado kernel seeds, Juniperus procera sawdust, papaya peels	1.0	40			Freundlich	Pseudo second order	(Mekonnen et al., 2015)
Peach stone derived activated carbon					Freundlich	Pseudo second order	(Duranoglu & Beker, 2015)
Activated bamboo waste carbon (Oxytenanthera abyssinica)	2.0	27	59.2	98.3	Freundlich	Pseudo second order and exothermic reaction	(Dula et al., 2014)

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Palm kernel and coconut husk respectively			84.9-91.1	Freundlich	Pseudo second order	(Ju & Okoli, 2014)
Coffee polyphenol-formaldehyde acetaldehyde resins	2.0			Freundlich		(Mulani et al., 2013)
Groundnut shell					Pseudo second order	(Idris, 2012)
Dalbergia sissoo pods	2.0			Langmuir and Freundlich	Pseudo second order	(Mahajan & Sud, 2012)

Adsorbent	pH	Temp (°C)	Adsorption capacity (mg/g)	Removal rate %	Isotherm model	Kinetic model	Reference
Chemically activated olive stone carbon	1.5				Langmuir	Pseudo first order	(Attia et al., 2010)
Orange peel citrus sinensis	3.0	30	81		Langmuir		(Ekpete et al., 1970)
Coconut shell	2.0- 9.0				Langmuir & Freundlich	Pseudo first order and Pseudo second order	(Pinto et al., 2006)
Neem leaf powder	4.5- 7.5	27	0.1	87	Langmuir		(Sharma & Bhattacharyya, 2005)

5.0 Conclusion

The review of biomass and agro-based adsorbents has shown them to be a potentially eco-friendly, efficient, and low-cost technology for Cr (VI) removal from steel industrial wastewater. Therefore, biomass and agro-based adsorbents should be exploited to reduce cost and maximise Cr (VI) adsorption capabilities. The review revealed that efficient removal of Cr (VI) ions highly depends on the pH of the solution, contact time, adsorbent type, initial metal concentration, and temperature. From the review of relevant literature, the maximum Cr (VI) removal efficiency using the various biomass and agro-based adsorbents ranged from 95–100%, with optimum pH and contact times ranging from 2–5 and 30-120 min at a temperature of 30 °C. The pseudo-second kinetics and Langmuir and Freundlich isotherm best described the Cr (VI) equilibrium adsorption.

The review study also demonstrated that the Cr (VI) adsorption efficiency of various low-cost biomass and agro-based adsorbents could be improved through pretreatment of adsorbents and optimisation of adsorption parameters with complex wastewater systems. The waste/exhaustive adsorbent disposal problem could also be optimised through desorption process, making the process eco-friendly and reducing the cost associated with disposal.

Nevertheless, further studies should be carried out to improve adsorbents applicability on a commercial scale, for instance, through combined use with nanoparticles or hybrid technology. The exhaust adsorbent regeneration could also be improved using different eluants in order to prolong the adsorbent life cycle, thus reducing disposal costs.

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The authors declare there was no conflict of interest

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