Review Paper. Utilization of Low-Cost Adsorbents for the Adsorption Process of Chromium ions.

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Abstract. This review is aimed at exploring the possibilities of recovering Chromium ions using several low-cost adsorbents through wastewater processing. In the past, several traditional methods were employed for removing Chromium ions. These included precipitation, evaporation, electroplating and ion exchange. However, these processes were associated with various limitations, which included the treatments to be restricted to a certain concentration of the Chromium ions. Therefore, the process of using low-cost adsorbents can be deemed as an eco-friendly one. At the moment, an enormous amount of natural materials and agricultural waste are produced, which extremely harmful to the environment. Thus, adsorption is an alternate process for removing Chromium ions. Based on the enhanced characteristics of the process of adsorption, such as cost-effectiveness, improved adsorptive properties, and increased availability, the process is definitely an economical one for removing Chromium ions. This review provides a brief appraisal of the relevant literature which exists on the low-cost adsorption for removing Chromium from polluted wastewaters. Additionally, in order to comprehend the overall adsorption process of low-cost adsorbents, this review will also explore the various existing adsorption models. These include the isotherm, kinetics, and thermodynamics along with the impact of various factors on the process of adsorption.

Keywords: Adsorption, Chromium ions, Conventional methods, Low-cost adsorbent.

1. Introduction
Rapid industrial development causes huge pollution on our planet especially contamination in the heavy metals that result from the mining industries, large world traffic...etc, that Chromium to passive effects on the plants and ecology [1-3]. One of the higher toxicity elements is the Chromium, it's contaminating the water, soil and the ecology system in general. Chromium discharges mostly coming from the textile, leather tanning, metal extraction factory, and electroplating as effluents [4]. Common states of the Chromium oxidation are trivalent state and hexavalent state, they are more stable and constant [5]. The Chromium in the hexavalent state considered higher toxicity than the Chromium in the trivalent state due to form a complex compound with Oxygen such as Cr₂O³⁻ and CrO₄²⁻ oxoanions [6]. Experimentally, the trivalent Chromium state is highly related to the organic substances in soil, it has less toxicity and less activity. Literature review of the manuscripts showing that Chromium in the trivalent state easily enters in leaves and or the plant cell organs then changes to Chromium in the hexavalent state [7]. Analyzing, the Chromium is collecting in the plants' roots 100 once more than the leaf, branches, and Leg [8,9]. The high accumulation of Chromium in the plants that grow in the serpentine soils found Collecting was Herniaria hirsute [10]. Chromium is transferring to plant by
soil with different harvest crops like to the transfer mechanisms of the Chromium in the hexavalent state to the barley, this process has been notifying [11-13]. Hexavalent Chromium has a larger impact on environmental assessment due to the insoluble of the hexavalent Chromium is regularly engrossed further simply a cross the cell of body membranes more willingly [14-15]. Therefore, it is more poisonous and steadier than chromium (III). Global environmental values in wastewater involve chromium should not go above 0.05 mg/L for hexavalent Chromium and 5mg/L for trivalent Chromium. The chromium(III) compounds are mainly toxic components of tannery wastewater, and they are used in various industries such as metallurgy, production of pigments, electroplating, pesticide, painting aircraft, preservative of wood, Textile manufacturing, dyeing, cement industry, metal finishing, photography industry, leather tanning …etc, by applications for conservation stabilization of proteins of animal hides [16]. Chromium is a transition element, it’s in various oxidation states, the main popular ones are the trivalent chromium in formed ([Cr(H2O)6]3−) and hexavalent chromium in form (CrO2−4 or Cr2O7−2) states [17]. The first type is more stable, and small quantities are required for the satisfactory performance of certain biological functions in human beings [18]. Hexavalent Chromium has a harmful and adverse effect on human health, it founds at a low concentration in different products [19]. Therefore, chromium removal from wastewater excite big interest. Many approaches to remove chromium like chemical precipitation [20-21], ion exchange [22-23], electro-coagulation [24-28], Reverse Osmosis (Membrane) technique [29-32], foam flotation [33-35], electrolysis [36-37] and Electrochemical [38-39] these have been notified, however, these methods have disadvantages like costly, great feed solution input of chemicals and the removal incomplete [40]. Adsorption operation utilized for treating wastewater that contains toxic ions of heavy metal is a suitable and efficient mechanism [41-42]. Utilizing different materials for chromium removal by adsorption, there are many types of research and manuscript published deal with the adsorption process onto activated aluminum [43], activated carbon & elution with sulfuric acid [44], superabsorbant polymer [45], Iron Oxide/Mesoporous Silica Nanocomposites [46], aluminum oxide hydroxide [47], Metal-Organic Framework [48], Blast Furnace Dust [49], hydrous chromium dioxide [50], iron oxide magnetic nanoparticle [51], PEI-silica nanocomposite [52], polyacrylonitrile-based porous carbon [53], Synthetic and Natural Polymers [54], carboxymethyl cellulose-based hydrogel [55], diatomite treated with micro emulsion [56], nanoscale zero-valent metal [57], calcium alginate as biopolymer [58], Surface Modified Nanozeolite [59], Scolecite Natural Zeolite [60], Biosorbents Supported on Zeolites [61], Beta vulgaris or Celery (Apium graveolens) after the Addition of Modified Zeolites [62], Zeolite/Chitosan Hybrid Composite [63], zeolite NaX [64], natural clinoptilolite zeolite [65], HDTMA-modified zeolites [66], surfactant-modified Sabzevar nano-zeolite [67], Zeolite and Agro-industrial Waste [68], amino-functionalized nano-Fe3O4 magnetic polymers [69], crystalline hydrous titanium oxide [70], aminated polyacrylonitrile fibers [71], New cross linked hydrazide–based polymers [72], Different Nanoporous Materials [73] and manganese oxide coated sand grain (MOCSG) [74]. Many treatments for wastewater polluted with Chromium ions have been proposed, including an adsorption process, which does not have high effectiveness, unless the adsorbent material shows certain physicochemical and mechanical properties [75]. In recent years, some physical, chemical, and biological treatments on natural raw minerals have been performed in order to modify their structure, thus increasing the adsorption capacity or the selectivity [76]. Overall, adsorption is known as an efficient process for purifying contaminated waters. Also, treating wastes containing Chromium is an increasing concern for the industries and hence an effective solution needs to be found for the successful elimination of harmful metals from wastewaters. One solution is also to use activated granular carbon [77]. Over the last few years, several investigative works have been in order to explore an alternative to the expensive methods for treating wastewaters. Several kinds of materials have been used for the adsorption process to test their adsorption abilities. According to studies results it appears that the elimination of Chromium ions by utilize the low-cost adsorbents is increasingly favorable, especially in the long term [78]. This is because several materials are readily available, i.e. these exist naturally or are found in agricultural waste and manufacturing wastes, used as low-cost materials adsorbent. Previous researches show that there is increasing attention in investigating different materials that can serve as
low-cost adsorbent [79]. These include River bed Sediments [80], Sweet Orange (Citrus Sinensis) Peels [81], Orange Peel Activated with Potassium Carbonate [82], lignin [83], Spheroidal Cellulose [84], Arthrobacter nicotianae Cells [85], commercial grade granular activated carbon [86], Biogenic Magnetite Nanoparticles [87], modified sugarcane bagasse[88], vesicular basalt rock [89], bentonite [90], Composites of polyaniline with rice husk and sawdust [91], Cellulose from Wood Powder [92], Carbon [93], Sorghum Bicolor [94], Alligator weed [95], Boehmite Nano-powder [96], Ligand [97], goethite and kaolinite [98], Lewatit [99], Cane Papyrus [100], Rhizophora apiculata Tannins [101], modified coconut husk [102], tea residue [103], spent tea leaves [104], modified walnut shells [105], walnut, hazelnut and almond shell [106], groundnut hull [107], wheat bran [108], modified palm branches [109], natura and calcined rice husks [110], carrots (Daucus carota) [111], rice bran [112], soya cake [113], Tamarindus indica seeds [114], tamarind hull-based [115], coir pith [116], modified red pine sawdust [117], sugar beet pulp [118], Lentinus sajor-caju mycelia [119], activated carbon from the industrial waste of sugar [120], preparing activated carbon from biomass of walnut shell [121], preparing activated carbons from agricultural wastes [122], preparing activated carbon from bamboo locally available waste (Oxytenanthera abyssinica) [123], Hazelnut Shell Activated Carbon [124], Acid Activated Low Cost Carbon [125], activated carbon [126], microporous activated carbon from biomass [127], deriving activated carbon from agricultural waste materials and cloth fabric of activated carbon [128], Charcoal, Attapulgite and Date Palm Leaflet Powder [129], biomaterial-based activated carbons [130], bone charcoal [131], bentonite and expanded perlite [132], Natural Clay [133], kaolinite [134], inorganic-organic clay [135], green alga unicellular, Chlorella sorokiniana immobilized in loofa sponge [136], biological wastes and vermiculite [137], dead fungal biomass of Aspergillus niger [138], agricultural waste biomass [139], basalt-inhabiting bacteria [140], Aeromonas caviae [141], Arthrobacter oxydans [142], Brown Seaweed Ecklonia Biomass [143], fermentation waste [144], red, green and brown seaweed biomass [145], Arthrobacter Viscosus [146], shelled Morina oleifera seeds [147], bagasse fly-ash—a sugar industry waste [148], Potential tea factory waste [149], pomace-an olive oil industry waste [150], using response surface methodological approach treating sugarcane bagasse [151], membrane bioreactor with immobilized Pseudomonas cells [152], Lentinus sajor-caju, free and immobilized biomass [153], Eggshell [154], brown seaweed biomass [155], boehmite [156], biochars in different environmental conditions [157], Agave Lechuguilla biomass [158], multi-walled carbon nanotubes [159],nanohydroxyapatite [160], inorganic materials [161], moss chlorophyll fluorescence [162], graphite furnace atomic [163], natural biomaterials [164] and eucalyptus bark [165]. Using low-cost adsorbents like waste materials is very interesting because it's contributing to reducing waste disposal costs. Even though there has been a variety of adsorption studies. In addition, Chromium appears to be one of the major risk factors for several deadly diseases if the concentrations of Chromium go over the allowable boundary, as World Health Organization (WHO) recommendations. Basically, the health's effects can be largely specified by the oxidation condition, various values of guideline to trivalent chromium and hexavalent chromium must be derived. But the current analytical approach and the chromium changing speciation in water support the total chromium values guideline. Due to the carcinogenicity of hexavalent chromium through the inhalation and genotoxicity, questioned in current guideline value 0.05 mg/L, but for a new value, the available data of toxicology doesn't back up the conclusion. Practically measurement, 0.05 mg/L, that is considering to be away to give rise to significant risks to health, has been retained as a temporary guideline value until further information becomes available and chromium can be re-evaluated [166]. The presence of Chromium ions in waterways produced by industries can also affect water bodies, which further presents an unsafe effect on the marine and extra-terrestrial bodies. Amongst the exhaustive list of issues related to Chromium poisoning, one most common issue led by the exposure of Chromium is the occurrence of miscarriages and neonatal deaths [167]. Therefore, the utilization of these materials as low-cost adsorbents is acknowledged as a possible and economical application for wastewater treatment. This is reflected in the increasing numbers of periodicals, that show using low-cost materials as adsorbents in the literature. These mainly conclude the immense interest in finding appropriate adsorbents for the process of adsorption [168]. This review aims to provide an outline of the adsorption processes utilizing low-cost
adsorbents for eliminating Chromium ions from different sources. This will be achieved by underlining the characteristics of the adsorbents, their optimum parameters, and their adsorption capacity. The major aim of the manuscript is to offer a survey about the latest studies which are related to the adsorption processes using low-cost materials as adsorbents to Chromium ions elimination.

2. Sources and toxicity of Chromium ions.
A wide distribution for Chromium into a crust of the earth. It is found in oxidation conditions from +2 to +6. Little chromium quantities may consist in rocks and soils, it's in the trivalent state nearly. The compounds distribution content for trivalent chromium and chromium depending on potential redox, pH, the appearance of reducing or oxidizing compounds, total chromium concentration, trivalent chromium complexes formation of insoluble salts of trivalent chromium, and redox reactions kinetics. Trivalent chromium exists generally into an environment, as Cr(OH)₃(3-n)+ and hexavalent chromium as CrO₄²⁻ or Hcno₄⁻. Trivalent chromium predominates in the soil. in the organic matter, reduction of hexavalent chromium to trivalent chromium (III) is very easy, for instance, results of human's activities occurrences in soil [169]. Trivalent chromium forms a positive ion in water from complexes and hydroxides and at relatively high pH values is adsorb. The ratio is widely different between hexavalent chromium and trivalent chromium in the surface water, it's found in ecology at a high concentration. Generally, salts of hexavalent chromium are more dissolved from salts of trivalent chromium, which means the hexavalent chromium more activity. chromium present in the air into aerosols form and, it's possibly eliminated with dry and wet sedimentation from the atmosphere, it's emitted to the ambient both the hexavalent and trivalent chromium. Rarely available data of chromium types in the atmosphere due to the difficulties in analytical, but the hexavalent chromium percentage is about 0.01% to 30% that's according to the last studies. The chromium concentration in the rainwater is estimated between 0.2g/l to 1g/l. In seawater measured the natural concentration of chromium, it's between 0.04 µg/l to 0.5 µg/l, the chromium concentration has been found equal to 0.7 µg/l in the North Sea [170]. In natural surface water, the approximate total content of chromium is between 0.5 µg/l to 2 µg/l and between 0.02 µg/L to 0.3 µg/l for dissolved chromium content. In Antarctica lakes, the chromium concentration increases with depth between> (0.6 to 30) µg/l, and the chromium content in most surface waters is between (1-10) µg/l [171]. Industrial activity is the guide to polluted the surface water with chromium content, in the United States of America were found a levels of chromium up to 84µg/l; (0.2 to 44) µg/l the level of water concentrations in central Canada "[data from the National Water Quality Data Bank (NAQUADAT), Water Directorate Interior, Canadian Ministry of Environment, 1985]". Less than 10 µg/l of chromium levels in the Rhine and, in India, 50% of natural sewage contents less than 2µg/l of chromium [172]. Generally, in groundwater the concentration of chromium estimates to (> 1 µg/l). 0.7 µg/l is a mean concentration and 5 µg/L a maximum concentration has been measures in the Netherlands [170-172]. there are 1473 samples contains dug well water in India, only 50% had below 2 µg/l chromium concentration. In the USA groundwater, notified that the level of chromium concentration (>50 µg/l) in shallow groundwater also founds (2-10) µg/l media levels. Most providers in the USA contain fewer than 5 µg/L. In 1986, levels in 17 groundwater supplies and one surface water supply exceeded 50 µg/L. Approximately 18% of the population of the USA are exposed to drinking-water levels between 2 and 60 µg/l and > 0.1% to levels between 60 and 120 µg/l. In the Netherlands, the chromium concentration of 76% of the supplies was below 1 µg/land of 98% below 2 µg/l. A survey of Canadian drinking-water supplies gave an overall median level of 2 µg of chromium per liter, with maxima of 14 µg/l (raw water) and 9 µg/l (treated water) [173-174].The values for oral acute exposure (LD₅₀) to the rats are estimate between (185–615) mg of trivalent chromium per kg of body weight and (20–250) mg of hexanelant chromium per kg of body weight, depends on tests of the dichromats and chromic compounds, respectively [175]. Short-term exposure Three-month-old inbred BD rats (5–14 per sex per dose) were exposed for 90 days, 5 'd' per week, to 0, 2%, or 5% of insoluble, non-hydrated chromium (III) oxide (Cr₂O₃) pigment in feed [176]. The dose levels are equivalent to 0, 480, and 1210 mg of chromium (III) per kg of body weight per 20 'd'. Survival, body and organ weights, feed intake, blood analysis, and the macroscopic and microscopic appearance of major organs were not affected.
The only effect observed was a dose-related decrease in liver and spleen weights, ranging from 15% to 35% [177-178]. The emission of Chromium ions into the environment from industrial processes and car exhaust will pollute the surface and also underground waterways. This can result in pollution of soil while enhancing the overall pollution rate, especially when ores from mining processes are disposed of in landfill sites [179]. In addition, agricultural wastes in soils can consist of metals, which would then be consumed by plants thus resulting in the accumulation of such harmful substances in their tissues. It is expected that animals, which feed on the aquatic and plant life may also be poisoned due to the presence of harmful Chromium ions. Hence, it is vital that strict wastewater regulations are laid to reduce the environmental risks of dangerous substances [180].

3. Removal of Chromium ions

3.1 Traditional processes.

Several processes have been used for eliminating Chromium ions from polluted waters. These consist of biological, chemical and physical treatments. It is worth mentioning that usually these processes are mainly based on physical and chemical treatments. The overall traditional method to remove Chromium ions involves ion exchange, electro-dialysis, oxidation, chemical precipitation, membrane filtration, evaporation, reverse osmosis, solvent-extraction, and adsorption onto activated carbon. Chemical precipitation is the commonly used process for Chromium ions removal from inorganic effluents depending on the pH alteration in a basic solution [181]. Nevertheless, the chemical precipitation disadvantages are manifold. For details, discharge of too much sludge produced needs additional treatments, which slows the metal precipitation, chromium to inadequate settling and the aggregation of metal precipitates. In addition, there are several long-term ecological concerns with the disposal of sludge. Coagulation-flocculation is also used to process wastewaters with Chromium ions by adding a coagulant in the coagulation process. However, this treatment has the possibility of destabilizing colloidal particles and thus resulting in sedimentation. The several advantages and disadvantages of the conventional method are provided below in Table 1. In spite of these approaches being expensive, these are mostly the ones that can have a positive impact on the commonly occurring discharge issues [182]. Additionally, these methods are also feasible for treating polluted waters containing Chromium ions. It is known that issues usually are found in the traditional methods where there is increased usage of the reagent and energy, a low selectivity, increased operational costs and production of secondary pollutants taking place. Asides the traditional methods, it is vital now to explore alternatives for replacing these traditional approaches of eliminating Chromium ions from polluted water sources [183].

| Traditional treatments | Benefits | Limitations |
|------------------------|----------|-------------|
| Ion-exchange            | Metal-selective, Increased regeneration of materials. | Increased initial capital and maintenance costs |
| Chemical precipitation  | Simple operation, Non-metal selective, Cheap. | Increased production of sludge, Increased costs of disposal sludge. |
| Membrane filtration    | Reduced production of solid waste Reduced chemical consumption. | Increased initial capital and maintenance cost, Low flow rate. |
| Electro-chemical treatment | Metal-selective, Potential for treating effluent >2000 mg dm$^3$. | Increased initial capital cost. |
3.2 Adsorption

Over the past few years, the process of adsorption has attracted great interests, as it appears to be a favorable methodology for long-term effective treatments along with being an economical approach for chromium ions removal. Depending on the flexible design and simplicity of operation, adsorption is an important process nowadays. The term “adsorption” refers to the mass transfer from the solution (liquid phase) to adsorbent surface. Advantages of the adsorption approach in removing or minimizing the Chromium ions, also in soft concentrations involve the enhancement of the application of adsorption as a useful and practical approach. The effectiveness of the adsorption processes is almost categorized depend on the solution nature in which pollutants are spread, the molecule sizes and the polarity of the contaminant along with the type of adsorbent used. Adsorption also occurs based on the interactions between surfaces and species being adsorbed at certain molecular levels [184]. Adsorption can be classified for two methods; physical adsorption and chemisorption. Physical adsorption is a reversible phenomenon, which results due to intermolecular forces of attraction that take place in molecules of the adsorbent and the adsorbate. Meanwhile, chemisorption occurs as a result of the chemical interactions amongst solid and adsorbed substances. Chemisorption is an irreversible method that is also known by activated adsorption. Increased physical adsorption occurs at a temperature in the range of the critical temperature of a known gas while chemisorption takes place at a higher temperature than the critical temperature. Moreover, depending on the situation, it is probable that both processes take place either separately or at the same time [185]. It is important to ensure that various variables are monitored in the adsorption processes occurring between adsorbate and adsorbent. This includes the characteristics of physical and chemical for adsorbate and adsorbent, the concentration of adsorbate in a liquid solution, temperatures, pH and also contact times. In terms of pH, this accounts for the most important factor as compared to other parameters because of its capability for control on distribution of charge on surface of adsorbent among adsorbate ion. However, in most related studies, it's should be taking the zero-charge point (pHzpc) in the consideration for perform a comparison with pH as pHzpc regulates the limits of the pH of the adsorbent. pHzpc is the charge an adsorbent’s surface carries and can be known by the protonation and deprotonation of the adsorbate ion. Also, the surface charge density is dependent on the specific metal ions that respond in a direct manner with the adsorbent surface [186]. For instance, in cases of the pH values of the solutions being more than pHzpc, the adsorbent’s surface charge will be negative. Or else, the pH rise within a certain range can result in increasing the rate of adsorption rate. However, any additional increase in pH can result in the reduction of the adsorption rate. This is due to some adsorbate ions being unaltered by pH changes. As an alternative to pH, the adsorbent dosage is an additional feature, which influences the adsorption process. Moreover, with a rise in the adsorbent’s dosage, the adsorption rate also tends to increase. Nevertheless, the adsorption rates can reduce if the adsorbent’s dosage rises more. Due to available a larger number of occupied effective places, while the adsorbate concentration gradients are maintained constant. A higher adsorption rate can be obtained when the temperature increases, the surface area growth and adsorbent pore volume [146]. Initial metal concentration can be the mainspring to avoid mass transfers through the adsorbent surface and solution. The initial chromium concentration has an influence on the adsorption rate depending on the presence of the explicit functional groups surface and capability of functional groups surface for connect chromium ions (mostly when concentrations increases). Thus, any parameters influencing the adsorptive capacity of adsorbent should be considered during the adsorption process [185].

3.2.1 Adsorption Mechanism

Adsorption mechanisms are complex due to the non-existence of any simplified theory on Chromium ions adsorption onto surface of adsorbent. Earlier works have been observed to report on several models, which describe the mechanism between the adsorbute and the adsorbent. The Langmuir and Freundlich models, both, are often employed for describing the sorption isotherms [186]. In regard to kinetics, the pseudo-first-order and pseudo-second-order kinetic models can be employed for describing the sorption
kinetics. The thermodynamics of the metal ion sorption can be explained based on thermodynamic factors, for example, free energy ($\Delta G^\circ$), enthalpy ($\Delta H^\circ$) and entropy changes ($\Delta S^\circ$) based on the endo-thermal and exothermal sorption processes [187].

### 3.2.2 Adsorption Isotherm

Sorption isotherm can be referred to as the process of the interaction of adsorbate ions on the adsorbent’s surface. In the literature, various isotherm equations exist, which can be used to analyze the relevant experimental parameters. However, one of the well-known adsorption isotherm models, which is commonly employed for the single solute system, is the Langmuir [188] and Freundlich isotherm [189]. These are models more feasible to explaining the association between the adsorbed material quantity on equilibrium state, $q$, in mg/g and the remaining adsorbate concentrations at equilibrium for the bulk solution, $C$, in mg/l.

#### 3.2.2.1 Langmuir Isotherms

Depending on the Langmuir adsorption theory, particles tend to adsorb at known well-defined sites that are consistently dispersed over the adsorbent’s surface. These sites also have similar affinities for monomolecular layer adsorption along without interactions existing among molecules of adsorbed [18].

For Langmuir equation, it is written as:

$$\frac{1}{q_e} = \frac{1}{q_{\text{max}}} + \left(\frac{1}{b q_{\text{max}}}\right) \frac{1}{C_e} \quad \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdots \cdOTS
temperatures can be used to control the various thermodynamic variables. These include (ΔH°) enthalpy, (ΔG°) free energy change and and (ΔS°) entropy change [190-191].

The adsorption free energy (ΔG°) can be associated with the Langmuir adsorption constant through the following equations:

\[
\Delta G^o = -RT\ln K_c
\]

\[
\ln k_c = \frac{\Delta S^o}{R} - \frac{\Delta H^o}{RT}
\]

The value obtained from the thermodynamic parameters was numerically analyzed to forecast the sorption operations characteristics. The various heavy metal ions adsorption onto different adsorbents is a complex process where the thermodynamic variables of the metal ion sorption are influenced by the type of metal ion, type of sorbents, solution conditions, ionic strength and experimental circumstances [192].

For study the rate-limiting step in the adsorption operations, it’s should consider the contact time for experimental parameters as a based, relating to the kinetic energy. Overall adsorption processes can be regulated through steps such as pore diffusion, surface diffusion or a mix of more steps. Lagergen's first-order equation and Ho’s second-order equation are instances of kinetic models, which are oftentimes utilized to characterize these models of kinetic adsorption [193]. The pseudo-first-order kinetic equation of Lagergen's model is given as [193]:

\[
\frac{dq_t}{dt} = k_1(q_e - q_t)
\]

when \(q_e\) and \(q_t\) are adsorbed waste quantities(mg/g) in a balanced state, in any period instant \(t\) ‘min’, correspondingly. \(k_1\) is the pseudo-first-order rate constant of adsorption operation 'min'. Pseudo-first-order equation refers to "the assumption of the rate of change of solute’s uptake with time which is in direct relation to the change in the saturation concentration and the amounts of solid uptake overtime” [194]. The equation of pseudo-second-order kinetic given as:

\[
\frac{dq_t}{dt} = k(q_e-q_t)^2
\]

The model of pseudo-second-order is dependent on "proposition that the rate-limiting step may arise from the chemical adsorption", which involves the valence forces that take place due to the distribution or exchanging of electrons in the adsorbent and adsorbate [194]. In regard to removing Chromium ions, researchers in the past have also considered the soya cake removing these ions from the wastewaters. This was carried out at various optimal conditions for the initial metal concentrations, adsorbent doses, the solution’s pH and particle sizes. It was revealed that the ratio of adsorbent to the solution along with the metal ion concentration can have an effect into a metal ions quality removed. Most Chromium ions adsorption was about 98%, as the adsorbent doses were increased between (0.5 - 2.5) g per 500 ml at various concentrations of the ions, i.e., 100 mg/l, 200 mg/l, and 300 mg/l. The adsorption increases take place with the electrolyte concentration increases. It was noticed that the most metal uptake in soya cake waste took place at 125mg/gm, 105 mg/l and 85 mg/gm for Chromium ions, for pH reaching between (5.5-6.5), correspondingly. The isotherm adsorption analyzing for the data achieved at 25°C demonstrated that the equilibrium data for Chromium ions appropriate fit with both, the Langmuir and Freundlich isotherms. Also, Chromium ions were seen to have an increased affinity and adsorption rates at all experimental circumstances. Moreover, the study of kinetics revealed that Chromium ions uptake was faster with 95% or even a higher percentage of adsorption taking place within the first (20 – 30) 'min' of contact times. In addition, the kinetics statistics is suitable well with the model of pseudo-second-order, where correlation coefficients were found to be higher than 0.98 [113]. Table 2 lists some of the equation of the empirical models above [188-194].

The rise in the overall adsorption rate and capacity of Chromium was observed when smaller adsorbent particles(sawdust) [91]were used. In addition, investigative research was performed with varying pH (i.e. pH of 3, 6 and 7), varying temperatures (i.e.20°C,30°C, 40°C and 50°C) and adsorbent doses (i.e. 5gm to 20gm). The outcomes of this investigative study showed that adsorption capacities of clays were increased the chromium removal with increased in the solution’s temperature. The maximum capacity of adsorption revealed was 135mg/g in temperature of 50°C. Also, the adsorption process exhibited a Langmuir and Freundlich behavior, which was shown by the coefficient (i.e. R² > 0.985). An increase
in the percentage of trivalent chromium removal in the minimum pH solution is possible because of chromium ions lower content [91].

In modeling, the kinetic data is proportional to the first pseudo-rank model contrasted with the model of second pseudo-rank. Works on adsorption of Chromium (III) by inorganic and organic slurry waste in terms of isothermal, kinetic and thermodynamics, verified operation, which has endothermic (H°> 0), spontaneous (G° <0) and irreversible (S >> 0) [135] properties. Moreover, the Orange Peel Activated with Potassium Carbonate was also considered for removing Chromium from water and wastewater [82]. The works were performed as a function of: pH (i.e. with pH values in the range of 1.18 to 13.5), particle sizes (i.e. with sizes of 600, 420, 300, 150, 75 and <75 µm), doses (of 0.05, 0.1, 0.2, 0.5 and 1 g), contact time (of 3 'h') and temperature (in the range of 30-70°C). Findings revealed, adsorption optimum conditions are achieved when pH =6.5, particle size 75 µm, dosage =0.5g/100ml and 1 'h' contact time. Orange Peel Activated with Potassium Carbonate adsorption capacities used to Chromium removal reduces with a rise in the solution temperature, which shows that the adsorption process is impulsive [82]. The type of adsorbent is an important factor. Adsorption capacity depends on activated carbons, which is not feasible for use, currently, based on its highly expensive cost of production and operations. Activated carbon also needs a somewhat complicated mix of agents for improving the inorganic matters removal process from. As a result, the difficulties stated earlier, latest researches have looked into an alternative adsorbent with high regeneration capability, obtainability and cost-effectiveness to substitute the expensive activated carbons like (activated carbon prepared from walnut shell biomass) [121]. Up to the present time, several works have investigated the low-cost adsorbents to utilize. Agrarian wastelands along with natural materials all were examined as potentially low-cost adsorbents for treating wastewaters, plagued with Chromium ions [151].

| Types of mechanism | Equations | Nomenclature | Ref. |
|-------------------|-----------|--------------|-----|
| a. Adsorption     |           |              |     |
| Isotherm          |           |              |     |
| i) Langmuir       | \( \frac{1}{qe} = \frac{1}{q_{\text{max}}} + \frac{1}{(bq_{\text{max}})} \frac{1}{Ce} \) | qe is capacity of metal sorption in equilibrium, C is concentration of solute in solution at equilibrium, qmax and b are Langmuir constants linked to highest sorption capacity (monolayer capacity) and bonding energy of adsorption | [188] |
| ii) Freundlich    | \( qe = k_F C e^{ln} \) | KF is constant of a bio-sorption at equilibrium, qe is the capacity of sorption, n is a bio-sorption strength indicative constant. | [189] |
| b. Adsorption     |           |              |     |
| kinetics          |           |              |     |
| i) Pseudo first-order | \( (qe - qt) = logqe - \left( \frac{Kt}{2.303} \right) \) | qe & qt are the capacity of sorption in equilibrium and at time t, k is the constant of rate. | [193] |
| ii) Pseudo second-order | \( \frac{t}{qt} = \frac{1}{K2qt2} + \left( \frac{1}{qe} \right) \) | qe & qt are the capacity of sorption in equilibrium and at time t, k is the rate constant of pseudo-second-order sorption. | [194] |
3.2.4 Low-Cost Adsorbents

Recently, quite a significant amount of research has been carried out for obtaining materials, which can be utilized as low-cost adsorbents. These consist of natural materials, agricultural waste, and wastes produced from industries. Low-cost adsorbents refer to those materials, which are found abundantly in the environment or are byproducts or wastes from industries. Moreover, adsorbents are known as low-costs if they have reduced processing requirements. Previous adsorption works concentrated on plant wastes such as, the Peat and coconut fiber [195], carrots [111], rice husks [110], Cane papyrus [100], modified coconut husk [102], rice bran [112], and others, which can be utilized each on the normal compose or change several alterations physical or chemical. Converting these materials into adsorbents is an effective way of reducing the costs of waste disposal and for providing alternate treatments for replacing the commercially activated carbons [78]. Table 3 provides a summary of the outcomes of different works on adsorption, utilizing several adsorbents.

### Table 3. Adsorption capacities of Chromium (III) ions utilizing several different adsorbents.

| Type of adsorbent               | pH  | Contact time 'min' | Temperature (°C) | qmax (mg/g) | References |
|--------------------------------|-----|-------------------|------------------|-------------|------------|
| Peat and coconut fiber         | 7   | 80                | 25               | 1.25        | [195]      |
| Carrots                        | 2   | >120              | 22               | 26.2        | [111]      |
| Rice husk                      | 4   | >120              | 25               | 102.96      | [110]      |
| Cane papyrus                   | 6   | 90                | 25               | 154.76      | [100]      |
| Modified coconut husk          | 60  | 40                | 9.91             | 9.91        | [102]      |
| Rice bran                      | 60  | 6                 | 6.8              | 6.8         | [112]      |
| Sweet Orange Peels             | 60  | 40                | 82.31            | 82.31       | [81]       |
| Coir pith                      | 6   | 60                | 79.6             | 79.6        | [116]      |
| Groundnut hull                 | 6   | 60                | 40               | 40          | [107]      |
| Spherical Cellulose            | 6   | 60                | 16.35            | 16.35       | [84]       |
| kaolinite                      | 6   | 60                | 24.24            | 24.24       | [134]      |
| Carbon                         | 5.5 | 60                | 14.97            | 14.97       | [93]       |
| fermentation waste             | 6   | 90                | 35.17            | 35.17       | [144]      |
| eucalyptus bark                | 6.5 | 60                | 45.5             | 45.5        | [165]      |
| Soya cake                      | 5   | 90                | 23.4             | 23.4        | [113]      |
| Eggshell                       | 6   | 120               | 49.5             | 49.5        | [154]      |

3.2.5 Effects of pH

Metal ions adsorption from wastewaters is usually dominated by the solution’s pH. It is worth mentioning that the pH of the solution influences the surface charges on the adsorbent, the extent of ionization along the class of adsorbates. Over a known pH range, mostly metal sorption is improved with pH. However, this is valid for a known increase in pH, after which an additional rise in pH can lead
to a reduction in the metal sorption. The medium influences pH value on the system’s equilibrium, as the pH correlation can be expressed as Eq. [146]:

$$pH = pK_a - \log \frac{[AH]}{[A]}$$

where [A] and [AH], refer to deprotonated and protonated surface groups concentrations. Equilibrium constant, pKa, resembles carboxyl groups. The effects of pH on the Chromium ion uptake are also investigated for removing Chromium ions in utilizing the Cane Papyrus [100].

The pH values used were in the range of 3 to 7. It was seen that the highest bio-sorption occurs when the pH value was in the range of 2.5 to 5. The possibly by reason of a fact, a low pH value leads to the detachment of carboxylic acids, which further results in the production of carboxylate groups plus H+. A further rise in the pH results in an increase in the metallic ion bio-sorbed. Moreover, for a pH more than 6, a strong reduction is observed in the metal uptake. This is mainly because of the hydrolysis of the metal ion. The effects of pH on banana peels have also been looked into [100]. Furthermore, in terms of the Chromium Ions, the pH effects were also monitored, and it was observed that the adsorption abilities rose from 0.5 mg/g to around 3.25 mg/g with a rise in pH from 2 to 6. This can be based on the availability of the free ion, which exists at a pH lower than 6. Nevertheless, the adsorption capacities decrease after a further rise in pH (i.e. from a pH of 6 to 12). To elaborate, at lower pH values, the adsorption abilities are lesser than Chromium ions, which are competing with hydrogen ions for the binding site on the surfaces of the adsorbent. On the contrary, at increased pH values, the Chromium ions tend to precipitate in the solutions [196].

3.2.6 Effects of Temperature

Based on the adsorbent utilized, the relative temperatures can have an influence on the adsorption capacities. To elaborate, the thermal value is able to alter the adsorptive equilibrium based on the type of the procedure (i.e. whether it is exo or endothermic). Hence, it is vital to determine factors such as enthalpies, entropies and Gibbs free energies, prior to the conclusion of the procedure [74].

Gibbs free energy ($\Delta G^\circ$) is measured as the impulsiveness pointer of a chemical reaction. The connection between Gibbs free energy change, ($\Delta G^\circ$) [192], temperature and equilibrium constant, $K_a$, is expressed by Eq. (10):

$$\Delta G^\circ = -RT\ln K$$

The enthalpy, $\Delta H^\circ$, and entropy, $\Delta S^\circ$ changes on the adsorption procedure can be found from equilibrium constants as functions of temperature through the Van't Hoff equation, as can be referred to in Eq. (11):

$$\ln K_a = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT}$$

The percentage of Chromium ions adsorption by dried Gamma plant that increases with the rising temperatures from 25°C to 40°C has been investigated. Negative free energy change ($\Delta G^\circ$) values designate the impulsive characteristics of the adsorption process. Whereas, positive values of the enthalpy change ($\Delta H^\circ$) suggest the endothermic characteristics of the adsorption procedure. These findings are also reported due to a rise in the uptake capacities of the adsorbent with an increase in temperature. It has been found that the rising sorption capacities of the sorbent with temperature are due to the increase of pores and/or the activation of the sorbent surface [192]. Additionally, positive values of entropy ($\Delta S^\circ$) show the increased extents of free active sites at the solid-liquid interface during the adsorption of Chromium ions on dried plants.

3.2.7 Effects of Contact Time

Adsorption of Chromium ions adsorbent also is dependent on the connections of functional groups concerning the solution and the surface of the adsorbent. Adsorptions can be considered to conclude when an equilibrium stage is obtained with the solute of the solution and the adsorbent. Nevertheless, a certain period of time is required to maintain the equilibrium connections to confirm that the adsorption process has been completed. The effects of contact times on walnut, hazelnut and almond shell for the
adsorption elimination of Chromium ions from aqueous solutions are observed [106]. The experiment measures the effects of contact times under the initial concentrations of the batch adsorption as 25 mg/L and a pH of 5.5 for Chromium ions. The increased contact time improved the adsorption of chromium ions. Conversely, the quick adsorption had an initial effect on the overall required time to obtain equilibrium. For walnut, hazelnut and almond shell, the equilibrium time was 2.5 'h', for Chromium adsorption while for Cane papyrus, 2 'h' were required to attain an equilibrium for the adsorption of chromium ions. Hence, a 3 'h' contact time was maintained as an optimal time for future studies. The adsorption of Chromium ions on Cane Papyrus is observed to have taken 120 'min' as the optimal time for future studies. The ranges of contact times ranged from a minute to 3 'h'. However, the significant removal of Chromium ions occurred during the first 30 'min' where no considerable variations in terms of the removal were observed after 2 'h'. The adsorption of Chromium ions is originally higher mainly because of the existence of increased surface areas of Cane Papyrus for adsorption. All further experimental works maintained an equilibrium time of 2 'h' for removing Chromium ions onto Cane Papyrus [100].

3.2.8 Effects of Adsorbent Dose
Adsorbent dosage is a useful variable in determining the adsorbent’s capacities at known concentrations of the adsorbate. The effects of adsorbent doses on Cane Papyrus powder for adsorbing Chromium ions from aqueous solutions are examined. At room temperature, the adsorbent dosages were changed from (5 to 30) mg/l along with an initial concentration of 10 mg/l. Results achieved from this work describe the adsorption of Chromium ions, which improves and advances when doses of Cane Papyrus powder are amplified from (5 to 20) mg/l. This explains the increased accessibility of surface areas at increased adsorbent concentrations. Additional increases in the adsorbents will not have any effect on the adsorption because of the overlying adsorbent particles’ sites [197].

3.2.9 Effects of Initial Concentration
Initial concentrations of Chromium ions can modify the effectiveness in terms of removing metals based on a mix of features. These include the existence of specific surface functional groups in addition to the capability of these groups to bind Chromium ions. Moreover, this initial solution concentration can act as a vital factor in overcoming the mass transfer resistances of Chromium ions concerning the aqueous and solid phases [198-199]. The rapid adsorption of Chromium using Cane papyrus after 30 minutes before it continues at a faster rate and achieves saturation has been examined [100]. As the initial concentration of Chromium rises from 10, 20 and 30 ppm, the adsorption removal decreased, which was mainly because of the lower concentrations, where almost all Chromium ions were adsorbed rapidly on the outer surface. Nevertheless, a further rise in the Chromium ions initial concentration resulted in rapid adsorbent saturation [201].

4. Conclusions
The discharge of wastewater containing chromium from many different industrial processes to the environment has been on the rise. Chromium removal from wastewater varying techniques have been developed. Some effective electrochemical strategies for the reduction of Chromium in aqueous solutions were reported herein, such as electrode-ionization, electro-dialysis, reduction, and electro-coagulation. A detailed review of severing allow-cost adsorbents is discussed in this paper, which reveals the efficiency and scope of using low-cost adsorbents for removing Chromium ions utilizing an adsorption procedure. The adsorptive capacity depends on the adsorbent utilized nature also on type of wastewaters under treatment. Further investigative works need to be performed in order to develop an improved understanding of the adsorption processes of low-cost adsorbents as an alternative to endorsing the use of non-conventional adsorbents on a large scale. These works involve the structure works of adsorbents, batch investigations on the parameters that influence adsorption, adsorption modeling's like isotherm, kinetics, and thermodynamics, the Chromium ions recovery and the improvement of adsorption capacities through the alteration of adsorbents.
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