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Review Article

Low Cost Adsorbent Derived from Agricultural Byproduct and Its Application for the Removal of Cationic Dye from Waste Water: A Review

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Abstract

Water is the most important natural resource in the planet and is essential to life. But human activity such as industrial waste, and natural sources may all pollute this priceless resource. Among those, the effluent discharged from industries is the major pollutant in the water body. The development of industries is the backbone of the economic growth of countries, results in increased employment, and improves the living standards of human beings. But it is also the main source of environmental pollution. The primary cause of the cationic dye contamination in the water body is the organic effluent released by the textile industry. One of the cationic organic dyes generated by the textile industry is methylene blue (MB). This poisonous material hinders photosynthesis, keeps light from penetrating the water, and causes irreversible harm to people, animals, and plants. Therefore, different treatment methods were employed, including sedimentation, coagulation, oxidation, osmosis, electrolysis, and adsorption. Among them, adsorption is a highly effective technique for removing MB from waste water because of its affordability, environmental friendliness, accessibility, and high efficiency. Therefore, this review concentrated on different types of adsorbents made from agricultural products. Based on a study of many research, it appears that using agricultural wastes as an adsorbent can effectively remove MB from industrial waste water.

Keywords

Activated Carbon, Adsorption, Cationic Dye, Agricultural by Product, Waste Water

1. Introduction

One of the most important natural resources for all life on Earth is water. Water quality and availability have always been major factors in deciding not just where people may live but also how well they can live. The world we live in now has an environmental disorder with a significant problem of water pollution as a result of the fast rise of humanity, society, science, and technology [1].

The contamination of water bodies as a result of human activities, in such a manner that negatively affects its legitimate uses is called water pollution. It's the effect of alteration of the physical, chemical and biological properties, and the discharge of different waste such as liquid, solid and gaseous

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substance into water. Then it is harmful to public health, domestic, agricultural, animal and aquatic organism [2]. Since most animals and plants are constituted of 60% water by volume, water pollution is a prominent topic in environmental studies because of the importance of water and it's necessary for life. Water pollution can be categorized into two groups: the first group includes pollutants that come from a single source, like industrial wastes, mining operations, sewage and wastewater, pesticides and chemical fertilizers, energy consumption, radioactive waste, urban development, etc. The second group includes pollutants that come from multiple sources. The environment's pollutants include the effluents from industrial waste, which are one of the many sources of water pollution [3].

The foundation of a nation's economic expansion and development is industrialization, which raises the number and variety of manufactured goods produced, increasing employment and raising people's standards of life [4]. However, industrial operations may have detrimental effects on the environment, contributing to air and water pollution, resource depletion, and climate change [5]. The rise in industrialization has resulted in a greater release of hazardous chemicals and effluents into the air, land, and water. The many species that live in those particular habitats have been severely and irreversibly destroyed by these effluents [6]. Throughout their entire lifecycle, from the production of fibres through manufacturing, distribution, consumer use, and the end of the clothes' useful life at disposal, the textile industries are among those manufacturing sectors that consume enormous amounts of natural resources, including water and land. As a result, they contribute significantly to environmental pollution by releasing large volumes of waste water into the environment [7]. Since the textile industry is one of the most chemically demanding sectors of the world and a major polluter of clean air, its effluent has caused significant pollution problems. One of the main issues is how to dispose of wastewater from the dyeing industry because it contains various toxins, odours, and colour. [8]. Due to its visual nature, colour is the most easily identifiable pollutant among them. Fortunately, the negative impacts and illnesses caused by these hazardous colours in residential areas include allergies, jaundice, heart abnormalities, skin irritation, and tumours. [9]. Furthermore, because of their obvious colour, these harmful dyes are killing aquatic life by preventing enough sunlight from penetrating the cells for photosynthesis [10].

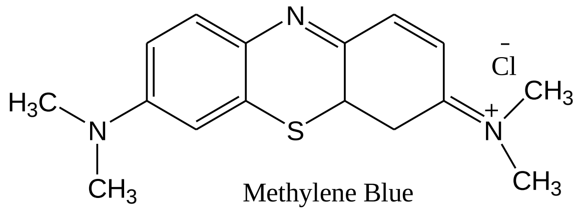
2. Textile Dyes and Effects

The bulk of textile dyes are complex organic compounds that must be resistant to deterioration. Textile dyes can be manmade or natural substances that have a tendency to stick to surfaces of materials like textiles to change their colour [11]. Many industries, including the food, rubber, plastic, paper, cosmetics, textile, leather, and pharmaceutical sectors, utilize a lot of dyes, which contribute significantly to water pollution [12]. Those industries may release wastewater that contains several types of organic chemicals and hazardous substances that can be harmful to animals and microbiological communities, as well as carcinogenic to them [13]. Dyes may interfere with bacterial growth and aquatic plant photosynthesis by absorbing and reflecting sunlight that enters the water. The complicated aromatic structures of the dyes make them ineffectual in the presence of heat, light, bacteria, and even oxidizing agents, making the breakdown of the dyes more difficult, making the issues worse [14]. Furthermore, it can seriously harm humans by impairing their kidney, liver, brain, reproductive system, and central nervous system. Dyes' harmful effects have led to a great deal of worry about their use. Thus, it is desirable to concentrate on certain techniques and technologies to remove colours from various types of wastewater streams as shown in shown below in Figure 1 [11].

Figure 1. *Effluent discharge from textile industries.*

3. Cationic Dye (Methylene Blue)

MB dyes are called cationic dyes, which are soluble in water and form cationic solutions. It has intensity of color and is highly visible even in very low concentration. While dyes consist of metals, aromatics, and other substances that can be poisonous to some aquatic species, they may also have an adverse effect on the photosynthetic activity of aquatic life by reducing light penetration. Although they are the most vivid and luminous of all the synthetic dyes, they are not very light- or wash-fast [11]. Methylene blue (MB) is a common dye used to colour cotton, silk, and other fabrics. MB may result in eye burns, which have the potential to permanently harm both human and animal eyes. Inhalation may result in brief episodes of fast or labored breathing, whereas oral consumption causes a burning feeling and may induce nausea, vomiting, excessive perspiration, and disorientation [13]. The chemical structure of MB and some physico-chemical properties are presented in scheme 1 and Table 1 respectively.



Scheme 1. *Structure of methylene blue [15].*

Table 1. *Physicochemical properties of MB dye.*

Molecular formula	$C_{16}H_{18}N_3SCl$
IUPAC name	3,7-bis[Dimethyl amino]-phenothiazin-5-ium chloride
Molar mass [g.mol^{-1}]	319.85
λ_{max}[nm]	664

4. Treatment Methods

Typically, industrial wastewater treatment procedures include the following steps: Prior to treatment Equalization and neutralization are pretreatment processes used on industrial wastewater streams before they are released into municipal or even central industrial sewage systems [16]; then undergo primary wastewater treatment is to eliminate contaminants from the system with the least amount of work possible. Chemical or physical separation techniques are used to remove suspended particles. Bacteria are the main microorganisms used in secondary treatment, which maintains the waste components. Adsorption, ion exchange, stripping, chemical oxidation, and membrane separations are among the procedures that make up the third phase, which is physical-chemical treatment [1].

Currently, many physical, biological, and chemical techniques are used for dye removal from waste water such as sedimentation, coagulation, oxidation, adsorption, reverse osmosis, ion exchange, electrolysis, etc., but these methods still have their own advantages and disadvantage for removal of dyes was shown below in table 2 [18]. Adsorption is one of these methods that works well for eliminating dyes by employing an adsorbent medium [15]. High surface area, strong mechanical, chemical, and physical stability, and a strong affinity to bind with pollution molecules are characteristics of the perfect adsorbent. Additionally, a number of other variables, including pH and temperature, the adsorbate's concentration and structure, its degree of ionization, the ionic strength of dispersion, and the adsorbent's surface charge and structure, all affect how well an adsorption process works [19].

Table 2. *Advantage and disadvantage of some technologies for dye removal [17].*

Technology	Advantage	Disadvantage
Chemical precipitation	Permanent, immediate result, efficient, easily implement, and easy to monitor	High cost, not applicable for all case, required power, and may generate waste
Ion exchange	Ability to remove at low concentration	Expensive, and easily fouled
Membrane filtration	Small space required, simple, rapid, and effective at high concentration	High cost, low throughput and rapid membrane clogging
Biological method	Simple, economical and well acceptable	Inefficient on non-degradable compounds and slow process
Oxidation	Rapid, good elimination of color, odor, and efficient process	Chemical requirement and a few dyes are more resistant to treatment
Adsorption	Simple, economical and adaptable to many treatment	Limited capacity, selectivity

5. Related Studies on Removal of Methylene Blue

Several literatures agricultural byproducts have underlined their importance in methylene blue removal. Different agricultural byproducts were used for the preparation of adsorbent. Among this some of them are, Water melon rind [20], date stones, [13], bamboo [21], tea waste [22], rice husk [23] and coconut shell [24]. Some of the literature is listed below in Table 3.

Table 3. *A variety of adsorbents that are used to remove toxic Methylene Blue dyes from industrial waste.*

No.	Raw Materials	q_{max} (mg/g)	References
1	Moringa Stenopetala seed husk	436.68	[49]
2	Coconut shell	320.50	[24]
3	Coffee husk	6.826	[46]
4	Moringa oleifera pod	78.53	[50]
5	Tea waste	238.1	[51]
6	Rise husk	344	[52]
7	Walnut shell	315	[53]
8	Prickly pear seed cake	336.12	[54]
9	Bamboo	454.2	[21]
10	Leaves of pine trees	40	[23]
11	Beech sawdust	9.87	[23]
12	Groundnut	164.90	[8]
13	Vetiver roots	423	[34]
14	Peach stone	412	[41]

Watermelon peel is a by-product from watermelon fruit and its fresh juice shop waste, which have no value among commercial and possibility to be converted into valuable adsorbent by activated with H_2SO_4 activation. Watermelon rind contains hydroxyl, carbonyl, and carboxyl groups. This makes it capable of binding cations from aqueous solutions [25]. The application of waste as a reusable raw material for preparing carbon contain adsorbent will decrease the high number of waste discharge, decrease environmental pollution and yield valuable products. The watermelon peel in carbonized form was reduced to a particle size of 250–500 µm.

The adsorption study was performed at contact time 180 min, pH 5.60, adsorbent dosage of 0.08 g, initial dye concentration, and 100 mg/L shaking speed of 110 rpm. The adsorption kinetics were best described by the pseudo-second order model. With a maximal adsorption capacity of 200 mg/g, the Langmuir model describes the isotherm adsorption equilibrium. According to thermodynamic studies, the adsorption process is endothermic and spontaneous. Good adsorption efficiency for the elimination of Mb was given by WMR. Therefore, carbonized watermelon peel is one of the potential agricultural wastes to remove MB dye from industrial effluent [20].

Date stones are one of agricultural wastes which have been used as raw material for activated carbon preparation through zinc chloride chemical activation. Activated carbon from Date stones is prepared by impregnating with activating reagent ($ZnCl_2$) and 500℃ carbonization temperatures for 1hr [26]. From the experimental result the moisture content, ash content, surface area and iodine number are 10.86%, 2.04%, 1045.61, and 1008.86 respectively. Equilibrium adsorption data was best fit with the Sips isotherm equation with a maximum adsorption capacity of 398.19 mg/g obtained at 4.5 hr contact time, 450 mg/L initial MB concentration, 7 pH value and adsorbent dose of 0.5 g. Kinetic adsorption model of methylene blue was best fit with Pseudo-second order. Generally, the date stone is the best raw material for activated carbon preparation with high adsorption efficiency [13].

Bamboo is one of the most varied plant families in the grass family. It belongs to the Poaceae family's subfamily Bambusoidae. According to Scurlock et al (2000) by about 1500, the uses of bamboo for commerce had been identified [27]. Bamboo takes many months to grow, making it a plentiful natural resource. Because bamboo is a sturdy, resilient, and affordable material, it has been employed as the structural material for stairs at building sites in China, India, Malaysia, and other nations. The issue of treating industrial wastewater can be resolved by turning bamboo into a valued commodity like activated carbon. After impregnating the raw bamboo material with potassium hydroxide, it was carbonized for one hour at 700 ℃ in a nitrogen environment. With an adsorbent dose of 0.2 g, a pH of 7, a contact duration of 48 hours, and an initial dye concentration ranging from 100 to 500 mg/L, batch equilibrium tests were carried out. With a maximum adsorption capacity of 454.20 mg/g, the homogenous nature of the bamboo carbon surface is shown by the Langmuir isotherm model as the conformation of the experimental results. The pseudo-second-order model provided the most accurate description of the methylene blue adsorption on bamboo activated carbon [21].

As tea is the most popular drink in the world, a significant amount of waste tea is produced every day. It takes a long time for it to biodegrade and is an oxygen-demanding contaminant. Tea waste is the best cost effective precursor for preparation of activated carbon by chemical activation method and a good adsorbent for methylene blue removal [28]. An efficient adsorbent for removing effluent from aqueous solutions has been developed using tea waste [29]. The tea waste, which had been dried and powdered, was combined with a 3.5:1 impregnation ratio and allow to stand at room temperature for 24 hr. After that, it was dried and neutralized with distilled water. The activated carbons that were synthesized were kept in sealed containers until they were needed. According to this study, tea waste activated carbon has a maximum adsorption capacity of 238.1 mg/g at room temperature (25℃) with an adsorbent dose of 0.1 g and an initial dye concentration of 10 mg/L. Freundlich isotherm and pseudo-second-order model, respectively, describe the study of equilibrium isotherm and kinetic model. Employing tea waste as a very potential adsorbent to remove methylene blue dye from an aqueous solution [22].

Rice husk is one of the widely available agricultural waste generated in various rice-cultivating countries [2]. Approximately 500 million metric tonnes of rice are produced worldwide, with 10 to 20 percent consisting of rice husk. The organic substance that makes up 70 to 85 percent of dry rice husk contains mostly carbohydrates, lignin, cellulose, and other materials. Its composition includes cellulose (32.24%), hemicelluloses (21.34%), lignin (21.44%), mineral ash (15.05%), and a significant amount of silica (about 96.34%) in the mineral ash [30]. Rice husk activated carbon is a cheap adsorbent that is made by activating sulfuric acid and zinc chloride and is used to remove methylene blue dye from aqueous solutions. The highest methylene blue uptakes by activated rice husk carbon were reported to be 97.15% under ideal circumstances, which included a pH of 10, an initial methylene blue volume of 50 mL, an adsorbent dose of 3 g, and an initial methylene blue concentration of 4.0 mg/L. According to the findings, commercial activated carbon is more expensive than rice husk activated carbon when it comes to treating waste water to remove basic dyes (MB) [23].

Coconut shell is one among agricultural waste and it's available in large quantities throughout tropical countries. It's one of the best agricultural waste used as a precursor for preparation activated carbon [31]. According to Oribayo *et al* (2021) the coconut shell activated carbon was prepared by impregnation with $Zncl_2$ activation and carbonized at 600 ℃ for 3hr [24]. The maximum adsorption capacity of Mb on CASC was 320.5 mg/g at a pH value of 7, 0.02 g adsorbent dose and 4.5 hr contact time. Adsorption isotherm of the equilibrium data for Mb was fits Langmuir isotherm with a correlation coefficient (R^2) value of 0.9923. The kinetic data was best fitted with pseudo-second order model. The calculated thermodynamic parameters indicate that the process is spontaneous and endothermic nature [24].

6. Type of Adsorption Processes

The process of atoms, ions, biomolecules, or molecules of gas, liquid, or dissolved solids adhering to a surface is called adsorption, and it leaves a film on the adsorbent's surface. Adsorbents can be attracted to the atoms on the adsorbent's surface because they are not completely encircled by other atoms. Although the specifics of the species involved determine the exact form of the bonding, the adsorption process is commonly categorized as follows [32]:

Physisorption: if there is a physical attraction between the adsorbed molecules and the solid surface. Reversible adsorption occurs when van der Waals forces, which are weak attractive interactions between adsorbed molecules and the solid surface, are present during physical adsorption [30].

Chemisorption: The adsorption process is known as chemisorption if the attraction forces are the result of chemical bonding. It is challenging to remove chemisorbed species from the solid surface due to the stronger affinity in chemisorption [33]. Adsorption mostly occurs on the pore walls or at certain locations inside the particle. Adsorbate and adsorbent are bound together by particular attractive forces during the adsorption process. These forces may be the result of strong chemical bonds or weak Van der Waals forces [30]. It has been discovered that adsorption is better than alternative methods in terms of initial cost, insensitivity to harmful contaminants, ease of operation, and flexibility and simplicity of design. Furthermore, hazardous chemicals are not produced via adsorption [11]. Two other factors that may be taken into account in the adsorption process are kinetics, which controls the pace of the adsorption process, and thermodynamics, which is related to the final equilibrium interfacial energy [34].

7. Mechanism of Adsorption

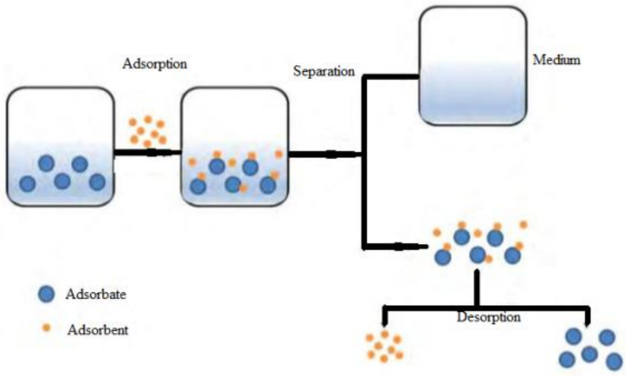


Figure 2. *Basic terms of adsorption theory [11].*

Three stages comprise the classical mechanism of adsorption: Adsorbate diffuses to the adsorbent surface, migrates into the adsorbent's pores, and builds up as a monolayer on the adsorbent. The process of adsorbate dispersion is shown in Figure 2 [35]. Adsorbate diffuses onto the adsorbent surface in the first stage due to intermolecular interactions between the adsorbent and adsorbate. Step two may be impacted by the agitation force and dye concentration. Step 4 is based on the characteristics of the dye molecules, such as their cationic and anionic structures, whereas step three is the rate-determining stage that will impact the adsorption of dye molecules on the substrate. It is essential to emphasize that step three may encompass two distinct occurrences [36].

8. Low Cost Adsorbents

Precursor selection for the development of cheaper adsorbents is affected by a number of characteristics, including availability, affordability, lack of toxicity, and cost, which is a crucial element to consider when comparing sorbent materials [10]. Considering their abundance in nature and little processing needs, it is reasonable to presume that waste materials and by-products from agriculture and other sectors are low-cost adsorbents [37]. Several low-cost adsorbents have been employed in the dye removal process. The majority have undergone testing and been suggested for the removal of dyes. Waste treatment by adsorption using low cost adsorbent is a demanding area as it has double benefits i.e. water treatment and waste management [14]. Agricultural wastes are abundant and have low economic value. Their current deposition causes significant environmental degradation. Agricultural wastes are also a rich source of activated carbon production due to their reasonable hardness and low ash content. Therefore, converting agricultural wastes into activated carbon is a promising solution to address environmental issues and lower the cost of preparing activated carbon [10, 38].

9. Activated Carbon

Activated carbon is a non-graphite form of carbon that can be made from any carbonaceous material. It is becoming more and more popular as an affordable and reliable mass separation agent for the removal of surfactants to improve the quality of the final product in many industrial processes. It is also used in catalysis, liquid and gas purification, and mixture separation [39]. Adsorption by activated carbon is one of the physical, chemical, and biological methods or a suitable combination of them that can be used to treat textile wastewater before it is safely released into the environment. This is because the adsorption process has been shown to be superior to other techniques in terms of its ease of operation, high removal efficiency, and easy design [40]. When AC is produced through different physical or chemical activation methods, the biochar develops qualities such as large surface area, extremely porous structure, high thermal stability, and high acid and basic stability with varying surface functional groups [41]. Activated carbon, which is produced from agricultural waste and has the desired chemical and physical properties, is a standout material that has gained significant interest. It can be used as an adsorbent in a variety of purification processes, including the removal of metal, cleaning of gas, and wastewater treatment. It also has a large surface area, high pore volume, and a high amount of surface functional groups [42].

9.1. Advantage of Activated Carbon over Other Methods

There are different treatment methods for effluent discharged from industrial waste containing dyes. The traditional methods for the treatment of colored wastewater are physical treatment which includes adsorption techniques, electrolysis, reverse osmosis and membrane filtration, chemical treatments which include coagulants and flocculants, and biological treatments include adsorption by microbial biomass, fungal decolorization, bioremediation systems and microbial degradation [43]. These technologies do, however, have benefits and drawbacks. The majority of these traditional techniques are inapplicable on a broad scale. Due to the high cost, short lifespan, inability to achieve sufficient colour elimination, and disposal issues brought on by the process's significant output of sludge at the end [18]. The adsorption procedure is reportedly the most efficient way to remove contaminants water out of all the treatment methods. Given its relative ease of use and low-cost application costs, adsorption is regarded as a promising technology with significant applicability in the discoloration process. Activated carbon derived from agriculture is thought to be comparatively better than other methods because to its low cost, availability, high efficiency, and capacity to separate huge amount [44].

9.2. Preparation of Activated Carbon

Chemical, biological, and thermochemical processes are used for the conversion of agricultural waste into sorbents among them the thermochemical approach is often preferred because of faster processing duration, higher product, employment of entire agricultural waste, and energy proficiency. Different low cost agricultural waste is used for the purpose of preparing activated carbon and used for the industrial waste water.

Coconut shell had been one of agricultural wastes used for preparation of activated carbon. A study has been conducted by Oribayo *et al* (2021) on preparation of activated carbon from coconut shell [24]. The authors impregnated the coconut shell with $ZnCl_2$ solution for 8 hr and carbonized it at 600 ℃ for 3 hr. Another optimization study conducted by Ayalew and Aragaw (2020) on coconut shell showed that the activated carbon prepared by using response surface method with H_3PO_4 as activating reagent has an optimum carbonizing temperature of 416 ℃ and time 19.5 min [45].

Coffee husk is another important agricultural waste used for preparation of activated carbon. Ayalew and Aragaw (2020) on their study prepared activated carbon from coffee husk by impregnating with H_3PO_4 (50% v/v) for 24 hr [46]. Then activated coffee husk was carbonized at a temperature of 400 to 600 ℃ with Muffle Furnace for 30 to 90 min of activation time to develop porosity and high adsorption efficiency. Faisal and Kana (2022) also reported that the coffee husk is used for preparation of activated carbon [47]. In this study the coffee husk was impregnated with $ZnCl_2$, $NaHCO_3$ and KOH with ratio of 1:1. The entire activated coffee husks were carbonized at 750 ℃ for 1 hr under inert atmosphere.

Moringa oleifera is the agricultural by product which is very important precursor for preparation of activated carbon and used for the treatment of different industrial effluents. It has the potential to be the promising precursor for the production of activated carbon. The preparation of activated carbon from Moringa oleifera seed pod were successfully done by chemical impregnation with $ZnCl_2$ and H_2SO_4 and carbonized at temperature of 600 and 800 ℃ in muffle furnace under inert atmosphere. The surface area and pore diameter of prepared activated carbon from Moringa oleifera seed pod has reported as 853.68 m^2g^{-1} and 2.13 nm respectively [48].

10. Conclusion

This review showed the promising findings of the research on getting more economically feasible and effective ways of treating wastewater using activated carbon prepared from agricultural waste. The various low-cost adsorbents made of agricultural waste material have been presented in this review. The efficiency of the removal of these low-cost adsorbents is recommended for the treatment of cationic dyes from industrial waste water due to their relatively low cost, ease of availability, reusable nature, and high adsorption efficiency. The preparation of adsorbent from agricultural products and the removal process through adsorption need further investigation by using different models to enhance efficiency and recovery. In addition to this, the reusability of this waste material requires more studies. However, this review predicts that the performance of those low-cost agricultural-based adsorbents is eco-friendly and has high efficiency to remove cationic dyes. Yet, more studies are needed in pursuit of preparing activated carbon from locally available agricultural wastes.

Abbreviation

MB Methylene Blue
WMR Water Melon Rind

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Data Availability

All data and materials are mentioned in the paper.

Conflict of Interest

The authors declare no conflicts of interest.

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