Review article

Agricultural-based Biomass as an Efficient Adsorbent in the Removal of Dyes in Dye-contaminated Wastewater: A Mini Review

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Abstract

1. Introduction

Water is vital for survival; however, people seem to have forgotten to take care of it. Due to industrialization and rapid population growth, water pollution problems have worsened over the years [1]. According to a scientific/statistical report, some 80% of the world's wastewater, most of which is not treated, is disposed of into the environment, polluting rivers, lakes, and oceans [2].

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Along with the rapid increase in the human population and the demand for industrialization, industries of various kinds have sprouted, contributing significantly to increased wastewater discharge in the environment [3, 4]. While we acknowledge the importance of water, many people still neglect the concept of sustainability in water resource usage, as evidenced by the high-water pollution presence globally that exacerbated the water scarcity problem. Because of this, the demand for safe freshwater has dramatically increased over the years on a global scale [5].

Apart from water scarcity, another significant problem is water pollution poses harm to human health. The United Nations World Water Development Report 2020 reported that 2 billion people currently do not have access to safe drinking water [6]. In the Philippines, nearly five million people rely on unsafe and unsustainable water sources [7]. Also, several experts have concluded that water contamination has created many health problems for humans due to the characteristics of the pollutants present in our water bodies [3, 8, 9]. Among these known pollutants, dyes contribute significantly to the global water pollution load.

One major industry that significantly contributes to water pollution is the dyeing industry. The dyeing industry involves the manufacture of dyes and the use of those dyes in a range of industries like textiles, printing, leather, and plastics industries [10]. The textile industry, for example, produces dye-contaminated wastewater that is harmful to both humans and the environment when it is released without proper treatment. Textile dyes significantly affect the aesthetic quality of water bodies and increase the biochemical oxygen demand (BOD) and chemical oxygen demand (COD). As reported by Lellis *et al*. [11], dyes also impair photosynthesis, inhibit plant growth, penetrates the food chain, offer recalcitrance and bioaccumulation, and may cause toxicity, mutagenicity, and carcinogenicity. Although this industry is undeniably vital as it contributes about 7% of the total world exports amounting to about 1 trillion dollars [12], it is also among the top contributors to pollution, including untreated dye-contaminated effluents that get released into the environment. As long as this industry is present, the problem of dye-contaminated wastewater will exist.

Several technologies, such as the advanced oxidation process [13], ion exchange [14], coagulation/flocculation [15], ozonation [16], and adsorption [4, 9, 17], are currently used in the treatment of dye-contaminated wastewater. However, given its convenience, ease of operation, easy designs, and cost-efficiency, adsorption is one of the best methods to eliminate different dyes present in the water system [18-23]. Although many researchers use various adsorbents in adsorption, activated carbon is still one of the most commonly used methods. Activated carbons are usually produced from finite resources, such as coal, lignite, peat, and petroleum residue materials, which are expensive and need intensive regeneration [24]. This is why many researchers nowadays look for alternative sources of activated carbon that are less costly, environment-friendly, and sustainable [3]. One of these alternatives is the use of agricultural waste and residues.

This work reviews relevant literature that describes the efficiency of agricultural wastes and residues used as adsorbents in removing dyes in dye-contaminated wastewater. The efficiency of the different agricultural biomass-derived adsorbents in treating different dye substances will also be compared. Hence, researchers may be able to conceive an idea for their future research work related to the utilization of agricultural waste and residue as adsorbents in treating wastewater.

2. Overview of Dyes as a Pollution Source

Dyes are soluble organic compounds with a very high solubility in water, making them difficult to remove by conventional methods. They are mainly used to impart color to a given substrate due to the existence of chromophoric groups in their molecular structures [11]. Many industries worldwide contribute to the release of dye pollutants into the environment (Figure 1). The textile

and clothing industries, among other known dyeing industries, are undeniably essential to our lives. These industries do not only provide us with our needs in terms of their products, but they also offer jobs to many people. The global textile industry is estimated to be worth US\$1 trillion worldwide, accounting for 7% of total world exports and employing about 35 million people [12]. Nevertheless, the textile industry is considered one of the critical causes of pollution globally.

Figure 1. Industries that release dyes into the environment [25]

Textile industries generate 125-150 L of wastewater/kg of products, with an annual production of over 7×10^5 tons/year, and 100 tons/year of dye is discharged into water streams [13, 26]. These industries are also responsible for consuming large quantities of water and chemical products, and generating effluents with high staining, turbidity, BOD, and toxicity [15]. The level of dye pollution in wastewater has reached an alarming level that necessitates urgent action.

3. Various Techniques and Technologies in the Removal of Dyes in Wastewater

Various techniques and technologies have already been used to remove dyes in wastewater. Included are advanced oxidation process [13], ion exchange [14], coagulation/flocculation [15], chemical precipitation [27], membrane technologies [28], electrochemical techniques [29], and adsorption [4, 9, 17]. These technologies have their own advantages and disadvantages, and these were summarized and are presented in Table 1 and Figure 2.

Treatment Technology	Advantages	Disadvantages	Reference
Advanced Oxidation Process	• In situ production of reactive radicals • Little or no consumption of chemicals • Mineralization of the pollutants • No production of sludge · Rapid degradation • Efficient for recalcitrant molecules (dyes, drugs, etc.) • Very good abatement of chemical oxygen demand and total oxygen demand	• Laboratory scale • Economically non-viable for small and medium industries • Technical constraints • Formation of by-products • Low throughput	$[30]$
Ion Exchange	• Wide range of commercial products available • Technologically simple • Procedures are already established. • High regeneration • No loss of sorbents during regeneration	• Economic constraints • Large volume requires large column. • Not effective for disperse dyes	[30, 31]
Chemical Precipitation	• Technologically simple • Simple process • Economically efficient • Adapted to high pollutant loads	• Chemical consumption • Sludge generation • Additional cost for sludge disposal • Ineffective in removal of the metal ions at low concentration	[30, 31]
Coagulation/Flocculation	• Process simplicity • Low capital cost • Very efficient for SS and colloidal particles • Good sludge settling	• Requires adjunction of non-reusable chemicals • Increased sludge volume generation; problem in handling and disposal • Low removal of arsenic	[30, 31]
Membrane Technologies	• Removal of all dye types • Resistance to temperature and chemical environments	• Investment costs are often too high for small and medium industries. • Limited lifetime before membrane fouling occurs. • High energy requirement	[30, 31]

Table 1. Advantages and disadvantages of the different wastewater treatment technologies

Treatment Technology	Advantages	Disadvantages	Reference
Electrochemical Techniques	• The system is robust, efficient, and can be easily controlled. • Efficient elimination of SS, oils, greases, color, and metals • Increases biodegradability • More effective and rapid organic matter separation than in traditional coagulation	• Periodic maintenance \bullet Cost	[30, 31]
Adsorption	• Dyes have a high adsorption capacity. • Technologically simple • Convenient, ease of operation, easy designs, and cost-efficiency	• Non-destructive processes • Cost of regeneration • Generation of sludge	[30, 31]

Table 1. Advantages and disadvantages of the different wastewater treatment technologies (continued)

Figure 2. Schematic representation of dye removal technologies

4. Adsorption as a Wastewater Treatment Process

Adsorption is one of the unique mass transfer mechanisms in which a fluid, gas, or liquid component(s) adheres to a solid surface without the solid atoms being intimately mixed [32]. This popular separation technique has been used for adsorbing water pollutants in numerous applications, including wastewater treatment [33]. Adsorption is a physicochemical reaction with the adsorbate on the surface of the adsorbent. It can be categorized as physical adsorption or physisorption and chemical adsorption or chemisorption, depending on the interactions between the adsorbate and adsorbent molecules [34]. The intermolecular force occurring between adsorbates and adsorbents induces physisorption. This is known as the van der Waals adsorption, and the force is called the van der Waals force. Physical adsorption can occur on any solid surface since van der Waals force occurs between any two molecules. The binding force is weak with less adsorption heat since intermolecular forces cause physisorption and rapid adsorption and desorption rates. Chemisorption involves the transfer, exchange, or distribution of electrons between adsorbates and adsorbents (atoms or molecules), and the adsorption of adsorbents is caused by the formation of chemical bonds between adsorbents and specific adsorbate groups [35]. Over the years, several wastewater treatment technologies have been studied; however, adsorption has become more popular and commonly applied [36]. It is a widely used method for removing dye in wastewater because it is relatively cheap, stable, environmentally safe, and easy to use. [37]. Activated carbon (AC) is the most common adsorbent in adsorption processes. However, while AC is efficient in treating wastewater, preparing, and regenerating activated carbons is relatively expensive, limiting their use [38]. This led to exploration of less expensive alternative sources of activated carbon that could have the same efficiency in eliminating contaminants such as dyes in wastewater. Researchers have recently investigated using agricultural wastes as a source of activated carbon adsorbents.

Figure 3 presents the general schematic flow of the adsorption of dye-contaminated wastewater using agricultural-based biomass as adsorbent. In the Figure, it can be noted that several methods are being used in the utilization of agricultural biomass as adsorbent, including conversion of the biomass to activated carbon [39, 40], chemical activation and modification of the biomass [41-43], hybridization [44, 45], impregnation [46], and others.

Figure 3. Overview of adsorption of dyes in contaminated water using agricultural-based biomass

5. Agricultural Wastes and Residues as Adsorbents in Dye-Contaminated Wastewater Treatment

In searching for less expensive activated carbon adsorbents, researchers eyed the potential of agricultural wastes as raw materials in developing adsorbents. Agricultural waste was generated in high amounts, especially in food industries that have been associated with the increasing demand for food because of the growing population. Thus, because of the high volume readily available, these wastes were exploited in adsorbent preparation experiments [47]. In recent years, for four key reasons, many researchers have used agricultural waste as a precursor when developing activated carbon because it is sustainable, cheap, readily accessible, and environmentally friendly [24].

Numerous researchers have proven the potential of agricultural waste as an adsorbent in removing dyes in wastewater. For instance, Pagalan *et al*. [4] and Liang *et al*. [48] reported the potential of coffee grounds as an adsorbent in removing aniline yellow and congo red dyes, respectively, from contaminated water samples, with a remarkable removal efficiency, as reported in Table 2.

Table 2. Agricultural waste biomass adsorbents and their efficiency in the removal of dyes in dyecontaminated wastewater

Table 2. Agricultural waste biomass adsorbents and their efficiency in the removal of dyes in dyecontaminated wastewater (continued)

Biomass	Dye	Experimental Conditions	Removal Efficiency/ Adsorption Capacity	Reference
Wheat Bran	Crystal Violet	Adsorbate Concentration: 100 mg/L Adsorbent Dose: 10 mg/L Contact Time: 180 min pH: 8	88%	[59]
Lime Peel	Crystal Violet	Different quantities $(1, 3, 5, 7,$ and 10 g) were tested to 50 mL of CV solution $(1,000 \text{ mg/L})$ at 3-h interval incubation.	98%	[60]
Pineapple Leaves	Crystal Violet	Different quantities $(1, 3, 5, 7,$ and 10 g) were tested to 50 mL of CV solution $(1,000 \text{ mg/L})$ at 3-h interval incubation.	97%	[60]

Table 2. Agricultural waste biomass adsorbents and their efficiency in the removal of dyes in dyecontaminated wastewater (continued)

Moreover, several types of agricultural biomasses have been reported to have the potential to remove crystal violet dye in contaminated wastewater. These include coconut flower sheath [52] with a removal efficiency of 94%, rice husk [53] with 85.89-96.16% efficiency, *Zea mays* L. (sweet corn) cobs [58] with an adsorption capacity of 700 mg/g, wheat bran [59] with the removal of 88%, peanut hull $[61]$ with adsorption capacity of 100.6 mg/g, lime peel $[60]$ with 98% removal efficiency, and pineapple leaves [60] with 97% removal. These values are high enough to support the idea that agricultural waste biomass can be an alternative adsorbent in treating wastewater dyes.

Preparations and pre-treatments of these agricultural biomasses vary from one to the other and can affect adsorption capacity. For instance, Rondina *et al*. [18] prepared a novel activated carbon from sugarcane press mud by activating the biomass using 2% (v/v) HNO₃ before it was carbonized. This novel press mud activated carbon resulted in a 98.68% removal of methyl orange dye from wastewater. Meanwhile, Homagai *et al*. [53] explored the modification of rice husk to remove crystal violet dye from aqueous solutions. In their paper, they used charred rice husk and a modified charred rice husk by introducing the xanthate group to make xanthated rice husk. With this modification, the researchers were able to increase the adsorption capacity of the charred rice husk for crystal violet dye in aqueous solution from 62.85 mg/g to 90.02 mg/g at pH 10.

Adsorption isotherm and kinetics are also important aspects of the adsorption studies that must be considered. The adsorption isotherm data is also an essential part of data analysis since it can be used to find the adsorption capacity and the mechanism of adsorption. The principle of monolayer adsorption at homogeneous and energetically active adsorbent sites is based on the Langmuir isotherm [62]. On the other hand, the Freundlich isotherm assumes that the active sites have their energies distributed heterogeneously and exponentially and follow a multilayer adsorption process [63]. Adsorption kinetics is used to measure the adsorption uptake with respect to time at constant pressure or concentration and is employed to measure the diffusion of adsorbate in the pore. Pseudo-first-order and pseudo-second-order kinetics are the two most common kinetic models. The pseudo-first-order kinetic model indicates that the reaction is more inclined towards

physisorption, while pseudo-second-order kinetic suggests an inclination towards chemisorption. Table 3 shows the best-fitted isotherm and kinetic models for some research studies that utilized agricultural biomass as adsorbents for dye removal in wastewater.

Based on Table 3, it can be observed that most of the findings reported that the Langmuir isotherm model and the pseudo-second-order kinetic model are the best-fit models that describe the adsorption mechanism of dyes using agricultural-based adsorbents. This implies that adsorption occurs at well-defined localized sites in agricultural-based biomass adsorbent active sites, meaning that saturation coverage equates to full utilization of these localized sites in most active sites. Furthermore, it can be interpreted that in most cases, chemisorption takes place by forming a single layer of adsorbate attached to the adsorbent surface by chemical bonds.

6. Recent Advancements in the Use of Algal, Fungal, and Bacterial Biomass as Adsorbents in Dye-Contaminated Wastewater Treatment

Recent advancements in the adsorption process have evolved from the use of plant-based biomass to the use of some other biological biomass such as algae, fungi, and bacteria. This is because using biological methods in wastewater treatment is considered the most promising because the treatment is environmentally safe [30]. Among the widely researched bio-sorbents with proven efficiency in removing dyes in wastewater are the algal, fungal, and bacterial biomass types.

The wide availability and the efficiency of algal biomass in the biosorption of wastewater dyes make it one of the most favorable bio-sorbent sources [30]. There have been several reports already published on the internet that proved the efficiency of the algae bio-sorbents in dyecontaminated wastewater treatment. The composition and the structure of the algal cell wall and the proteins present in algae are essential components in the biosorption process as these components provide acid-binding sites such as amino, amine, hydroxyl, imidazole, phosphate, and sulfate groups [30, 64]. The most common algal species that have been reported to have a high removal efficiency rate of up to 99% are the *Spirulina* and *Chlorella* species. Meanwhile, the most common dye that has been removed by the microalgae and algae bio-sorbents is methylene blue dye. These findings were reported by Aragaw and Bogale [30] in their article on biomass-based adsorbents for removing dyes from wastewater. The same observation was reported by Mishra *et al*. [65]. The list of algal biomasses with reported high efficiency rates in the removal of dyes ainclude *Sargassum muticum, Sargassum horneri, Spirulina platensis, Scenedesmus* sp*., Nizamuddinia zanardinii, Sargassum hemiphyllum, Dictyota cervicornis, Durvillaea antarctica, Ulothrix zonata, Chlorella pyrenoidosa Chlorella vulgaris, Chlamydomonas variabilis,* and *Ulva fasciata* [65-75]. Overall, it can be noted and described that the pH of the solution is a great contributory factor to the interaction of the dyes and the bio-sorbents, and thus to removal efficiency. According to Aragaw and Bogale [30], cationic dyes are generally removed in basic conditions. In contrast, anionic dyes are removed in acidic conditions due to the involvement of the H+ ions in the interaction process between dyes and the bio-sorbents. Accordingly, adsorption tends to decrease with the hydrogen ions (H+) competing with the cations present in the dye solution when the adsorbent possesses a positive charge on its surface.

Fungi are also among the most commonly studied biological adsorbents, as they possess significant components that are essential in dye-contaminated wastewater adsorption process. Generally, the most reported compounds that compose the fungal cell walls are carbohydrates such as sugars, D-glucose, D-galactose, glucan and chitin, and other compounds such as proteins, lipids, and in some cases, phosphorus and uronic acid are present [76]. These compounds have been widely observed in several fungal biomass species such as *Aspergillus* sp., *Penicillium* sp., *Trichoderma* sp., *Rhizopus* sp., and *Saccharomyces* sp. For instance, it was previously reported that *Trichoderma* biomass efficiently removed Congo red (CR) and malachite green (MG) dyes in wastewater solution. As the researchers noted, physicochemical factors such as the concentration and size of the dye molecule, ionic charge, pH, and temperature, significantly affect the dye-removal efficiency of fungal biomass [77]. Furthermore, it is worth to be noted that in the study of Chen *et al*. [78], *Penicillium simplicissimum* had a better efficiency when the cells were living rather than dead. The live cells of *Penicillium simplicissimum* treated crystal violet (CV), methyl violet (MV), malachite green (MG) and cotton blue (CB) (75.6-90.8%, 0.5-2h) more efficiently than dead cells (43.9- 75.2%, 4 h).

Meanwhile, with a maximum capacity of dye adsorption obtained of 58.48 mg g^{-1} , *Sarocladium* sp. dried biomass was found to be a novel, low-cost and efficient adsorbent for reducing water and environment dye contamination (e.g., Remazol Black dye) [79]. Generally, reports showed that fungal biomass is a promising substitute for the current dye removal technologies. Aside from parameter optimizations, the genotype and the biomass preparation are essential factors to consider for effective dye adsorption performance [30].

Lastly, bacterial biomass has become one of the recently explored and promising biological adsorbents. The small size of bacteria and their omnipresence and ability to grow under erratic environmental conditions make them good adsorbents [30, 64]. Among the bacterial biomasses that were reported to have high removal efficiency was the consortium of *Neisseria* sp., *Vibrio* sp., *Bacillus* sp., and *Aeromonas* sp., with consortium efficiency of 65-90% in treating reactive dyes such as Novacron Orange FN-R, Novacron Brilliant Blue FN- R, Novacron Super Black G, Bezema Yellow S8-G and Bezema Red S2-B [80]. It can also be noted that in the study of Velayutham *et al*. [81], 100% removal of Remazol Brilliant Blue R (RBBR) dye was recorded at 100 mg/L, 37°C, 12 h, and static conditions, using *Staphylococcus* sp. K2204 bacterial strain. A methyl orange dye removal of 100% was also recorded in the study of Akansha *et al*. [82] at pH 7, 35 °C, 12 h, 150 mg/L, static conditions using *Bacillus stratosphericus* SCA1007 bacterial strain. Several more bacterial strains effectively removed various dyes in wastewater such as *Alcaligenes faecalis AZ26, Bacillus cereus AZ27 and Bacillus* sp*, Pseudomonas aeruginosa (RS1),* and *Thiosphaera pantotropha ATCC 35512, Bacillus megaterium KY848339.1,* and *M. yunnaenensis*, among many others [83].

7. Future Challenges in the Utilization of Agricultural-Based Adsorbents

Numerous works have been mentioned in this paper about the potential use of agricultural biomass as an adsorbent in removing dyes in wastewater. However, there are challenges worth to be noted for future works of this field.

7.1 Upscaling of the use of agricultural-based adsorbents

Although studies have already proved the efficiency of agricultural-based adsorbents in removing dyes in wastewater, most of these studies are conducted at a laboratory scale. One future challenge will be to upscale the use of these adsorbents in large-scale and industrial setups.

7.2 Exploration of the application of these adsorbents in removing mixed dye wastes

In an actual scenario, industries have a combination of dyes in their wastewater treatment facility. As observed, many studies were conducted to test the adsorbent's efficiency in removing one dye contaminant in an aqueous solution alone. Thus, it is a good research area in the future to explore whether the adsorption capacity of the adsorbent is still the same when the wastewater sample contains more than one dye contaminant.

7.3 Waste management of the used adsorbents

The used adsorbents must be managed appropriately, especially if the application becomes largescale. One of the main reasons for using agricultural biomass as an adsorbent is to establish an environmental-friendly treatment. However, improper waste management and handling of the used adsorbents defeat this purpose.

8. Conclusions

Agricultural-based and biological adsorbents are indeed promising alternatives in the treatment of dye-contaminated wastewater, as reported in various published research work. This can be attributed to the claim that these agricultural-based adsorbents are sustainable, cheap, readily accessible, and environmentally friendly. Various treatment technologies are already used in the treatment of dyecontaminated wastewater; however, each of these technologies has its own advantages and disadvantages. In this paper, we tried to reveal the advantages of using agricultural biomass as adsorbents owing to the high removal efficiency and adsorption capacity of various agricultural wastes as reported in various literature. The use of agricultural wastes as a source of activated carbon and bio-sorbents has become a promising trend with recorded high removal efficiency; however, depending on the characteristics of the adsorbent, the removal is greatly affected. The same is also observed in a lot of research work concerned with the use of biological adsorbents such as algae, fungi, and bacteria. Overall, for algae biomass, it can be noted and described that the solution's pH is a great contributor to the removal efficiency as it affects the interaction of the dyes and the biosorbents. Meanwhile, fungal biomass is a promising substitute for the current dye removal technologies. Aside from parameter optimizations, genotype and biomass preparation are essential factors to consider for effective dye adsorption performance. However, some challenges and concerns for future work also need to be considered. This includes the challenge of upscaling agricultural-based adsorbents from laboratory to plant scale. Hence, future work should include the integration of these adsorbents into treatment plants. Another challenge is to explore the application of these adsorbents in removing mixed dye wastes in industry wastewater. Lastly, it is important that waste management strategies that are to be implemented should facilitate the development of agricultural biomass as adsorbents that are sustainable, cost-effective, accessible, and environmentally friendly.

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