

Removal of Azo and Anthraquinone Dye by Plant Biomass as Adsorbent – A Review

Peck Kah Yeow¹, Sie Wei Wong¹, Tony Hadibarata^{1,*} 

¹ Department of Environmental Engineering, Faculty of Engineering and Science, Curtin University Malaysia, CDT 250, Miri, Malaysia

* Correspondence: hadibarata@curtin.edu.my;

Scopus Author ID 16233109100

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Abstract: Acting as a key element for the survival of human and nature, clean water also contributes tremendously to the ever-growing industries in a country. However, the supply of clean water had led to a decrease as pollutants such as dyes had caused a major negative impact on pure and clean main water bodies. In recent years, textile industries have developed and contributed to more than 50% of dye wastewaters in the world. The improper method of discharging dye effluent to the aquatic environment caused the destruction of habitat and degradation of water quality. Advanced treatments such as photocatalysis, electrooxidation, the Fenton process, and biological treatment via bacterium are often used for dye wastewaters. However, these treatment processes are often expensive in operation and maintenance. In conjunction, adsorption is one of the efficient, cost-effective, and environmentally friendly treatment methods. The adsorbent most widely used is the activated carbon adsorbent. Activated carbon comes in two forms, granular activated carbon (GAC) and powdered activated carbon (PAC). There are two methods to activation of carbon that are physical activation and chemical activation. The factors affecting the efficiency of adsorption are the adsorbent dosage, dye concentration, pH value, and temperature. In this article, the efficiency of dye wastewater treatment via adsorption is discussed. Several waste materials are being studied especially agricultural biomass as it has little or no economic value and often poses disposal issues. Some low-cost agricultural biomass-based adsorbents such as tea waste, hazelnut husk, bacteria aggregate, rice ash, pineapple leaf, and fruit waste have been tested to be effective in the dye removal process. There are mainly four categories of agricultural biomass-derived adsorbent, such as leaf-based, peel-based, stem-based, and seed-based adsorbents.

Keywords: Adsorption; Activated carbon; Dye wastewaters; Agricultural waste.

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1. Introduction

Life on Earth is highly dependent on clean water as it serves as one of the requirements needed to conduct activities that have huge implications for the daily life chain. To ensure water to have of any beneficial value for human usage, it is required that a good quality of water meets the favorable level of physical and chemical requirements that is comparable to national water quality guidelines. Water pollution is the change of any beneficial use of water due to the existence of any biological, physical, or chemical factors that could lead to a hazardous impact on safety and public health. Dyes have become one of the major pollutants to the environment due to the high amount of discharge of wastewater containing high concentrations of dye. There are various types of dye; however, the most commonly used dye

is the azo dyes. In textile industries, azo dyes are highly used and have accounted for a minimum of 50% of the synthetic dyes used in the world. Azo dyes are normally in red, orange, and yellow colors. They are also toxic and can cause recalcitrance and salination of water resources. The high frequency of usage of azo dye also caused the abundance of wastewater in the environment. Hence, it is important to treat dye wastewater before discharging into the environment [1, 2]. The treatment of dye wastewater can be classified into three categories, which are physical, chemical, or biological methods. However, studies found that some physical and chemical treatments do not mineralize dyes completely, which may cause secondary pollution when discharged into the environment. On the other hand, biological treatments are often recommended in treating dye wastewaters. Biological treatment is coming into a trend as it is cost-effective, eco-friendlier, and produces less sludge compared to physical and chemical treatments. The only downside of biological treatment is the inefficiency of decolorization during treatment. The efficiency of biological treatment often depends on the microorganism used in treatment. There are several methods to classify dyes, and one of them is, according to the manufacturing source of being natural or synthesized. Natural dyes are derived from natural resources, which can be categorized into the plant, animal, mineral, microbial and fungal origins. The majority of natural dyes are extracted from plants, and the parts utilized include leaves, twigs, flowers, bark, roots, fruits, and stems. In 1856, the first synthetic dye named mauve was accidentally created by William Perkin while searching for a cure for malaria. Ever since, more chemical dyes have been discovered and extensively used in industries as synthetic dyes are less expensive and have better color fastness compared to natural dyes. Nevertheless, harmful chemicals are often found in synthetic dyes such as lead, copper, benzene, mercury, and toluene. As shown in Table 1, synthetic dyes can be classified by their types of surfaces to which they can be applied, method of application, and chemical composition [3]. Dyes are also categorized based on their chemical structures, which consist of a group of atoms known as chromophores. Chromophores are organic molecules that absorb light at specific frequencies formed by atoms and electrons, which cause dyes to be colored [4]. There are several chromophores for dyes that are present in the form of azo, anthraquinone, triarylmethane, heterocyclic, indigoid, nitroso, nitro, and phthalein. It is reported that azo dyes are the largest class of dyes being applied in the industries because of the simplicity of the pairing reaction and the extensive possibilities for structural variations to adapt to the needs of complex applications, as shown in Table 2.


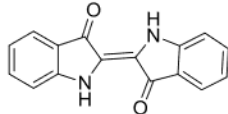

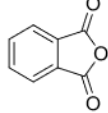
Table 1. Types of dyes used on suitable surfaces.

Class	Application	Example of dye	
Acid	Silk, wool, leather, nylon, modified acrylic fibers, ink, paper	Acid Yellow 23 Acid Yellow 36 Acid Red 52 Acid Red 88 Acid Black 1 Acid Blue 25 Acid Blue 45	Congo Red Allura Red Erythrosine Tartrazine Sunset Yellow Acid Orange 3 Acid Orange 19
Basic/ Cationic	Leather, paper, wool, straw, acrylics, silk, linen, hemp, cotton, rayon, jute, modacrylic fiber, coir	Basic Brown 1 Basic Violet 10 Basic Violet 14 Basic Blue 9 Basic Blue 54 Basic Violet 3 Basic Green 4	Basic Yellow 2 Basic Yellow 82 Methylene Blue Toluidine Blue Thionine Crystal Violet Basic Fuchsin Safranin
Direct	Cotton, paper, leather, wool, silk, nylon, linen, rayon, viscose	Martius Yellow Direct Yellow 50 Direct Green 28 Direct Orange 26	Direct Red 23 Direct Red 81 Direct Blue 67 Direct Black 19

Class	Application	Example of dye	
		Direct Orange 28 Direct Violet 51	Direct Black 22
Disperse	Nylon, orlon, polyester, cellulose acetate, triacetate fibers, diacetate fibers, acrylic, plastics	C.I. Basic Blue 3 C.I. Basic Green 4 Disperse Yellow 3 Disperse Yellow 49 Disperse Yellow 126 Disperse Red 4 Disperse Red 54	Disperse Red 134 Disperse Red 202 Disperse Red 210 Disperse Blue 27 Disperse Blue 73 Disperse Blue 296
Reactive	Cotton, wool, silk, viscose, nylon, polyamide fiber	Reactive Orange 1 Reactive Orange 86 Reactive Red 5 Reactive Red 2 Reactive Red 11	Reactive Red 6 Reactive Yellow 22 Reactive Blue 1 Reactive Blue 5 Reactive Blue 161
Vat	Cotton, fibers, linen, rayon, wool, silk, nylon, polyester	Indigo Vat Red 10 Vat Blue 4 Vat Blue 36 Vat Blue 64 Vat Brown 3 Vat Brown 45 Vat Violet 13 Vat Violet 18	Vat Orange 1 Vat Orange 3 Vat Orange 17 Vat Green 1 Vat Green 11 Vat Green 12 Vat Black 25 Vat Black 27 Vat Yellow 28
Sulfur	Cotton, viscose, rayon	Sulfur Red 7 Sulfur Black 1 Sulfur Yellow GC Sulfur Brilliant Green Sulfur Red Brown B3R Sulfur Yellow Brown 6G Thiazoles Thianthrene	Sulphur Red 10 Sulphur Blue 10 Sulphur Black 2 Sulphur Orange 1 Sulphur Green 14 Sulphur Yellow 9 Sulphur Brown 10
Mordant	Wool, nylon, fiber, cotton, silk, leather, anodized aluminum	Mordant Red 5 Mordant Red 11 Mordant Black 11 Mordant Orange 1	Turkey Red Acid Orange 10 Acid Mordant Orange 14
Solvent	Synthetics, plastics, gasoline, oil, wax, vanish, lacquer, ink, stain	Solvent Red 24 Solvent Red 43 Solvent Red 164 Solvent Blue 35 Solvent Blue 67 Solvent Green 28 Solvent Orange 2	Solvent Orange 45 Solvent Yellow 32 Solvent Yellow 124 Solvent Violet 31 Solvent Violet 38 Solvent Black 27 Solvent Brown 43

Table 2. Classification and example of dyes according to the chemical structure.

Class	Structure/ Chromophore	Example of dye	
Azo		Procion Yellow H-EXL Procion Navy H-EXL Sulphonyl Blue TLE Sulphonyl Green BLE Maxilon Blue Entrazol Blue IBC BNLE	Acid Red 337 Congo red Reactive Orange 16 Reactive Black 5 Remazol Black B Remazol Black N Sulphonyl Scarlet
Anthraquinone		Alizarin Red S Uniblue Reactive Blue 4 Reactive Blue 19 Remazol Brilliant Blue R Pigment Yellow 24	Vat Yellow 28 Vat Green 1 Acid Blue 129 Acid Green 25 Disperse Red 3B Disperse Blue 3
Triarylmethane		Acid Blue 90 Acid Blue 119 Acid Blue 48 Patent Blue V Brilliant Blue FCF Basic Green 4 Malachite Green	Basic Red 9 Basic Violet 14 Basic Violet 3 Basic Violet 1 Basic Violet 2 Green S

Class	Structure/ Chromophore	Example of dye	
Heterocyclic		Disperse Red 356 Fluorol Yellow 88 Solvent Green 4 Basic Violet 10	
Indigoid		Acid Blue 71 CI. Vat Blue 35 CI. Basic Violet 14	Tyrian Purple Thioindigo Indigo Carmine
Nitroso	—N=O	Martins Yellow Palatine Orange Fast Green O	Pigment Green B CI. Pigment Green 8
Nitro		Disperse Yellow 14 CI. Acid Yellow 1 CI. Pigment Yellow 11	Martius Yellow Naphthol Yellow
Phthalein		Phenolphthalein	

2. Toxicity effects of dyes

Wastewater from dyeing industries is unarguably a huge pollutant around the world greatly because of the low ability of degradation and release of harmful substances as the dyes decompose. The release of dyeing effluents poses direct effects on water transparency, aesthetic merit as well as dissolved oxygen content. Furthermore, the water bodies contaminated by dyes usually contain high chemical oxygen demand (COD), biochemical oxygen demand (BOD), total dissolved solids (TDS), inorganic salts, pH, turbidity, salinity and temperature [5]. These conditions are related to the incomplete degradation of dyes as the color from the dye wastewater impedes light penetration into the water bodies. The growth of biota was eventually interrupted as the photosynthetic activity is slowed down due to sunlight insufficiency. In addition, the foul smell was released to the atmosphere when the aquatic flora and fauna beneath the water surface are decomposing [6]. The discharge of dyeing effluents into the rivers induces soil infertility as the dye components are responsible for stabilizing and retaining the soils. As mentioned by Ajmal and Khan [7], a higher concentration of dye effluents used for irrigation has distinctly increased the delay and inhibition rates of seed germination as well as the overall growth of plants by observing the length of roots, shoots, dry weight of plants and the conditions of leaves. Even though new methods of reducing negative impacts on soil microbial pollutions such as organic amendments to soils were discovered, the effectiveness and post effects still remain unrecognized [8]. Thus, it is suggested to treat the contaminated wastewater before being released into the water bodies. Besides that, high content of chemicals, especially heavy metals such as cadmium, chromium, copper, lead, and nickel is often found in dyeing effluents. These heavy metals will bioaccumulate in the aquatic life and trapped in the soil, which was then be deposited into the human body through consumption [9]. Prolonged exposure to dye chemicals may contribute to the disruption of varied processing operations, which can cause serious health problems such as cancer, emphysema, heart diseases, kidney failure, and negative effects on metabolism. Based on a research done in 91 locations, more than 70 percent of the fish were detected with highly toxic dyes in their muscle tissues and has appeared to be an additional threat to some endangered

species [10]. The occurrence of dyes not only poses health hazards to humans and aquatic plants but an environmental impact on the ecological balance.

3. Current trend of dye wastewater treatment

Textile industries involve the use of dye to colorize materials such as fabric; they are also the largest contributing factor to dye wastewaters. At the present time, the removal of dye particles from water sources has become a significant environmental concern, as well as a challenge such as dyes are extremely difficult to treat. This is due to the nature of dyes being stable molecules, produced primarily to resist degradation of light, various biological and chemical reactions. In addition, most dyes do not degrade naturally and require proper treatment prior to discharge. The treatment of dyeing effluent plays a vital role in ensuring safe water quality where the water will be recycled to the environment, then for industrial and domestic uses. Hence, effective and thorough technologies for textile wastewater treatment are essential to reduce environmental and health threats. A wide variety of dye removal techniques have been discovered in recent years and claimed to be successful dye remediations. Nevertheless, most of them suffer from drawbacks such as the ineffective removal against certain dyes or high cost of manufacturing [11]. The optimal method to eliminate dye from wastewater should be able to effectively extract large amounts of dye over a short time, without creating secondary pollution [12]. The classifications of dye removal technologies are divided into three groups, which are biological, chemical, and physical (Table 3).

Table 3. Advantages and disadvantages of biological, chemical, and physical methods for synthetic dye removal [17, 44, 45].

Treatment	Advantages	Disadvantages
Biological methods		
Activated sludge	<ul style="list-style-type: none"> - Low cost of installation - Low requirement of land area 	<ul style="list-style-type: none"> - Generation of sludge - Rigid process method
Fungal decolorization	<ul style="list-style-type: none"> - Capable of degrading dyes using enzymes - Flexible method 	<ul style="list-style-type: none"> - Unreliable enzyme production - Lengthy growth phase
Microbial cultures (Mixed bacterial)	<ul style="list-style-type: none"> - Decolourization occurs in 23-30 hr which is considered fast 	<ul style="list-style-type: none"> - Cationic dyes are not ready for metabolism under aerobic conditions
Adsorption (Microbial biomass)	<ul style="list-style-type: none"> - Some dyes bind to microbial species with have high affinity 	<ul style="list-style-type: none"> - Effective for a limited type of dyes
Aerobic-anaerobic remediation	<ul style="list-style-type: none"> - Allow decolorization of azo and water-soluble dyes - No foam formation 	<ul style="list-style-type: none"> - Formation of hydrogen sulfide and methane - Require a large land area - Generation of sludge - Time-consuming
Algae degradation	<ul style="list-style-type: none"> - Easily assessable - Low operational cost 	<ul style="list-style-type: none"> - Unstable system
Enzyme degradation	<ul style="list-style-type: none"> - High efficiency - Non-toxic - Reusable 	<ul style="list-style-type: none"> - Unreliable enzyme production
Chemical methods		
Electrokinetic coagulation	<ul style="list-style-type: none"> - Economically expedient 	<ul style="list-style-type: none"> - High generation of sludge
Electrochemical destruction	<ul style="list-style-type: none"> - Compounds broken down are non-toxic - No consumption of chemicals - No sludge generation 	<ul style="list-style-type: none"> - High electricity consumption - High flow rates cause a reduction in dye removal
Fenton's oxidation	<ul style="list-style-type: none"> - Effective decolorization of soluble and insoluble dyes - Removal of all toxins 	<ul style="list-style-type: none"> - Generation of sludge - Narrow working pH range - High cost and risk with handling reagents
Ozonation	<ul style="list-style-type: none"> - Can be applied in a gaseous state - No increase in sludge and wastewater volume 	<ul style="list-style-type: none"> - Short half-life (20 mins) - Generation of toxic by-products - Unstable system

Treatment	Advantages	Disadvantages
Biological methods		
	- Fast reaction	- High cost
Ion exchange	- No loss of adsorbent during regeneration	- Effective for a limited type of dyes
Photochemical	- Reduction in foul odors - No sludge generation	- Generation of by-products
Sodium hypochlorite (NaOCl)	- Initiates and accelerates azo-bond cleavage	- Discharge of aromatic amines
Ultraviolet irradiation	- Require hazardous chemical - No sludge generation - Reduction in foul odors	- High cost - High energy consumption - Limited treatment times
Physical methods		
Adsorption (Activated carbon)	Effective removal of wide ranges of dyes	- Very expensive - 15% loss of sorbent upon reactivation
Adsorption (Peat)	Effective removal of wide ranges of dyes	- Low surface area of adsorption
Adsorption (Wood chips)	Effective removal of acid dyes	- Require long retention time
Adsorption (Silica gel)	Effective removal of basic dyes	- Side reactions (air binding & fouling) prevent a commercial application
Membrane filtration	Effective for water recovery	- High capital cost - Fouling of membrane - Generation of concentrated sludge
Nano-filtration & Ultra-filtration	Effective removal of every type of dye	- High electricity consumption - Require high pressure - Constant blockage of membrane pores - Short life span
Reverse osmosis	Effective desalination and decolorization	- High electricity consumption - Require high pressure
Coagulation & Flocculation	- Low cost Effective removal of sulfur, vat and disperse dyes	- Large generation of concentrated sludge - pH-dependent system

Biological treatment is often known as the most affordable green technique that eliminates organic dyes from textile wastewater with an optimum operating period, compared to chemical and physical remediations [13]. Examples of biologically based methods are activated sludge process, fungal decolorization, microbial cultures, adsorption, aerobic-anaerobic remediation, algae, and enzyme degradation. Microorganisms such as algae, fungi, bacteria, and yeast are usually applied as biological materials to disintegrate, absorb, and accumulate dyes. It is notable that activated sludge treatment is capable of dealing with BOD and COD reduction [14] as well as removing more than 90% of dyes from the wastewater [15]. Dyes are designed to resist light-induced oxidative fading and chemical fading, such that they have fused aromatic ring, high water solubility, and molecular weight. These are the factors that cause dyes to be resilient to microbial degradation and restricted from permeation through biological cell membranes. Furthermore, some commercial dyes are hazardous to certain microorganisms, which will induce a distinct reduction in the biological degradation of dyes and result in sludge bulking. Generally, biological methods are not suitable to be applied to most textile effluents as they are unable to degrade complicated dyes, and the processes are time-consuming [16].

Chemical remediations are methods that apply chemistry and their theories to eliminate dyes. Chemical-based methods used to treat industrial dye effluents consist of electrokinetic coagulation, electrochemical destruction, Fenton's oxidation, ozonation, photochemical, and ion exchange. In comparison with biological and physical treatments for dye removal, chemical methods are more expensive, with an exception to electrochemical degradation [17]. For the

treatment of wastewaters that are toxic to living biomass or immune to biological remediation, Fenton's reagent is a suitable chemical to be utilized [18]. Despite that, García-Montaña, Ruiz, Muñoz, Domènech, García-Hortal, Torrades, and Peral [19] mentioned that Fenton's oxidation is the least environmental-friendly dye removal process as it produces significant iron sludge through the flocculation of the reagent and dye particles. This is for the reason that cationic dyes are unable to coagulate at all, which will result in a poor quality of floc to be disposed of [11].

4. Adsorption

Adsorption through activated carbon has become a popular equilibrium separation technique due to its ability to produce high-quality treated effluent. Although activated carbon is an excellent adsorbent, it is also expensive, and hence, cheaper adsorbent alternatives such as peat, wood chips, and silica gel are developed. The basic types of membrane filtration processes in ascending order of allowable particle size are reverse osmosis, nano-filtration, ultrafiltration, and microfiltration. The filtration mechanisms of these operations are similar but can be differentiated by the relative size of particles being filtered and the pressure required to remove charged ion across the semi-permeable membranes [20]. This removal technique benefits from its resistance to temperature, microbial attack as well as the chemical environment. Nevertheless, membrane filtration requires a high capital cost on membrane replacement as the performance of a membrane drops over time and has a high possibility of clogging [11]. Besides that, the generation of thick residue poses disposal issues. Adsorption is the deposition of molecules at the interface of two phases, which can be liquid-solid, liquid-gas, liquid-liquid, or gas-solid. For instance, the occurrence of adsorption in a solid-liquid system is capable of removing solutes from the solution and accumulating them at the solid surface [21]. Adsorption stands out as one of the most successful treatments in removing dye wastewaters. In recent years, not only the treatment of textile dye wastewaters has been using adsorption for treatment, large industrial companies that purify water from wastewater to drinking water uses the same technique as well. Due to the inability to remove dye from wastewaters through conventional methods, adsorption has become the most reliable treatment method. Initially, adsorption was not highly recommended due to the adsorbents being too expensive. However, researches and development these years have led to the discovery of low cost and high-efficiency adsorbents. Thus, it has become one of the most economical treatment methods in removing dye wastewaters [22]. There are many types of materials that can be used as adsorbents to treat dye wastewater, but the most general type is activated carbon.

4.1. Types of activated carbon.

Adsorption is the process of capturing pollutants known as adsorbate by attaching adsorbates on the adsorbent. Adsorbents are materials that have many insoluble sponge-like pores. A good adsorbent has characteristics such as high adsorption capacity, short adsorption period, size of adsorbent, and the pollutants that can be removed by the adsorbent [23]. The adsorbent capacity is based on the porosity of an adsorbent, and an increase in porous holes increased the total surface area in contact with adsorbates. The adsorption time required to treat wastewater should be shorter as the longer the time required, the lower the efficiency of the adsorbent. In addition, adsorbents that can remove various pollutants can help reduce the type of adsorbent required to treat different types of dye wastewater. However, the capacity is

dependent on the type of carbonaceous source used. The lowest capacity of activated carbon is 500 mg/g, while the highest capacity can reach up to 2000mg/g. One of the most well-known activated carbon is derived from coal. Many previous studies and researches had proved that coal-derived activated carbon has a high adsorbent capacity and can remove various types of dye from wastewaters. In addition, activated carbon can be beneficial as it can remove other pollutants such as heavy metals and organic and inorganic compounds. There are two sizes of activated carbon, that is microporous and macroporous. Microporous activated carbons are used to treat pollutants that have a molecule size smaller than 1nm. In contrast, macroporous activated carbon is used to treat larger sized molecules than are bigger than 1nm. In addition, there are two types of activated carbon. The two types of activated carbon are granular activated carbon (GAC) and powdered activated carbon (PAC). Generally, PAC is more advantageous as it is cheaper than GAC and has higher efficiency. This is because PAC has finer particles, which increases the total surface area in contact with the adsorbate. However, due to the fine particle size of PAC, it can be hard when it comes to designing equipment that can fully utilize PAC. Also, GAC and macroporous activated carbon are more suitable in treating dye as they have better dye adsorption ability [24-26].

4.2. Activation of carbon.

To obtain activated carbon, raw materials like coal and bamboo is treated under specific conditions. There are two types of activation, that is chemical activation and physical activation. The activation period for chemical activation is short, while the activation period for physical activation takes a long time. The activation of carbon through chemical activation is a one-step action. Raw materials are saturated with carbon activation chemicals such as potassium hydroxide and sodium hydroxide solution. The saturated chemical and raw materials are then simultaneously heated under the flow of nitrogen gas. After a short period of time, the raw materials are taken out and washed with distilled water to remove chemicals, and activated carbon is obtained. The advantage of chemical activation is that only a short period of time is required for the overall process, and the less activated carbon obtained is burnt. Physical activation of carbon requires two steps. The first step is pyrolysis, where raw materials are carbonized. The second step is the carbon activation of raw material with the use of steam or oxidation gases. The advantage of physical activation is the activated carbon obtained is macroporous in size, and they are suitable for dye wastewater treatment [27].

4.3. Advantages and disadvantages of adsorption.

The advantages of removal of dye through adsorption of activated carbon are that the efficiency is high. The success rate of removing dye by adsorption of activated carbon is 99.8%. In addition, adsorption does not require large industrial area or processes which would require high maintenance and operation cost. The operation for adsorption is also easy and does not require high professional skills. During the process of activation of carbon also reduces unwanted waste as the raw materials used sometimes are unwanted waste. However, the disadvantage of adsorption is that activated carbon can become saturated to adsorb the pollutant and to decrease the ability after some time [28].

5. Factors affecting adsorption

The efficiency of adsorption is not only dependent on the type of adsorbent used but also on several other factors. Without proper handling and planning, adsorption can be inefficient as well. The most common factors that are affecting the efficiency of adsorbent are adsorbent dosage, contact time, pH value, dye concentration, and temperature. Any change in one of these factors would disturb the optimum working condition for the adsorbent.

5.1. Adsorbent dosage.

The efficiency of adsorption depends on the number of adsorbents used during the treatment process. An optimum number of adsorbents can be determined by preparing sample tests with a fixed concentration of dye and pH value. With optimum adsorbent dosage, then only can the efficiency be at maximum [29]. The removal rate of dye increases with increasing adsorbent dosage. This can also be explained as the increased mass of adsorbent provides a larger surface area, which enhances the adsorption process. This theory was supported by numerous studies focused on the effect of adsorbent dosage, and the results are demonstrated in Table 4.

Table 4. Effect of solution pH on the adsorption of RBBR and RBV-5R dyes by various biomass.

Dye	Adsorbent	pH range	Removal (%)	Reference
RBBR	Pinecone	2-13	35-96	[46]
	Purging nut (<i>Jatropha curcas</i>)	1-8	40-95	
	Pinang frond (<i>Areca catechu</i>)	3-11	40-80	
	Watermelon peel	3-11	31-71	
	Corn peel	3-11	9-29	
	Pomegranate peel	2-12	40-94	[47]
	Peanut shell	3-9	84-100	[48]
RBV-5R	White rot fungus	3-7	15-100	[49]
	Cocoa pod husk	3-11	92-100	[35]
	Orange peel	2-6	69-90	[50]
	Chicken eggshell	2-11	55-94	[51]
	Calcined eggshell	3-11	96-97	[52]
	Sawdust	1-6	10-85	[53]
	Coconut mesocarp	1-6	8-85	[53]
	Natura rice hull	1-6	15-46	[54]
	Treated rice hull	1-6	60-74	[54]
	Watermelon peel	3-11	39-68	
Corn peel	3-11	14-44		

5.2. pH value.

The pH value controls the movement of the electrostatic charge. The rate of transmission of the dye molecules from the wastewater to the adsorbent varies depending on the degree of electrostatic charge. The optimum pH value for adsorption is near to 6, which is slightly below neutral [29]. A previous study showed that high pH value causes a change at the solution interface, inducing the adsorbent surface to be negatively charged. Thus, the electrostatic repulsion of negatively charged functional groups of reactive dyes from the adsorbent surface will eventually decrease the rate of adsorption [30]. On the contrary, solutions with low pH content enhance the attraction forces between the molecules of reactive dye and adsorbent surface. This phenomenon will then allow the rate of anionic adsorption to

increase [31]. The results of various studies on the effect of pH content with different adsorbents are tabulated in Table 5.

Table 5. Effect of initial dye concentration on the adsorption of RBBR and RBV-5R dyes by various biomass.

Dye	Adsorbent	Concentration range (mg/L)	Removal (%)	Reference
RBBR	Pinecone	200	48-98	[46]
	<i>Jatropha curcas</i> pods	50-250	48-94	[55]
	Watermelon peel	30-50	64-48	
	Corn peel	30-50	11.4-11.5	
	White rot fungus (<i>Dichomitus squalens</i>)	500-3000	94-95	[56]
RBV-5R	Cocoa pod husk	20-100	76-97	[35]
	Chicken eggshell	20-100	72-88	[51]
	Calcined eggshell	20-100	92-97	[52]

5.3. Dye concentration.

A higher concentration of dye would require a much larger adsorbent. The number of binding sites for the dye molecules to attach to the adsorbent would need to be increased in order to fully remove high concentrated dye from the solution [17]. The initial concentration of dye plays a significant role in resolving the mass transfer resistance of the dye in both solid and aqueous states. Mahmoud, Salleh, Karim, Idris and Abidin [32] states that high adsorption capacity can be achieved if the initial concentration of dye is high. This is due to the strong driving force provided to the concentration gradient. Nevertheless, the percentage of color removal will decrease as the initial dye concentration increases as the majority of binding sites of adsorbents are occupied [33]. Table 6 shows the effect of initial dye concentration on dye adsorption by various adsorbents.

Table 6. Effect of temperature on the adsorption of RBBR and RBV-5R dyes by various biomass.

Dye	Adsorbent	Temperature range (°C)	Removal (%)	Reference
RBBR	Pinecone	25-45	61.5-94.6	[46]
	Peanut shell	20-60	52-100	[48]
	White rot fungus	4-60	15-100	[49]
RBV-5R	Chicken eggshell	20-0	90-94	[51]
	Calcined eggshell	20-40	89-97	[52]

5.4. Temperature.

Temperature affects the treatment process by the shifting of nature of reaction from endothermic to exothermic or vice versa. The rate of adsorption drops when the temperature is too high or low. The optimum temperature is tested to be around 30°C [17]. Adsorption reactions are usually indicated as exothermic processes as heat energy is being released to the surroundings [34]. An increase of temperature improves the adsorption capacity by enhancing the rate of diffusion of adsorbate molecules across the external boundary layer caused by the low viscosity of the solution [35]. This may also be a result of the rising number of active sites in the adsorbent surfaces, as well as the enhanced mobility of the dye molecules at higher temperatures [36]. However, the adsorption rate will decrease after some time when the temperature has achieved its equilibrium [37]. Table 7 indicates the results from several studies on the effect of temperature with different adsorbents.

Table 7. Effect of adsorbent dosage on the adsorption of RBBR and RBV-5R dyes by various biomass.

Dye	Adsorbent	Adsorbent dosage (g)	Removal (%)	Reference
RBBR	Pinecone	0.5-2.0	48-98	[46]
	<i>Jatropha curcas</i> pods	0.1-0.5	62-98	[55]
	Tea leaves	5	11	[57]
RBV5R	Orange peel	0.05-2	25-80	[50]
	Chicken eggshell	0.5-2.5	75-95	[51]
	Calcined eggshell	0.5-2.0	90-97	[52]

6. Utilization of agricultural biomass and future development

The challenges for coal-derived activated carbon is a limited resource due to the limited availability of fossil fuel. Industries cannot fully rely on the usage of coal activated carbon. In addition, adsorption beds become weaker as time passes. The efficiency drops due to the dye removal rate become weaker, and the adsorbent capacity decreases [38]. Adsorption on activated carbon has been widely applied in wastewater treatment industries as it is regarded as a highly efficient process. Nevertheless, the high production and regeneration cost of activated carbon, as well as its effect, which may produce other pollutants, has prompted researchers to develop cheaper alternatives [39]. A number of waste materials are being studied especially agricultural biomass as it has little or no economic value and often poses disposal issues. Some low-cost agricultural biomass-based adsorbents such as tea waste, hazelnut husk, bacteria aggregate, rice ash, pineapple leaf, and fruit waste have been tested to be effective in dye removal process [40]. Used activated carbon would have fewer active pores to trap adsorbates as well. However, technologies nowadays have found a way to regenerate the adsorbent, allowing it to be used again [41]. Other than that, the development of activated carbon derived from renewable materials has been studied thoroughly in recent years. A renewable resource such as biomass and specific wastes has been put to research and to activate carbon. Studies found that the cost of using renewable resource derived activated carbon can be cheaper than coal-derived activated carbon [42]. In addition, deriving activated carbon from wastes can also help reduce current unwanted wastes and further protect the environment. Furthermore, the cost to develop activated carbon from waste is rather cheap, making it a viable treatment option nowadays. Moreover, scientists have found new treatment methods involving adsorption with biological treatment processes. Bio-adsorption is a combination of a biological method using biomass such as agricultural waste with adsorption. The adsorption capacity and sustainability are better than normal adsorption [43]. However, the tests carried out are on a laboratory scale. The difficulty in advancing this treatment to industrial-sized is because of the high utilization cost. It should be further studied such that a more economical treatment method can be found. There are mainly four categories of agricultural biomass-derived adsorbent, such as leaf-based, peel-based, stem-based, and seed-based adsorbents. The percentage of color removal by various agricultural biomass-based adsorbent are tabulated in Table 8.

Table 8. Various plant biomass as an adsorbent for RBBR and RBV-5R dye removal.

Dye	Adsorbent	Plant parts	Removal (%)	Reference
RBBR	Xanthium italicum	leaf	95	[58]
	Tea leaves	leaf	11.39	[57]
	Mangosteen peel	peel	80.35	[59]
	Orange peel	peel	14.92	[57]
	Rambutan peel	peel	40.5-90.3	[57]
	Pomegranate peel	peel	81.35	[60]
	Watermelon rind	peel	41.8-97.2	[61]
	Pinecone	seed	48-98	[46]
	Green coconut	seed	79	[62]

Dye	Adsorbent	Plant parts	Removal (%)	Reference
	Orange peel	peel	75-80	[63]
	Bagassa <i>guianensis</i> Aubl	stem	10-85	[53]
	Coconut mesocarp	seed	8-85	[53]
	Cocoa pod husk	seed	92-100	[35]

7. Conclusions

To conclude, it is essential to develop an effective treatment method on dye wastewaters such that the environment is not damaged or disturbed. Although there are many types of treatment methods, not all are viable options. The limited source of funding can be a major problem as dye wastewater treatment is a complex process. Some equipment required can be expensive. Furthermore, without proper planning and treatment process, and inefficient treatment method could be wasteful. Thus, adsorption using activated carbon becomes a better choice for removing dye from wastewater. Adsorption mainly uses the porous sponge-like holes in activated carbons to trap and capture dye molecules. Factors such as pH value, dye concentration, temperature, adsorbent dosage, and contact time should be measured properly such that the removal rate can be optimized. In adsorption, the most efficient type of activated carbon is coal-derived activated carbon. The efficiency of treatment is 99.8%, and it can treat various types of dyes and even heavy metals. The regeneration of activated carbon can be done to renew the adsorption capacity of the activated carbon such that less adsorbent would be disposed of. However, the disposal of adsorbents is not discussed in this paper. Therefore, more research is to be conducted on how to dispose of unwanted adsorbent in case of regeneration or reactivation failure of the adsorbent. The consequences of disposing of adsorbent should also be taken into consideration such that no environmental damage is done.

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Conflicts of Interest

The authors declare no conflict of interest.

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