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Evaluation of the use of agricultural waste materials as low-cost and eco-friendly sorbents to remove dyes from water: a review

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ABSTRACT

Toxic pollutants, especially dyes, are the main source of chemical pollutants and pose a major challenge to conventional water treatment systems such as activated sludge. Therefore, there is a need to develop appropriate techniques to eliminate these toxic dyes from different media. There are several limitations to using different techniques such as coagulation, chemical oxidation, membrane separation process, electrochemical and aerobic and anaerobic microbial degradation to remove these pollutants from wastewater. Adsorption is a promising method for removing dyes due to its high efficiency and selectivity, simplicity, low cost and recyclability of the adsorbents. It has recently been demonstrated that using activated carbon is a highly efficient technology in removing dyes from wastewater. However, its extensive use is restricted because of its relatively high cost encouraging researchers to investigate alternative sources of unconventional, efficient, low-cost and environmental-friendly adsorbents. Agricultural waste materials are the most widely used bio-sorbents for removing dyes. These materials were characterized by different techniques such as adsorption–desorption of nitrogen at 77 K, Fourier-transform infrared spectroscopy, X-ray diffraction, scanning electron microscopy-energy-dispersive X-ray spectroscopy etc, and it was found that they have a loose and porous structure and contain many functional groups in addition to being low-cost, high adsorption capacity as well as efficiency and low energy demand. This made them materials capable of adsorption of various contaminants. But one of their main drawbacks is that they present a small surface area, often less than 50 m^2/g. The dye adsorption efficiency of these adsorbents varies depending on the pH of the solution, initial dye concentration, adsorbent dosage and process temperature. It was found that the most efficient agricultural wastes for the adsorption of Methylene Blue and Malachite Green were papaya seeds (555.56 mg/g; ~80%) and orange peels (483.63 mg/g), respectively. This review highlights the evaluation of the use of various agricultural waste materials as low-cost adsorbents for dye removal from wastewater, which is a safer alternative to traditional adsorbents. Some of the fundamental principles of dye adsorption were also outlined. These adsorbents are recommended for removing dyes from real wastewater under a continuous design to achieve commercial objectives.

Keywords: Agricultural waste materials; Dyes removal; Efficiency; Low-cost adsorbent

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1. Introduction

Although about 70% of the Earth's surface is covered by water [1], only 2.5% is useable as freshwater [2]. Freshwater demand increases with population density, urbanization, agriculture, and industrialization [3–13]. Freshwater scarcity has become a worldwide concern [14]. The World Health Organization estimates that 1.8 billion people will be under water stress by 2025 [15] and may reach 6 billion by 2050 [16]. Water pollution is a highly controversial issue on a global scale due to its long-term consequences [17]. Dyes are one of the most important water pollutants in effluents from various industrial activities such as textile, pesticide, food, pharmaceutical, paints, rubber, paper, varnish, tanning and cosmetics [18–38]. Among these industries, the textile industry ranks first as producing the largest amount of wastewater, beyond 56% [39–41]. According to recent estimates, the annual production is 10^6 tons of more than 100,000 commercial dyes [42–45]. During the dyeing process, 10%–15% of the dye is lost, and approximately 20% of the lost dye is found in industrial wastewater [45,46]. The colour concentration in the wastewater can exceed 1 mg/L [47,48]. In addition to high toxicity, many dyes are highly carcinogenic [49], such as acid dyes (e.g., Acid Blue 25), azo dyes (e.g., Methyl Red), direct dyes (e.g., Congo Red), reactive dyes (e.g., Reactive red 120), nitro dyes (e.g., Martins yellow), sulfur dyes (e.g., Sulfur Brilliant) etc. Dissolved in the water, the presence of dyes reduces the dissolved oxygen, which greatly affects aquatic plants and species [50]. Conventional physical and chemical methods cannot destroy these dyes and are difficult to degrade using biological methods [21]. The aromatic structure of the dye molecules makes them more stable and difficult for biodegradation. However, during decomposition, synthetic dyes can create toxic secondary chemicals that are destructive to the aquatic environment [51–53] and can cause serious damage to the aquatic environment and the health of living organisms if left untreated [54–56]. Therefore, remediation of the dyeing wastewater poses challenges to researchers and requires them to meet the laws imposed by the current legislation related to the quality of the treated water before discharging it into the environment [57]. Various physical, chemical and biological techniques, such as adsorption, ion-exchange, solvent extraction, liquid–liquid extraction, membrane separation, chemical oxidation, precipitation, ozonation, coagulation, electrochemical, biological degradation, etc. are used for the removal of dyes from wastewater [33,41,58–70]. Most of these technologies are associated with disadvantages such as toxicity, by-products, energy consumption, smelly, long processing period, high cost, and secondary sludge production [71–85]. Unlike other technologies, adsorption can be considered as a promising and effective physico-chemical method and an attractive alternative for wastewater treatment. This process has some advantages compared to other methods, such as cost-effectiveness, high removal efficiency and selectivity, high adsorption capacity, simple operation process, easy operation technique, sludge-free process, high quality of the treated wastewater, reversibility, recyclability of the adsorbents, low energy consumption. It does not produce deleterious substances [13,14,21,86–95]. Adsorption is affected by many factors, such as the type of adsorbent and its functional groups, surface area, adsorbent particles size, the type and size of the pores, adsorbate–adsorbent interactions, contact time, agitation speed, temperature, and pH [14,96–101]. Accordingly, this work reviews the challenges and opportunities of the adsorption methodology available as the current state of the art in removing dyes from wastewater. Adsorbents usually used to remove various pollutants, particularly dyes, from wastewater include activated carbon, biochar, fly-ash, clays, silica gel, chitin, chitosan, zeolites, ion-exchange resins, agricultural waste materials (AWM), industrial by-products, and biological and polymeric materials [102–112]. The adsorbent selection is the most important step in the adsorption process, which depends on many factors such as cost-effectiveness, accessibility, non-toxicity and high efficiency [113]. Activated carbon is one of the most widely used adsorbents due to its large active surface area, high porosity, various functional groups on the surface, and high adsorption capacity when removing dyes from wastewater. However, it is not applicable for removing dyes in a large scale due to its high cost [13,14,21,86–89,114–116]. Therefore, researchers are always focused on finding an alternative to activated carbon with effective and low-cost sorbents. Since almost two decades ago, various non-traditional and cost-effective alternative adsorbents derived from natural materials and industrial transformation are explored [117–119]. AWM are considered as very effective bio-adsorbents when applied to remove dyes. The advantages of these materials lie in their inexpensive, eco-friendly, high efficiency and have high adsorption capacity and low energy consumption. They are also accessible in everywhere, considered as sustainable sources, can be used as a mixture of inorganic and organic substances and present different functional groups (such as hydroxyl, carboxyl, carboxyl, amino, nitro, etc.) and porous and loose structures [120–125]. The adsorption process for dye removal from wastewater is an ideal and effective alternative to other expensive treatment methods, especially when the adsorbent is low-cost, such as AWM. Low energy consumption and cost are the most important reasons for favouring adsorption. Several examples of case studies of agricultural waste materials, including coir pith, orange peel, banana peel, rice husk, straw, date pit, oil palm trunk fibre etc. were considered [109,111,112,118,126–155].

Studies on using agricultural waste as low-cost adsorbents have been ongoing for over two decades. In this review, a wide list of low-cost sorbent has been collected. The review evaluates different agricultural waste materials as inexpensive adsorbents for removing dyes from wastewater. The review also outlines some of the essential principles of dye adsorption onto adsorbents. The present review is expected to be useful in identifying cost-effective and efficient adsorption methods for the remediation of dyeing wastewater. Finally, the current review also targets various research gaps and their possible solution.

2. Classification of dyes

Dyes are mainly derived from natural (plants and animals) and synthetic (in which organic and inorganic compounds are included). Synthetic dyes are classified based on their chemical structure, application, and particle charge. They can be classified classically according to the Color Index (CI) number, where each dye is given a unique number of

five digits depending on their chemical structure. However, due to the colour nomenclature's difficulties, the application classification is often preferable [156]. Depending on the application, synthetic dyes can be divided into acid, basic, azo, direct, reactive, disperse, nitro, fluorescent bleach, mordant, food, grain, pigment, natural, oxidation base, reagent, solvent, sulphur and vat dyes [156,157]. According to the chemical composition, synthetic dyes can be classified as, anthraquinone, azo, triarylmethane, basic, acidic, formazan, oxazine, phthalocyanine, stilbene, cyanine, diphenylmethane, xanthenes, diazahemicyanine, azine, acridine, hemicyaninem, indigoids, benzodifuranone, nitro, nitroso, and styryl [89,158,159]. Moreover, dyes can be classified based on their particle charge into cationic, anionic, and non-ionic. All basic dyes such as Methylene Blue (MB), Malachite Green (MG), Crystal Violet, Original Red 29 (OR29), Blue Base 159 (BB159), Yellow Base 28 (YB), Rhodamine B etc. are cationic dyes. These dyes are used in paper, paint, nylon and polyester industries. Anionic dyes can also be divided into direct, acidic, and reactive dyes. Congo Red (CR), Methyl Orange, Direct Red 23 (DR23), Reactive Black 5 (RB5), etc. are among the most common anionic dyes. In addition, non-ionic dyes include dispersed dyes such as Direct Green 97 (DG97) [158,160,161]. The classification, chemical structure, applications, and eco-toxicological effects of some common dyes are shown in Table 1.

3. Dye removal techniques

Each dye has three parts: chromophore, chromogen and auxochrome. Chromophore is an unsaturated group that provides colour, and chromogen holds chromophore. It plays an important role in determining the final colour and its stability. While auxochromes are acidic or basic substituted groups that intensify the colour of the dye molecules and increase their water solubility, thus, it increases their ability to adhere to the fibres [179].

Current dye removal treatment techniques involve biological, physical, physico-chemical, and chemical processes (Fig. 1). However, these techniques have advantages and disadvantages. Most of these traditional methods are not widely applicable due to the high costs and large quantities of sludge formed at the end of the treatment processes [180].

- Biological processes: may involve the use of microorganisms under aerobic (in the presence of oxygen) or anaerobic (without oxygen) conditions to reduce dilute organic dyes [13].
  - Advantages: Remove several types of dyes at once, cheap and no foam formation, environmental-friendly and cost-competitive, less sludge production, non-dangerous product, less consumption of water, and the ability of enzymes to degrade dyes [79,88].
  - Disadvantages: Low reliability, long duration times, large land area, not effective for all dyes such as azo dyes, not eliminate all dye particles, and low biodegradability of recalcitrant chemicals [88,179,181–183].
- Advanced oxidation processes: this is one of the physico-chemical treatment processes for the degradation of synthetic dyes that has attracted the attention of researchers in recent years. The photocatalysis process is one of the most important because of its high efficiency and short treatment time. The mechanism of this method is based on the generation of hydroxyl radicals ($OH^{\bullet}$) to oxidize the pollutants present in the wastewater. Hydroxyl radicals are strong, highly reactive oxidizing agents (E° = 2.8 V) that attack organic pollutants and convert them to simpler organic compounds or fully mineralize in CO_2 and H_2O. A disadvantage of hydroxyl radical is that it has a short half-life and is non-selective in nature, in addition to being only effective in the acidic pH range 2–5 [186–191].
  - Advantages: Effective decolourisation of both soluble and insoluble dyes, in situ reactive radical development, little to no chemical intake, pollutant mineralization, and effective for molecules recalcitrant (drugs, dyes, etc.) [88,184].
  - Disadvantages: Long time, sludge generation, economically unsustainable for small and medium enterprises, and technical limitations [88,184].
- Ion-exchange method: involves removing undesirable ionic contaminants from the water by exchanging them with another ionic substance.
  - Advantages: Produce high-quality water, ion-exchangers can be regenerated, no loss of adsorption after regeneration, and low maintenance cost.
  - Disadvantages: Effective to a limited number of dyes, very sensitive to pH, conventional non-selective resins, limited selective commercial resins, and long-time of regeneration [88–91].
- Membrane filtration method: is an assessment of water quality through a special filter, that is, membrane filter, to trap the microorganisms. Membranes also concentrate and purify many types of dye from their aqueous solutions. It is now used extensively in process industries to concentrate, purify and improve the finished product.
  - Advantages: Removes all dye types, effective for water recovery and reusing, small space, easy, fast, practical, and suitable even at high concentrations levels, products with high quality, effluent care and low solid waste output.
  - Disadvantages: Concentrated sludge production, high cost, high energy demands, different design of membrane filtration systems, high maintenance, rapid membrane obstruction, fouling with high concentrations [88,185].
- Nanofiltration and ultrafiltration processes: Nanofiltration is a filtration process using a nanoporous membrane that is used in waters with low total dissolved solids. Ultrafiltration is a membrane technique that removes the dissolved and colloidal material in low transmembrane pressure.
  - Advantages: Remove any dye, separation of low molecular organics and divalent ions and environmental-friendly membrane.
  - Disadvantages: High cost, high energy consumption, and high pressure [88–90,185].
- Electrochemical process: The electrochemical treatment of wastewater is considered as one of the advanced oxidation processes, potentially a powerful pollution control method.

Table 1
Characteristics and application of some common dyes

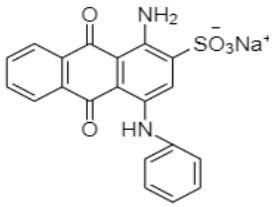
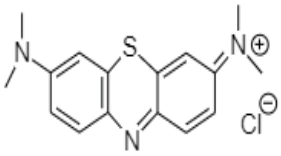
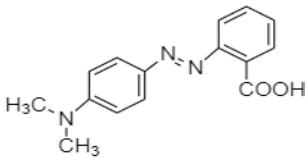
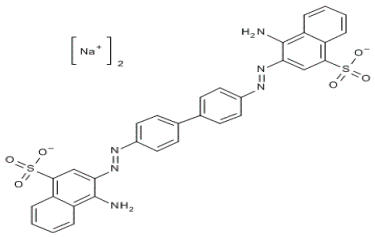
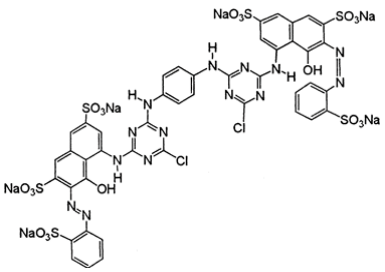
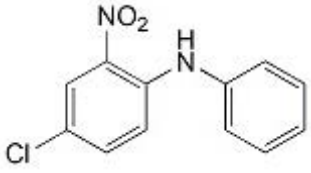
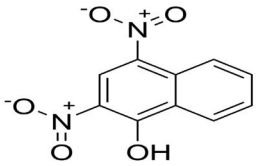
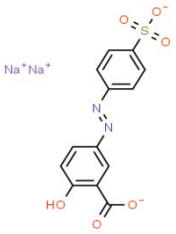
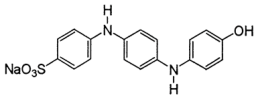
Types of dye	Examples of dyes	Chemical structure	Applications	Toxic effects	References
Acid dye	Acid Blue 25 Acid Yellow 132 Acid Blue 25 Acid Violet Acid Brilliant Blue	 Acid Blue 25	Textile, wool, silk, leather, pharmaceutical, nylon, and modified acrylics	Vomiting, nausea, diarrhoea, cancer	[162–170]
Basic dye	Methylene Blue Malachite Green Methyl Violet Rhodamine B Basic Red 1 Basic Yellow 2 Crystal Violet Original Red 29 Blue Base 159 Yellow Base 28	 Methylene Blue	Paper, nylon, polyacrylonitrile, and polyesters	Agitation, bluish-coloured lips, confusion, dark urine, fever, difficulty breathing, headache and dizziness	[162,163,166,170]
Azo dye	Methyl Red Methyl Orange Trypan Blue Direct Black 22 Acid Orange 20	 Methyl Red	Textile and food industries	Carcinogenic human and animal tumours, it has effects on the eyes, skin and digestive system	[37162,164,168–171]
Direct dye	Congo Red Direct Red 28 Direct Black 38 Brilliant Yellow Direct Blue 151 Direct Dark Green NB Direct Brown 44	 Congo Red	Cotton, paper, leather, wool, silk and nylon. They are also used as pH indicators and as biological stains	Respiratory tract irritation and cancer	[162,163,172–174]
Reactive dye	Reactive red 120 Reactive Red 147 Reactive Black 5 Reactive Blue 19	 Reactive red 120	Textile, wool, silk, cotton	Skin irritation, skin cancer, genotoxicity, mutagenicity, cytotoxicity, high and undesired levels of dissolved solids in the effluent. Allergic reaction in eyes, skin, mucous membrane, and the upper respiratory tract	[162,163]
Disperse dye	Disperse Red 9 Disperse yellow 26 Disperse Violet 1 Disperse Red 60	 Disperse yellow 26	Coloring synthetic fibres such as polyester, nylon, acrylic, and acetate rayon	Sensitization and elicitation to the body	[162,163,175]

Table 1 (Continued)

Table 1

Types of dye	Examples of dyes	Chemical structure	Applications	Toxic effects	References
Nitro dye	Martins yellow Palatine Orange Nitroso Green	 Martins yellow	Dyeing of human hair, dye wool	Decreases light, penetration and photosynthetic activity, carcinogenic and mutagenic	[162,163,176]
Mordant dye	Mordant Red 11 Mordant yellow 10 Mordant Black 17	 Mordant yellow 10	Textile fibres such as wool, silk, and leather	Allergic reactions	[162,163,177,178]
Sulfur dye	Sulfur Brilliant Green Sulfur blue 10 Sulfur Black 1 Leuco Sulfur Black 1 Phthalic anhydride	 Sulfur blue 10	Dyeing cellulosic fibres and cotton	Skin irritation, itchy or blocked noses, sneezing, sore eyes, carcinogenic	[162,163,166]

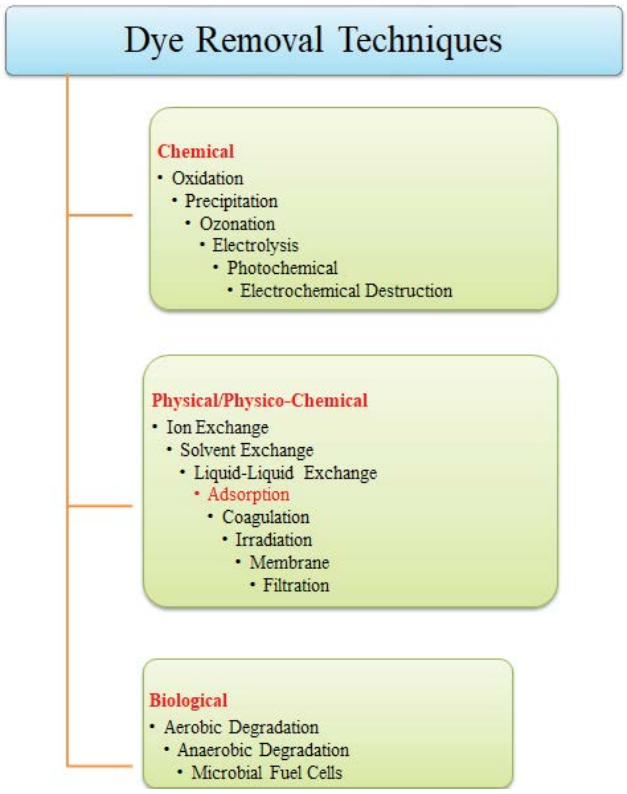


Fig. 1. Various wastewater treatment techniques for dyes removal.

- Advantages: No chemicals, no produce of sludge, direct or indirect oxidation flexibility, space saving, high efficiencies, environmental-friendly.
- Disadvantages: High cost of electricity, ferrous oxide sludge [88,185–192].

- Flocculation and coagulation process: Coagulation is the destabilization of colloidal particles brought about by adding a chemical reagent called coagulant. Flocculation is the agglomeration of destabilized particles into microfloc and after into bulky floccules which can be settled called floc.
  - Advantages: Significant reduction in biochemical oxygen demand (BOD), chemical oxygen demand (COD), and a wide range of commercial chemicals.
  - Disadvantages: Addition of non-reusable chemical substances (coagulants, flocculants, aid chemicals), production a large volume of sludge, high residual aluminium concentration [88].
- Irradiation method: reduces or eliminates pests and the risk of food-borne illnesses and prevents or slows spoilage and plant maturation or sprouting.
  - Advantages: Effective at laboratory scale, adsorbent loss, effective laboratory oxidation.
  - Disadvantages: Expensive, the need for a lot of dissolved O_2 [88].
- Biodegradation process: consists of three stages: bio-deterioration, bio-fragmentation, and assimilation.
  - Advantages: Reusable, high efficiency, nontoxic, elimination of various types of azo dyes at once, and short time (maximum of 30 h).

- Disadvantages: Producing sludge, unreliable enzyme production, needs nitrogen, confined area to grow, effective to a limited number of dyes (suitable only for azo dye removal) [88].

- Ozonation method: is an advanced oxidation process using ozone, a reactive gas with low solubility, usually generated on-site. Once dissolved in water, it undergoes complex decomposition and oxidation reactions.
  - Advantages: Good decolourisation, COD reduction, no sludge, automated already at lab and pilot scale, effective dye removal technique, quick reaction, ozone can be applied gaseously.
  - Disadvantages: Very expensive, short half-life (20 min), unstable method [88].

As a result, many of these techniques effectively remove dye in the laboratory scale. However, because they are expensive, huge energy consumption, and long processing time, large amounts of sludge may be produced in some cases, which must be handled optimally to avoid further environmental contamination, making their use commercially restricted [192]. Among the various treatment techniques, adsorption is considered promising, attractive and the most efficient method [191–197].

4. Adsorption process

Adsorption is a surface phenomenon in which a mass transfer occurs between two immiscible forms at a biphasic interface, such as a gas–solid, gas–liquid, or liquid–solid. The solid is called adsorbent while the accumulated form is called adsorbate. Adsorption processes have gained importance as a viable method for wastewater treatment. Cost-effective, eco-friendly, flexibility in design and use, reversibility and recyclability, high efficiencies and simplicity are the most important advantages that make adsorption superior to other traditional methods [191–198]. Adsorption is a physical, chemical, or physico-chemical process. Table 2 and Fig. 2 show the difference between physical and chemical adsorption [13,21,199–202].

4.1. Agricultural waste materials as low-cost adsorbents for dyes removal

Activated carbon is one of the most common adsorbents due to its active surface and high adsorption capacity in removing various pollutants from aqueous solutions. However, it is not practical to apply to remove dyes because of the high cost of production [64,194]. It is known that the most important features of adsorbents are large surface area, high adsorption capacity, high adsorption efficiency, large porosity, stability, feasibility, compatibility, eco-friendliness, low cost, and easy availability [203–111]. Among the investigated adsorbents in dye removal that have received increasing attention are AWM. The advantages of utilizing these materials are low cost, high efficiency, accessible in almost every part of the world, minimum energy consumption, can be used without treatment, simple maintenance, high adsorption capacity, and absence of dangerous by-products [112–122]. The use of AWM as adsorbents is very important for removing many pollutants from wastewater on the one hand and for the disposal of AWM themselves on the other hand [123]. AWM have a porous structure [123], and its composition can include various functional groups such as hydroxyl, carboxylate, amino, nitro, etc. [5,123–204]. The presence of such functional groups can enhance the removal of dyes, and can play a role in the selective removal of some dyes (cationic, anionic or non-ionic) [205,206]. The performance of AWM can also be improved by modifying it with some chemical reagents such as acids, bases, salts, oxidants, etc. [206,207].

The most common AWM to remove dyes are pistachio peels, rice hulls ash, coffee powder, pomelo peels, garlic peels, pomegranate peels, coconut shells, orange peel, banana peel, tea waste, squash coal, peat bagasse, and coal fly ash [109,111,112,118,126–155,208–215].

Determining the physico-chemical surface properties of adsorbents is essential in predicting the adsorption capacity. Specific surface area, particle size, pore size distribution, pore size, point zero charge (pH_{pzc}), and functional groups on the surface of the adsorbent are the key parameters that

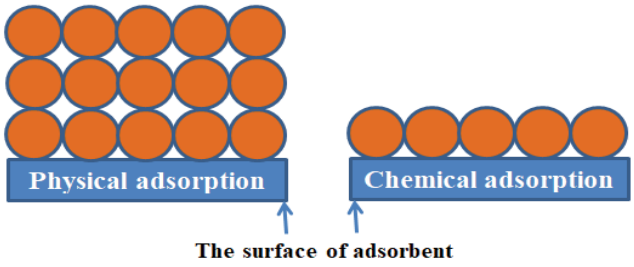


Fig. 2. Difference between physical and chemical adsorption.

Table 2
Differences between physical and chemical adsorption

Physical adsorption	Chemical adsorption
Adsorbate and adsorbent bonding is weak physical (van der Waals forces, electrostatics, hydrogen bonds)	Adsorbate and adsorbent bonding is strong chemically (ion-exchange, covalent bond, electron or proton donating or receiving)
Reversible in nature	Irreversible in nature
Process efficiency decreases with increasing temperature (exothermic process, $\Delta H < 0$)	Process efficiency increases with decreasing temperature (endothermic process, $\Delta H > 0$)
Multi-layered process	Mono-layered process
Heat of adsorption is low (0–20 kJ/mol)	Heat of adsorption is high (80–400 kJ/mol)

determine the efficiency of absorbents [216]. Therefore, it is necessary to understand the properties of the adsorbents and the adsorbent–adsorbent relationship to achieve the desired removal of the adsorbate. Therefore, several techniques (such as scanning electron microscopy (SEM), X-ray diffraction spectrum, Brunauer–Emmett–Teller (BET), Fourier-transform infrared spectroscopy (FTIR)) are used to characterize the physico-chemical surface properties of the adsorbent to determine the feasibility of use.

Morphological investigation by SEM analysis is a widely used technique to describe the detailed morphology, topology and structure of the surface of an adsorbent. All studies that used AWM as adsorbents proved that they have a porous and loose structure and that their surfaces contain active sites suitable for adsorption of the dye from aqueous solutions and wastewater [210–228]. For example, the SEM images of coconut bunch waste [126], neem bark, and mango bark [127] exhibit a caves-like, uneven and rough surface morphology. The meranti sawdust has considerable layers of pores where there is a good possibility for dye to be adsorbed [135]. The pores within the banana stalk waste particles were highly heterogeneous. All previous research proved that the surface of the adsorbent before adsorption differed from that after, which indicates the capability of dye adsorption. The functional groups on the surface of the adsorbent are usually determined by using FTIR spectra in the region of 4,000–400 cm^{-1}. For example, the functional groups (–OH, C–H, –COOH, $–NH_2$) present on the surface of the Olive Pomace are variable [100]. The main surface functional groups present in the rice husk Ash are carbonyl and siloxane groups [211]. The hydrophobic functional group of the C–H and the C–O group were also found on the surface of coffee ground powder [212]. Various functional groups such as hydroxyl, carbonyl, phenolic, amino, carboxylate, nitro, and azo groups in the AWM make them selective and highly capable for adsorption of dyes from wastewater [208–228]. The specific surface area is one of the main characteristics of the adsorbents because the larger surface area can contain more active sites and also presents a larger pore size which enhances the interaction between the adsorbents and the adsorbates and then greatly affects the efficiency and capacity of the adsorption [13]. Some important surface properties of AWM are mentioned in Table 3. The relatively small surface area of AWM is one of the main drawbacks of these adsorbents, ranging from 0.034 to 123 m^2/g in the scope of the research we have covered.

The dye solution's initial pH, which strongly influences the surface charge density and the degree of ionization and adsorption efficiency of the adsorbent, plays a considerable role throughout the adsorption process. The degree of adsorption differs with the change in the pH of the medium. At high pH, the removal efficiency of the cationic dye increases because of the adsorbent's negatively charged surface that favours the cationic groups' adsorption, resulting in an improvement in adsorption. However, at low pH, it shows reverse order [79].

The pH at the point of zero charges (pH_{pzc}) of the adsorbent is defined where the net charge of the adsorbent surface is equal to zero. In other words, pH_{pzc} describes the state when the positive charge equals the negative charge on the surface of the adsorbent. When pH is less than pH_{pzc}, favourable anionic dye adsorption is achieved. However, the adsorption of cationic dye is observed when the surface

Table 3
Surface properties of agricultural waste materials for dye removal

Agricultural waste materials	S_{BET} (m^2/g)	V_{total} (cm^3/g)	D_m (μm)	References
Activated carbon	1,400	–	–	[229]
Rice husk	2.4	–	–	[209]
Rice husk	33.5	0.0166	9.9×10^{-4}	[211]
Rice husk	36.4	0.1049	4.3×10^{-3}	[26]
Pomelo peels	0.034	–	1.3×10^{-3}	[213]
Eucalyptus bark	6.6	0.00343	–	[217]
Hazelnut shells	14.6–21.6	–	12.5–50.0	[129]
Walnut	13.3	–	12.5	[129]
Cherry	11.5	–	12.5	[129]
Oak sawdust	10.0	–	12.5	[129]
Pitch pine	9.4	–	12.5	[129]
Banana peels	20.6–23.5	–	–	[130]
Orange peels	20.6–23.5	–	–	[130]
Pumpkin seed hull	91.8	–	–	[149]
Garlic peels	0.561	1,120,000	8.0×10^{-3}	[131]
Luffa cylindrica fiber	123	–	1.0–10.0	[228]
Yellow passion fruit waste	30	0.07	3.0×10^{-3}	[132]
Wheat shells	0.67	–	–	[133]
Cucumber peels	0.855	–	–	[229]
Potato peels	0.614	–	–	[229]

becomes negatively charged (pH > pH_{pzc}) [79]. The pH_{pzc} of the adsorbent is a good indicator of the electro-chemical properties of functional groups present on its surface [230].

The contact time, which represents the time required for adsorption to reach the equilibrium state [231], is another important factor for the separation system design [232]. Studying the effect of the initial adsorbent dose and dye concentration on the adsorption process is also necessary. Previous literature studies indicated that adsorption capacity (q_e) increases with dye concentration and decreases with adsorbent dose. However, it is reversed for the process efficiency. The separation efficiency increases with the adsorbent dose due to the increased surface area and available binding sites [233,234]. However, it decreases with adsorbate concentration due to the increasing ratio of dye molecules to the available adsorbent surface area [130].

The amount of dye adsorbed by AWM is calculated by mass balance. The adsorption efficiency (E, %) and capacity (q_e, mg/g) are determined by using Eqs. (1) and (2):

$$E(\%) = \left(\frac{C_o - C_e}{C_o}\right) \times 100 \tag{1}$$

$$q_e = \left(\frac{C_0 - C_e}{m}\right) \times V \tag{2}$$

where C_0 (mg/L) is the initial dye concentration, C_e (mg/L) is the dye concentration at the equilibrium, m (g) is the weight of absorbent used, and V (L) is the volume of the dye solution. The adsorption capacities of some AWM and studied conditions are summarized in Table 4.

4.2. Isotherm equilibrium and kinetic models

The adsorption isotherm of a solid–liquid system generally provides information about the adsorption mechanisms, the affinity of the adsorbent, the interactions between the adsorbent and adsorbate as well as surface properties and internal bonding [244]. Adsorption isotherm curves describe the relationship between adsorbed dye (q_e) capacity and the residual dye concentration in the solution at equilibrium [245,246]. Adsorption isotherms generally plateau at high dosages, corresponding to complete surface coverage [245,246]. In most cases, the equilibrium-state behavior of an adsorbent is studied based on Langmuir, Freundlich, and Temkin models [244–252]. The Langmuir isotherm assumes the formation of a monolayer on the sorbent based on chemical interactions. In addition, the energy at each site is assumed to be equal and no interaction occurs between molecules adsorbed on the surface [244–250]. The Freundlich isotherm describes physical sorption processes that occur on heterogeneous surfaces. In other words, this model can describe the surface heterogeneity. Generally, formation of multi-layers of solute molecules is expected during the process [244–248,251]. According to the value of "1/n", the process is deemed unfavourable ($1/n > 1$), irreversible ($1/n = 0$) or favourable ($0 < 1/n < 1$).

The Temkin isotherm model suggests that indirect contact between solute molecules during adsorption are significant and assumes that adsorption heat decreases as the coating of the surface progresses [244–248,252]. Table 5 shows the non-linear and linear expressions of these models.

One of the most important criteria for selecting an appropriate adsorbent is the rate of the adsorption process. The study of the kinetics of the process reveals the variation of the adsorption capacity with time. The kinetic measurements estimate the time required to reach process equilibrium and understand the adsorption mechanism. The most common kinetic models are pseudo-first-order, pseudo-second-order, Elovich, and intraparticle diffusion kinetic models [253–256].

The pseudo-first-order kinetic model is mainly applicable to describe the process in its first half [100,257]. The pseudo-second-order model indicates that adsorption occurs according to chemical reactions such as donating/receiving, or sharing electrons [100,257]. The dose of adsorbent and the specific surface area play an essential role in this model [258]. Although the Elovich model is commonly used to describe the heterogeneous chemical absorption of gases on solid surfaces, its applicability to liquid processes can help to predict the diffusion of dye molecules on the surface and the activation energy of the system [101,259]. The intraparticle diffusion model is widely used to determine the mechanism that controls the adsorption process either film diffusion or particle diffusion [101,260]. The expressions of kinetic models are exhibited in Table 6.

The surface of agricultural wastes may contain polar functional groups such as hydroxyl, carbonyl, carboxyl etc. These functional groups can play a role in the adsorption process between dye ions and polar sites on the surface of the adsorbent. Thus, the adsorption reaction is the rate-limiting step in most desorption processes by agricultural wastes. The adsorbent's specific surface area and the dye's amount are two major factors affecting the adsorption rate. Thus, it was concluded that most adsorption processes that use agricultural waste as adsorbents follow the Langmuir isotherm- and the pseudo-second-order kinetic models [261]. Table 7 summarises the studies performed on applying isotherm and kinetic models for removing dyes using agricultural waste.

4.3. Effect of temperature and thermodynamics

The study of the effect of temperature and thermodynamics is important in describing the adsorption process and achieving an equilibrium state. Thermodynamic parameters include Gibbs free energy change ($\Delta G°$), enthalpy change ($\Delta H°$), and entropy change ($\Delta S°$). The value of $\Delta G°$ can be calculated from Eq. (3):

$$\Delta G° = -RT \ln K \tag{3}$$

where R is the general gas constant (8.314 J/mol·K) and K is the distribution constant [equilibrium constant; Eq. (4)] at temperature T (K). The $\Delta G°$ for physical adsorption processes generally ranges from 0 to 20 kJ/mol, while it varies between 80 and 400 kJ/mol for chemical processes [99,261].

$$K = \frac{q_e}{C_e} \tag{4}$$

Table 4
Adsorption efficiencies and capacities of some agricultural waste materials at the different studied conditions

Agricultural waste materials	Dye	Contact time (min)	Temperature (K)	pH	Dye concentration (mg/L)	Adsorbent dose (g)	E (%)	q_{max} (mg/g)	References
Activated carbon	Indigo Blue	140	298–353	2–11	5–40	0.1–1.0	~75	53.00	[227]
Coconut bunch	Methylene Blue	330	303	2–12	50–500	0.2	–	70.92	[126]
Sunflower seed husk	Methylene Blue	420	298–353	2–11	25–100	0.1	~100	45.25	[128]
Hazelnut shells	Methylene Blue	60	293	2.5–4.2	250–1,000	0.5	–	76.9	[129]
Walnut sawdust	Methylene Blue	60	293	2.5–4.2	250–1,000	0.5	–	59.17	[129]
Cherry sawdust	Methylene Blue	120	293	2.5–4.2	250–1,000	0.5	–	39.84	[129]
Oak sawdust	Methylene Blue	60	293	2.5–4.2	250–1,000	0.5	–	29.94	[129]
Pitch pine sawdust	Methylene Blue	60	293	2.5–4.2	250–1,000	0.5	–	27.78	[129]
Hazelnut shells	Acid Blue 25	60	293	2.5–4.2	50–500	0.5	–	60.20	[129]
Walnut sawdust	Acid Blue 25	–	293	2.5–4.2	50–500	0.5	–	36.98	[129]
Cherry sawdust	Acid Blue 25	–	293	2.5–4.2	50–500	0.5	–	31.98	[129]
Oak sawdust	Acid Blue 25	–	293	2.5–4.2	50–500	0.5	–	27.85	[129]
Pitch pine sawdust	Acid Blue 25	–	293	2.5–4.2	50–500	0.5	–	26.19	[129]
Banana peels	Methylene Blue	1,440	303	4–10	10–120	0.1	–	15.9	[130]
Banana peels	Methyl Orange	1,440	303	4–10	10–120	0.1	–	17.2	[130]
Banana peels	Rhodamine B	1,440	303	4–10	10–120	0.1	–	13.2	[130]
Banana peels	Congo Red	1,440	303	4–10	10–120	0.1	–	11.2	[130]
Banana peels	Methyl Violet	1,440	303	4–10	10–120	0.1	–	7.9	[130]
Banana peels	Amido Black 10B	1,440	303	4–10	10–120	0.1	–	7.9	[130]
Orange peels	Methylene Blue	1,440	303	4–10	10–120	0.1	–	13.9	[130]
Orange peels	Methyl Orange	1,440	303	4–10	10–120	0.1	–	15.8	[130]
Orange peels	Rhodamine B	1,440	303	4–10	10–120	0.1	–	9.1	[130]
Orange peels	Congo Red	1,440	303	4–10	10–120	0.1	–	7.9	[130]
Orange peels	Methyl violet	1,440	303	4–10	10–120	0.1	–	6.1	[130]
Orange peels	Amido Black 10B	1,440	303	4–10	10–120	0.1	–	3.8	[130]
Garlic peels	Methylene Blue	210	303–323	4–12	25–200	0.3	–	142.86	[131]
Yellow passion fruit waste	Methylene Blue	4,320	298	2–11	5–600	0.1–1.0	~100	74.70	[132]
Wheat shells	Methylene Blue	75	303–323	2–9	100–400	0.1–3.0	~100	21.50	[133]
Cedar sawdust	Methylene Blue	45	293	2–11	40	0.001–0.015	~97	142.36	[134]
Crushed brick	Methylene Blue	45	293	2–11	40	0.001–0.015	~96	96.61	[134]
Meranti sawdust	Methylene Blue	180	303–333	3–12	50–200	0.1–1.2	–	158.73	[135]
Mansonia wood sawdust	Methylene Blue	–	299	–	120	–	93.6	28.07	[136]
Beech sawdust	Methylene Blue	190	296	1.7–13	1.4–14	–	–	11.40	[137]
Rice husk	Methylene Blue	2,880	305	3–8	10–125	0.02–0.12	–	40.58	[138]
Papaya seeds	Methylene Blue	180	303	3–10	50–360	0.05–1	–	555.56	[139]
Grass waste	Methylene Blue	180	303	3–10	70–380	0.05–1.2	~80	457.64	[140]
Paspalum notatum	Methylene Blue	360	303	3–8	30–100	0.01–0.08	~50	31.00	[141]
Pomelo peels	Methylene Blue	315	303	2–10	50–500	0.2	–	344.83	[142]
Guava leaf powder	Methylene Blue	20	303	7.5	100–800	2	~100	295.04	[143]
Jackfruit peels	Methylene Blue	180	303	2–11	35–400	0.05–1.2	–	285.71	[144]
Banana stalk waste	Methylene Blue	330	303	4–12	50–500	0.2	–	243.90	[145]
Palm kernel fibre	Methylene Blue	120	299–339	7.1	100–550	0.1	~100	233.41	[146]
Broad bean peels	Methylene Blue	320	303	2–10	30–325	0.3	–	192.70	[147]
Rubber seed shell	Methylene Blue	60	Room temperature	–	5–100	0.1–0.5	–	82.64	[148]
Castor seed shell	Methylene Blue	120	309	–	25–300	0.1–1	–	158.73	[149]
Pumpkin seed hull	Methylene Blue	110	303	2–11	25–300	0.3	–	141.92	[150]

Table 4 (Continued)

Table 4

Agricultural waste materials	Dye	Contact time (min)	Temperature (K)	pH	Dye concentration (mg/L)	Adsorbent dose (g)	E (%)	q_{max} (mg/g)	References
Pineapple stem	Methylene Blue	330	303	2–10	25–300	0.3	–	119.05	[151]
Coconut husk	Methylene Blue	20–180	–	2–12	50–500	0.25–1.5	100	99.00	[152]
Coffee husks	Methylene Blue	120–720	303–323	3–11	50–500	0.5–3.75	96	90.10	[153]
Fallen Phoenix tree's leaf	Methylene Blue	180	295–323	4.5–10	30–180	0.02	95	89.70	[154]
Hazelnut shells	Methylene Blue	1,440	298–328	3–9	–	0.25	–	87.98	[155]
Neem bark	Malachite Green	120	298	2–9	0.365–36.5	0.5–2	–	0.36	[223]
Tamarind shell	Congo Red	60–360	303	5.5–12	20–80	0.05–0.9	–	10.48	[235]
Azadirachta indica leaf	Congo Red	60–300	300	6.7	10–50	0.01–0.05	99	72.00	[236]
Teak tree bark powder	Methylene Blue	60	303–323	3–11	100–400	0.025–0.15	99.4	333.33	[237]
Tree fern	Acid Yellow 132	180	298	3.5–11.5	500–2,000	0.5–3	–	280.30	[238]
Pine sawdust	Acid Blue 256	180	298	3.5–11.5	500–2,000	0.5–3	–	398.80	[238]
Peanut hull	Methylene Blue	720	293	2–11	25–200	0.05–0.5	~100	68.03	[239]
Peanut hull	Brilliant Cresyl Blue	720	293	2–11	25–200	0.05–0.5	~100	–	[239]
Peanut hull	Neutral Red	720	293	2–11	25–200	0.05–0.5	~100	87.72	[239]
Coir pith	Acid Violet	120	303	3–11	40	0.05–1	~95	7.34	[240]
Coir pith	Rhodamine B	120	303	3–11	125	0.05–1	~95	94.73	[240]
Coir pith	Methylene Blue	80	303	3–11	125	0.05–1	~95	120.43	[240]
Coir pith	Acid Brilliant Blue	220	303	3–11	40	0.05–1	~76.6	5.57	[240]
Rice husk	Methylene Blue	150	305	3–8	10–125	0.02–0.12	88.7	40.58	[138]
Rice husk	Indigo Carmine	480	293–323	2–10	50–500	0.2–2.5	–	65.91	[241]
Rice husk	Safranin	2,880	305	–	10–125	0.06	–	178.08	[242]
Orange peels	Malachite Green	1,440	305	–	50–200	0.009	–	483.63	[243]
Jujuba seeds	Congo Red	300	303–333	2–12	25–100	0.01–0.3	~88	55.56	[133]

Table 5
Non-linear and linear equations of the most isotherm models used

Model name	Non-linear equation	Linear equation	Plot
Langmuir	$q_e = q_{max}\frac{K_L \cdot C_e}{1+K_L \cdot C_e}$	$\frac{1}{q_e} = \frac{1}{q_{max}K_L}\cdot\frac{1}{C_e}+\frac{1}{q_{max}}$ $R_L = 1/\left(1+\left(K_L \cdot C_o\right)\right)$	$1/q_e$ vs. $1/C_e$
Freundlich	$q_e = K_F \cdot \left(C_e\right)^{1/n}$	$\log q_e = \frac{1}{n}\cdot\log C_e + \log K_F$	$\log q_e$ vs. $\log C_e$
Temkin	$q_e = \frac{RT}{b}\ln\left(K_T \cdot C_e\right)$	$q_e = B\ln K_T + B\ln C_e$ $B = \frac{RT}{b}$	q_e vs. $\ln C_e$

The values of $\Delta H°$ and $\Delta S°$ can be obtained from the van't Hoff equation [Eq. (5)] as the slope and intercept of the plot of $\ln K$ vs. $1/T$, respectively.

$$\ln K = -\frac{\Delta H°}{RT} + \frac{\Delta S°}{R} \quad (5)$$

A negative value of $\Delta H°$ indicates the exothermic nature of the adsorbate's adsorption on the adsorbent's surface. On the other hand, a positive value of $\Delta H°$ refers to the endothermic nature of the adsorption process and the formation of strong bonds between the adsorbate and adsorbent. A positive value of $\Delta S°$ signifies an increase in randomness

Table 6
Non-linear and linear equations of the kinetic models studied

Model name	Non-linear equation	Linear equation	Plot
Pseudo-first-order	$\frac{dq_t}{dt} = k_1\left(q_e - q_t\right)$	$\ln\left(q_e - q_t\right) = \ln q_e - k_1 \cdot t$	$\ln(q_e - q_t)$ vs. t
Pseudo-second-order	$\frac{dq_t}{dt} = k_2\left(q_e - q_t\right)^2$	$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}t$	t/q_t vs. t
Elovich	$\frac{dq_t}{dt} = \alpha \cdot \exp\left(-\beta \cdot q_t\right)$	$q_t = \frac{\ln(\alpha\beta)}{\beta} + \frac{\ln t}{\beta}$	q_t vs. $\ln t$
Intraparticle diffusion	–	$q_t = k_{id}t^{0.5} + I$	q_t vs. $t^{0.5}$

Table 7
Isotherm and kinetic models used in various studies for dyes removal

Adsorbent	Dye	Fitted isotherm model	Fitted kinetic model	References
Activated carbon	Indigo Blue	Freundlich	Pseudo-second-order	[227]
Mango bark	Malachite Green	Langmuir	Pseudo-second-order	[126]
Neem bark	Malachite Green	Langmuir	Pseudo-second-order	[127]
Sunflower seed husk	Methylene Blue	Langmuir	Pseudo-second-order	[128]
Hazelnut shells	Methylene Blue	Langmuir	Pseudo-second-order	[129]
Walnut sawdust	Methylene Blue	Langmuir	Pseudo-second-order	[129]
Cherry sawdust	Methylene Blue	Langmuir	Pseudo-second-order	[129]
Oak sawdust	Methylene Blue	Langmuir	Pseudo-second-order	[129]
Pitch pine sawdust	Methylene Blue	Langmuir	Pseudo-second-order	[129]
Hazelnut shells	Acid Blue 25	Langmuir	Pseudo-second-order	[129]
Walnut sawdust	Acid Blue 25	Langmuir	Pseudo-second-order	[129]
Cherry sawdust	Acid Blue 25	Langmuir	Pseudo-second-order	[129]
Oak sawdust	Acid Blue 25	Langmuir	Pseudo-second-order	[129]
Pitch pine sawdust	Acid Blue 25	Langmuir	Pseudo-second-order	[129]
Banana peels	Methylene Blue	Freundlich	–	[130]
Banana peels	Methyl Orange	Freundlich	–	[130]
Banana peels	Rhodamine B	Freundlich	–	[130]
Banana peels	Congo Red	Freundlich	–	[130]
Banana peels	Methyl violet	Freundlich	–	[130]
Banana peels	Amido Black 10B	Freundlich	–	[130]
Orange peels	Methylene Blue	Langmuir	–	[130]
Orange peels	Methyl Orange	Langmuir	–	[130]
Orange peels	Rhodamine B	Langmuir	–	[130]
Orange peels	Congo Red	Langmuir	–	[130]
Orange peels	Methyl violet	Langmuir	–	[130]
Orange peels	Amido Black 10B	Langmuir	–	[130]
Garlic peels	Methylene Blue	Freundlich	Pseudo-second-order	[131]
Yellow passion fruit waste	Methylene Blue	Langmuir	Pseudo-first-order	[132]
Wheat shells	Methylene Blue	Langmuir	Pseudo-second-order	[133]
Cedar sawdust	Methylene Blue	Langmuir	Pseudo-second-order	[134]
Crushed brick	Methylene Blue	Langmuir	Pseudo-second-order	[134]
Meranti sawdust	Methylene Blue	Langmuir	Pseudo-second-order	[135]
Mansonia wood sawdust	Methylene Blue	Langmuir	–	[136]
Beech sawdust	Methylene Blue	Freundlich	Pseudo-second-order	[137]
Rice husk	Methylene Blue	Langmuir	Pseudo-second-order	[138]

Table 7 (Continued)

Table 7

Adsorbent	Dye	Fitted isotherm model	Fitted kinetic model	References
Papaya seeds	Methylene Blue	Langmuir	Pseudo-second-order	[139]
Grass waste	Methylene Blue	Langmuir	Pseudo-second-order	[140]
Paspalum notatum	Methylene Blue	Langmuir	Pseudo-second-order	[141]
Pomelo peels	Methylene Blue	Langmuir	Pseudo-second-order	[142]
Guava leaf powder	Methylene Blue	Langmuir	Pseudo-second-order	[143]
Jackfruit peels	Methylene Blue	Langmuir	Pseudo-second-order	[144]
Banana stalk waste	Methylene Blue	Langmuir	Pseudo-second-order	[145]
Palm kernel fibre	Methylene Blue	Langmuir	Pseudo-second-order	[146]
Broad bean peels	Methylene Blue	Langmuir	Pseudo-first-order	[147]
Castor seed shell	Methylene Blue	Multi-layer adsorption model	Pseudo-first-order	[149]
Pumpkin seed hull	Methylene Blue	Langmuir	Pseudo-second-order	[150]
Pineapple stem	Methylene Blue	Langmuir	Pseudo-second-order	[151]
Coconut husk	Methylene Blue	Langmuir	Pseudo-second-order	[152]
Coffee husks	Methylene Blue	Langmuir	Pseudo-second-order	[153]
Fallen Phoenix tree's leaf	Methylene Blue	Langmuir	Pseudo-second-order	[154]
Hazelnut shells	Methylene Blue	Langmuir	Pseudo-second-order	[155]
Tamarind shell	Congo Red	Langmuir, Freundlich	Pseudo-first-order	[235]
Azadirachta indica leaf	Congo Red	Langmuir	Pseudo-second-order	[236]
Tree fern	Acid Yellow 132	Langmuir	Pseudo-second-order	[238]
Pine sawdust	Acid Blue 25	Langmuir	Pseudo-second-order	[238]
Peanut hull	Methylene Blue	Langmuir	Pseudo-first-order	[239]
Peanut hull	Brilliant Cresyl Blue	Freundlich	Pseudo-first-order	[239]
Peanut hull	Neutral Red	Langmuir	Pseudo-second-order	[239]
Coir pith	Acid Violet	Langmuir	Pseudo-second-order	[240]
Coir pith	Rhodamine B	Langmuir	Pseudo-second-order	[240]
Coir pith	Methylene Blue	Langmuir	Pseudo-second-order	[240]
Coir pith	Acid Brilliant Blue	Langmuir	Pseudo-second-order	[240]
Rice husk	Methylene Blue	Langmuir	Pseudo-second-order	[138]
Rice husk	Safranin	Langmuir	Pseudo-second-order	[242]
Orange peels	Malachite Green	Langmuir	Pseudo-second-order	[243]
Jujuba seeds	Congo Red	Langmuir	Pseudo-first-order	[133]

at the solid–liquid interface during adsorption, while a negative value of $\Delta S°$ indicates a decrease in irregularity upon the adsorption [99–101]. Table 8 provides a summary of thermodynamic studies performed on the adsorption of dyes on agricultural waste materials.

AWM, such as papaya seeds, grass waste, pomelo peels, guava leaf powder, jackfruit peels, banana stalk waste, palm kernel fibre, teak tree bark powder, tree fern, pine sawdust has been used by many researchers in dye remediation of contaminated water [139,140,142–156,237,238]. Hameed [139] used papaya seeds for the adsorption of MB (555.56 mg/g) present in wastewater at the equilibrium time of 180 min. The effect of increasing the initial dye concentration on dye adsorption showed positive effects, and the dye adsorption followed the Langmuir isotherm and pseudo-second-order kinetic models. The adsorption of MB on grass waste achieved an adsorption capacity of up to 457.64 mg/g [140]. In another study by Hameed et al., pomelo peels were used for the adsorption of MB (adsorption capacity: 344.83 mg/g at pH 8.0 and 303 K). The Langmuir isotherm best described the process of adsorption [142]. Guava leaf powder [143], jackfruit peels [144], banana stalk waste [145] and palm kernel fibre [146] as adsorbents achieved the maximum removal capacities of 295.04, 285.71, 243.90 and 233.41 mg/g, corresponding to the contact time of 20, 180, 330 and 120 min, respectively in the removal of MB dye. Teak tree bark powder was also used for the adsorption of MB dye, which showed 333.33 mg/g adsorption capacity within 60 min [237]. Tree fern and pine sawdust exhibited a maximum adsorption capacity of 280.30 and 398.80 mg/g for Acid Yellow 132 and Acid Blue 25 adsorption in a contact time of 180 min, respectively [238]. The adsorptive behavior of MG on orange peels showed an adsorption capacity of 483.63 mg/g at a contact time of 24 h and 305 K [243]. According to the listed results, AWM, in addition to their many advantages, competes with activated carbon and nanocomposites in their effectiveness in removing the dyes by the adsorption process.

Previously, conventional treatment processes (such as biological, coagulation/flocculation, and advanced physico-chemical processes) were used to treat dyes in wastewater. Biological processes were used to remove dyes using

Table 8
Effect of temperature and thermodynamic parameters on some dyes removal by agricultural waste materials

Adsorbent	Dye	Effect of temperature	$\Delta G°$ (kJ mol^{-1})	$\Delta H°$ (kJ mol^{-1})	$\Delta S°$ (J mol^{-1} K^{-1})	References
Activated carbon	Indigo Blue	Negative	–4.21 to –4.86	–8.40	–11.74	[227]
Mango bark	Malachite Green	Positive	–21.45	20.21	–150	[126]
Neem bark	Malachite Green	Positive	–21.45	20.21	180	[127]
Sunflower seed husk	Methylene Blue	Positive	–	3.27	18.3	[128]
Wheat shells	Methylene Blue	Negative	–22.64	–24.11	–26.35	[133]
Cedar sawdust	Methylene Blue	Negative	–	–	–	[134]
Crushed brick	Methylene Blue	Negative	–	–	–	[134]
Meranti sawdust	Methylene Blue	Positive	–6.99	86.62	308	[136]
Guava leaf powder	Methylene Blue	Positive	–29,254	33.20	192.966	[143]
Palm kernel fibre	Methylene Blue	Positive	–13.99	37.61	172.564	[146]
Coffee husks	Methylene Blue		–6.55	17.69	–3.1	[153]
Fallen Phoenix tree's leaf	Methylene Blue	Negative	–4.62	7.77	–40	[154]
Hazelnut shells	Methylene Blue	Positive	–	—	–	[155]
Azadirachta indica leaf	Congo Red	Negative	–22.17	–218.70	–56.32	[236]
Almond shells	Crystal Violet	Positive	–0.35 to –0.68	3.59	13.23	[262]
Banana peels	Acid Blue 25	Positive	–14.66 to –14.74	–12.10	–8.02 × 10^3	[263]
Durian peel	Acid Blue 25	Positive	–13.62 to –13.64	–13.40	–0.82 × 10^3	[263]
Anchote peel	Methyl Orange	Positive	–8.21 to –9.05	–14.14	–27.90 × 10^3	[264]

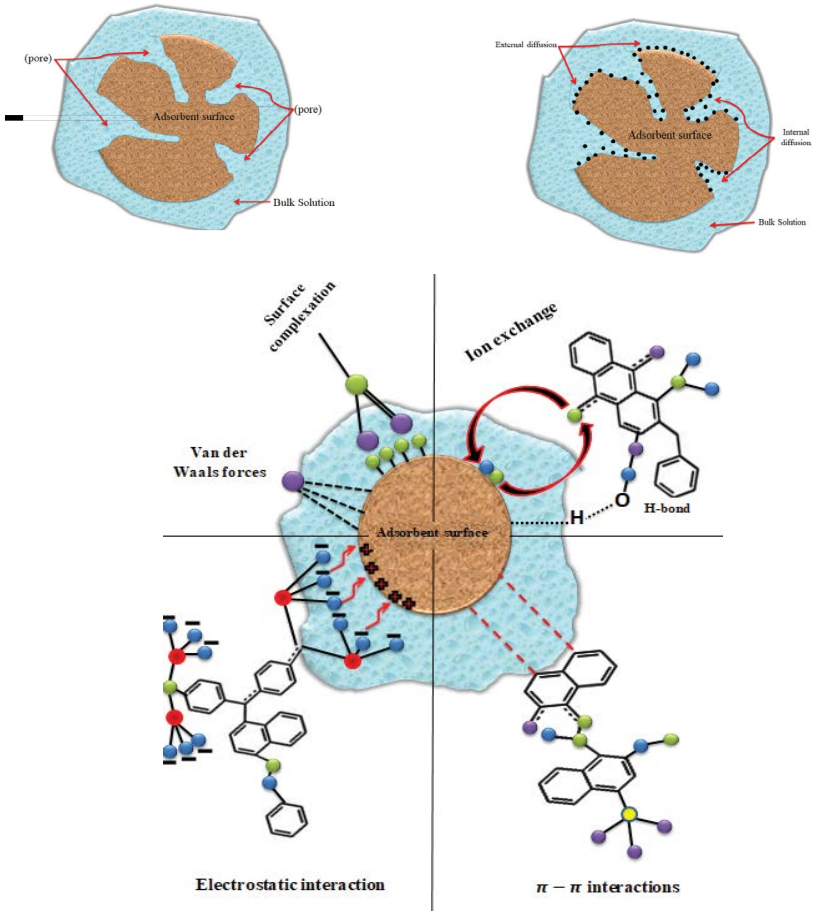


Fig. 3. Adsorption process and mechanism for the dye removal [89–94].

aerobic and anaerobic or facultative bacteria. The special conditions of use of biological processes and coagulants required to destabilize charged and suspended colloidal impurities are among the most important reasons for their lack of widespread use. The advanced physico-chemical processes include adsorption, filtration, photocatalysis, Fenton reaction, ozonation, UV/H_2O_2, and anodic oxidation. Adsorption is one of the widely used physio-chemical processes in dye waste treatment. Although removal efficiencies can reach over 90%, pore blockages and film fouling during this process are still the major drawbacks. However, the adsorption process is beneficial for wastewater treatment because of its operation ease, low cost, high efficiency, possibility of recycling of adsorbents, and suitability for the treatment of dye compounds. In general, when comparing previous studies, it is evident that adsorption is the most studied process for dye removal.

4.4. Adsorption mechanisms

Adsorption of dyes on the surface of the adsorbent can occur in two ways depending on the interaction between the solid surface and the adsorbent molecule (physical and chemical adsorption). There are several mechanisms of adsorption which may work in conjunction with each other (Fig. 3).

4.4.1. Physical adsorption mechanisms

Some factors such as adsorbent surface area, porosity, and aromaticity affect physical adsorption. Adsorption of dyes onto AWM surfaces mainly occurs through physical interactions such as van der Waals forces (intermolecular gravity), electrostatic interactions, pore filling, π–π interactions, hydrophobic interactions, surface diffusion, intraparticle pore diffusion, and hydrogen bonding [265–274].

4.4.2. Chemical adsorption mechanisms

Removing dyes from wastewater via AWM follows numerous chemical mechanisms such as surface complexation, ion-exchange, and precipitation [34,88,89,275–278]. The surface complexation concept describes the interfacial equilibrium caused by specific chemical reactions between the bulk species and active sites (functional groups) on the surface of the adsorbent. They can explain the effects of changing chemical conditions, such as pH, on adsorption. It is also concerned with specific types of chemical reactions and defines equilibrium constants, mass balances, and charge balances [277,278].

The removal of dyes by adsorption on the AWM surface may occur via exchange of dye ions/or the replacement of functional groups present on the adsorbent surface. In other words, the ion-exchange mechanism involves anion or cation exchange between the dye solution and the surface of the adsorbent [277,278].

5. Conclusion

This review article presents a wide range of agricultural waste materials, as cost-effective adsorbent. These low-cost bio-sorbents are recommended since they are relatively cheap or of no cost, good efficiency, high adsorption capacity, minimum energy consumption, and simple maintenance compared to more expensive adsorbents.

Parameters of optimal values such as pH, temperature, initial dye concentration and adsorbent dose, reported in previous studies, are discussed. These factors determine to some extent, the adsorption mechanism. According to the study results related to the isotherms and kinetics of the adsorption process, agricultural wastes often follow Langmuir and Freundlich isotherms and pseudo-second-order kinetic models. It was also found that the adsorption process is usually suitable, spontaneous, and either exothermic or endothermic based on thermodynamic parameters. The biosorbent process requires further research in modelling, biosorbent regeneration and waste material stabilization to enhance efficiency and recovery. At the end of this review, we recommend that more attention be paid by researchers to the dye removal procedure from real industrial effluents. This can be achieved by applying models to reach the optimal conditions for the removal process, then conducting the process continuously within separation columns to market it commercially, and finally renewing the adsorbent materials using one of the techniques used in this field, such as making a change in the pH, adding water, adding acidic or alkaline substance, or adding an organic substance. It is also possible to increase the effectiveness of these materials by modifying them in an acidic or basic medium or through the synthesis of composites to direct adsorption towards selective removal.

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Symbols and abbreviations

AWM	—	Agricultural waste materials
B	—	Constant related to heat of sorption, J/mol
b	—	Temkin isotherm constant
BB	—	Blue base
BOD	—	Biochemical oxygen demand
C_0	—	Initial concentration of dye, mg/L
CI	—	Color index
C_e	—	Equilibrium concentration of dye, mg/L
COD	—	Chemical oxygen demand
CR	—	Congo Red
CV	—	Crystal Violet
DG	—	Direct Green
DR	—	Direct Red
EDX	—	Energy-dispersive X-ray spectroscopy
FTIR	—	Fourier-transform infrared spectroscopy
I	—	Boundary layer diffusion effects (external film resistance), mg/g
K	—	Distribution constant or equilibrium constant, L/mg
k_1	—	Pseudo-first-order rate constant, 1/min
k_2	—	Pseudo-second-order rate constant, g/mg·min

k_{id}	—	Intraparticle diffusion rate constant, mg/g·min$^{0.5}$
K_F	—	Freundlich adsorption capacity, L/mg
K_L	—	Langmuir equilibrium constant, L/mg
K_T	—	Temkin constant related to the adsorption heat, L/mg
m	—	Mass of adsorbent, g
MB	—	Methylene Blue
MG	—	Malachite Green
MO	—	Methyl Orange
n	—	Freundlich adsorption intensity
OR	—	Original Red
PFO	—	Pseudo-first-order
PSO	—	Pseudo-second-order
q_e	—	Amount of dye adsorbed per gram adsorbent at equilibrium, mg/g
q_t	—	Amount of dye adsorbed per gram adsorbent at time t, mg/g
q_{max}	—	Maximum amount of dye adsorbed per gram adsorbent, mg/g
R	—	Gas constant, 8.314 J/mol·K
RB	—	Rhodamine B
RB5	—	Reactive Black 5
R_L	—	Dimensionless separation factor
R^2	—	Correlation coefficient
SEM	—	Scanning electron microscopy
t	—	Time, min
T	—	Temperature, K
V	—	Volume of the aqueous solution, L
YB	—	Yellow Base 28
α	—	Elovich initial adsorption rate, mg/g·min
β	—	Elovich desorption constant, g/mg
$\Delta G°$	—	Change in Gibbs free energy, kJ/mol
$\Delta H°$	—	Change in enthalpy, kJ/mol
$\Delta S°$	—	Change in entropy, kJ/mol·K

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