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Dye adsorption using biomass wastes and natural adsorbents: overview and future prospects

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ABSTRACT

The increase in number of modern industry in the twenty-first century that uses dye and ends up disposing it into the environment and water bodies causing diseases and other environmental risks to both aquatic life and human being is alarming. In response to the increasingly stringent discharge standards and discharge fee, textile industries must upgrade the performance of their wastewater treatment facilities with primary effort on reducing chemical oxygen demand and color. This paper presents a review of dye adsorption using activated carbon prepared from different sources and environmental implications. Also, its major challenges together with future prospects are summarized and discussed. Conclusively, the production of activated carbon from waste biomass for adsorption of these dyes is of great importance in pollution control and environmental conservation.

Keywords: Adsorption; Dyes; Activated carbon; Wastewater

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1. Introduction

Concern about environmental protection has increased over the years from global view point. In the past decades, the exponential population and social civilization growth changes in the productivity and consumption habits, increasingly affluent lifestyles, resource use, and continuing development of industrial technologies has been accompanied by the rapid generation of municipal and industrial solid wastes, which create the most intransigent paradox around the world [1–4]. The conventional method for the removal of toxic compound includes adsorption, precipitation, coagulation, flocculation, ion-exchange, reverse osmosis, complexation, sedimentation, electrochemical operation, and biological treatment [5]. Activated carbon, an adsorbent with large porous surface area, controllable pore structure, thermo-stability, and low acid/base reactivity [6], is currently receiving great attention owing to its superior and efficient ability in air pollution control [7], solvent recovery [8], wastewater treatment (dye, heavy metals, detergents, herbicides, pesticides, and polyaromatic hydrocarbons) [9–45], as well as of odor and taste [46]. As the demand for environmental protection increases every year, activated carbons are widely used in the

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industry for purification, separation, and recovery processes. Adsorption using activated carbon has been widely used because of its simplicity, low cost, and high removal efficiency which has been used for elimination of organic substances, or heavy metal ions from water and wastewater [4,47].

2. Adsorption as a process

Porous or finely divided solids can hold more adsorbate because of the relatively large exposed surface area. The adsorbent surface of a liquid is increased if the liquid is divided into fine droplets. In some cases, the atom of the adsorbate shares electrons with atoms of the adsorbent surface, forming a thin layer of chemical compound. Adsorbent is also an important part of catalysis and other chemical processes. Adsorption has been regarded as superior to other methods [48]. Adsorption used mostly in industries for the removal of waste depends on numerous factors [49]. Dyes are a kind of organic compounds with complex aromatic molecular structure that gave bright and firm color to other substances. However, the complex aromatic molecular structure of dyes makes them more stable and more difficult to biodegrade. The extensive use of dyes often poses pollution problems in the form of colored wastewater discharged into environmental water bodies [50,51]. Dyes are colored compounds suitable for coloring textiles, wools, and fibers; natural dyes such as indigo have been used for over 5,000 years [52]. There are more than 10,000 dyes with different chemical structures available commercially [53]. Dyes are classified into different classes: (a) anionic, direct, acid, and reactive dyes, (b) cationic: all basic dyes, and (c) nonionic, disperse dyes [54]. There are also synthetic and natural dyes but the synthetic dyes have replaced natural dyes because of their low cost and vast range of new colors [55]. Today, over 9,000 types of synthetic dyes have been incorporated. Discharge of dye from various industries such as textile, food processing, and ink production imposes threat to the environment and ecosystem resulting to toxicity in human and aquatic organisms. It also blocks sunlight penetration thereby inhibiting photosynthesis.

Table 1
Adsorption capacities of treated agricultural wastes for dye removal

Plant wastes	Dyes	Activation process	Equilibrium time	q_e (mg g^{-1})	Refs.
Banana bark	Rhodamine B	Carbonized by conc. H_2SO_4 and activated by heating at 600˚C	40 min	40.161	[57]
Rice husk	Direct F. Scarlet	Activated by 0.6 M citric acid for 200˚C	90 min	4.35	[58]
Mangrove bark	Direct red-23	Chemically treated with a mixture of 37% HCHO + 0.2 N H_2SO_4	4 h	21.55	[59]
Pandanus leaves	Malchite green	Carbonized by conc. H_2SO_4 and activated by heating at 400˚C for 12hrs	40 min	9.737	[60]
P. oceanica leaf sheath	Reactive red 23	Chemically treated with a mixture of 0.2 HNO_3	10 h	0.31	[61]
Rice husk	MB	Ash is formed by heating at 500˚C for 2 h	30 min	690	[62]
Tapioca peel	Red brown C4R	Activated by conc. H_2SO_4 and carbonized at 200˚C	45 min	121.47	[63]
Bagasse	Acid violet 17	Charcoal prepared by burning in open air	4 h	38.32	[64]
Groundnut shell	Acid violet 17	Charcoal prepared by burning in open air	100.57		[64]
Pea shell	Acid red 119	Charcoal prepared by burning in open air	4 h	44.48	[64]
Used tea leaves	Acid blue15	Charcoal prepared by burning in open air	4 h	126.53	[64]
Wheat straw	Acid red 119	Charcoal prepared by burning in open air	72.81		[64]
Rice husk	Malachite green	Carbonized in quartz tube in muffle furnace at 450ċ for 1 h & activated at 650ċ for 2 h.	40 min	63.85	[65]

Table 2
Adsorption capacities of raw agricultural waste for dye removal

Adsorbents	Dye	Equilibrium time	Temperature (°C)	q_e (mg g^{-1})	Refs.
Neem bark	Malchite green	7 h	25	0.36	[66]
Mango bark	Malchite green	7 h	25	0.53	[66]
Tamarind shell	CR	4 h	30 ± 1	10.48	[67]
Neem leaf powder	CR	5 h	27	72	[68]
Grape fruit peel	Reactive blue 19	45 min	25	12.53	[69]
Teak tree bark	MB	30 min	30	333.3	[70]
Wheat straw	Basic yellow 21	48 h	20 ± 2	71.43	[71]
Sunflower seed husk	MB	4 h	25 ± 2	45.25	[72]
Hazlenut shell	MB	60 min	20	76.9	[73]
Hazlenut shell	Acid blue	60 min	20	60.2	[73]
Cherry saw dust	MB	2 h	20	39	[73]
Walnut saw dust	MB	60 min	20	59.17	[73]
Oak saw dust	Acid blue	60 min	20	29.5	[73]
Pitch pine saw dust	Acid blue	60 min	20	27.5	[73]

The comparison between the raw and treated adsorbents is summarized briefly (Tables 1 and 2) [56]. From the literature reviewed, it is demonstrated that chemically treated agricultural waste showed comparatively significant removal efficiency than the raw agricultural waste. Decolorisation process is not specific and depends upon many factors. Although there are lots of agricultural adsorbents which can act as a substitute for the expensive commercial activated carbon, complete replacement is not possible. The factors which favor the selection of agricultural adsorbents are its low cost, widespread presence, and organic composition which shows strong affinity for some selected dyes.

3. Adsorbent

This is the surface to which an adsorbate adheres to. Adsorbents are usually in the form of spherical pellets, rods, molding, or monoliths with hydrodynamic diameters between 0.5 and 10 mm. It has high abrasion resistance, high thermal stability, and small pore diameter. Adsorbents mostly used are the agricultural waste. It is formed from the pyrolysis and carbonization of substrate so as to increase pore formation for adsorption. Activated carbon is the most common adsorbent due to its effectiveness, versatility, and good capacity. The preparation of activated carbon involves two stages, mainly pyrolysis and activation (chemical and physical activation) [74]. In the first stage, suitable carbon precursors are pyrolyzed under an inert or reducing gas at a moderate temperature (about 600–800°C) to release the volatile matter and produce chars with rudimentary pore structures [75]. Thereafter, the resulting chars are subjected to partial gasification at a higher temperature (usually above 90°C) with oxidizing agent such as CO_2 or steam or a mixture of both to produce activated carbon with well-developed and accessible internal porosities [76], while in chemical activation, a raw material is impregnated with an activating agent, this material is treated under inert atmosphere [77]. Chemical activation is often used to produce activated carbon from biomass. It involves pyrolyzing the feed stock in the presence of a chemical activating agent such as H_3PO_4, $ZnCl_2$, KOH, etc. Among the activation agents, H_3PO_4 offers several advantages [78,79]. The carbonization step and activation step are carried out simultaneously, with the precursor being mixed with chemical activating agents, or dehydrating agents and oxidants [79]. A very high temperature is invariably achieved by connective and conductive heating of the sample placed in a convectional heating such as a tubular furnace.

Activated carbon is advantageous for the adsorption of dyes; mainly its large porous surface area, controllable pore structure, thermo-stability, and low acid/base reactivity have been established in terms of its versatility for removal of different types of dyes dissolved in aqueous media [80,81]. The pore size of the activated carbon enhances the uptake of the dyes. This is a distinct property. The pore size distribution and the relative size of the adsorbate molecules have a significant effect on adsorption capacity [82]. The chemical properties of the dye and the activated carbon are also very important for the adsorption process because there has to be some interaction between the adsorbate and adsorbent. If the dye combines well with activated carbon, it will enhance the uptake of dye .The uptake of dyes onto the activated carbon is fostered as a result of the interaction between the functional groups of the dye and activated carbon. Adsorption greatly depends on the porous structure and surface functional groups of the activated carbon [83].

It is very difficult to understand which adsorbent is better because they have different properties (porosity, surface area, and physical strength) as well as different adsorption capacities related to experimental conditions [74]. Adsorbent cost is an important parameter to compare different materials. In Fig. 1, costs of several low-cost and commercial adsorbents are shown. They should be considered indicative because adsorbent costs depend on many factors such as its availability, its source (natural, industrial/agricultural/ domestic wastes or by-products or synthesized products), treatment conditions, recycling and lifetime issues. Furthermore, the cost also depends on where adsorbents are produced in (or for) developed, developing, or underdeveloped countries [84]. Finally, a right cost evaluation is related to the application scale, although many studies about nonconventional low-cost adsorbents are available in the literature, they are limited to laboratory scale. Thus, cost estimation is not strictly right and pilot-plant studies should also be conducted utilizing low-cost adsorbents to check their feasibility on commercial scale.

4. Alternatives to activated carbon

Activated carbon is a popular and effective dye sorbent, but its relatively high price, high operating costs, and problems with regeneration hamper its large-scale applications. Therefore, there is a growing need in finding low-cost, renewable, locally available materials as sorbent for the removal of dye colors [94–97]. Locally available materials and some low-cost agricultural waste materials are: nut shell [98], pecan shell [99], apple peal [90], coffee grounds [98–101], waste tea [102], coconut shell [103,104], apple pomace, wheat straw [105], orange peel [106], cellulose-based wastes [107], banana peel [108], maize stalk [109], wheat straw [110], corn cobs [110], barley husk [110], rice husk [6,111–119], peanut hull [120], wood chip [121], palm fruit bunch [122,123] sawdust [124], bark [125], leaf [126], coir pith [127], banana pith [128,129], bagasse pith [130], and aquatic plants [131,132]. Apple peal [133], cocoa pod husk [134,135], banana stalk [136], groundnut hull [137], rice hull ash [138], *Azadirachta indica* leaf powder [139], palm kernel fibre [140,141], fungi [142,143], bentonite [144–147], organo-attapulgite [148], olive seed and stone [149], bio-gas residual slurry [150], coffee beans [151], petroleum coke [152], date pits [153], and corn cob [154], all with a high carbon content have been used as raw material for the preparation of activated carbon and directly been used as sorbent for dye adsorption from wastewater. Adsorption processes provide a feasible treatment especially if the adsorbent is inexpensive and readily available [155]. Among various adsorbents used, commercially available activated carbon is the most commonly used adsorbent [156,157]. The use of activated carbon has been highlighted as an effective technique for dye removal. Due to its unique molecular structure, activated carbon has an extremely high affinity for many classes of dyes including basic dyes [158].

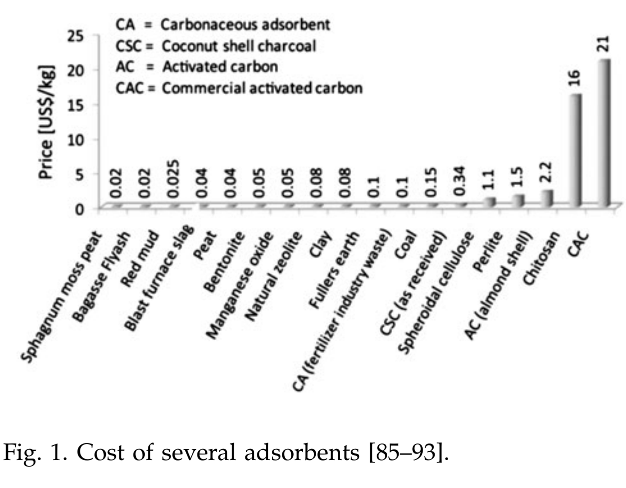

Fig. 1. Cost of several adsorbents [85–93].

5. Biomass waste materials and natural adsorbents used for dye removal

Although, activated carbon is undoubtedly considered as universal adsorbent for the removal of diverse kinds of pollutants from water, its widespread use is sometimes restricted due to the high costs [159,160]. Attempts have been made to develop low-cost alternative adsorbents which may be classified in two ways (Figs. 2 and 3): either (i) on basis of their availability, i.e. (a) natural materials (wood, peat, coal, lignite, etc.), (b) industrial/agricultural/domestic wastes or by-products (fly ash, bagasse, pearl, millet husk, bamboo, *Cunninghamella elegans* flyash, etc.), and (c) synthesized products or (ii) depending on their nature, i.e. (a) inorganic and (b) organic material [85,159,161,162].

Wastewaters contaminated by dyes represent a relevant issue associated with several industries. Dyes, even at very low concentrations, reduce wastewater transparency and oxygen solubility and are often toxic and recalcitrant; moreover, these chemicals are toxic, carcinogenic or mutagenic for various organisms [163].

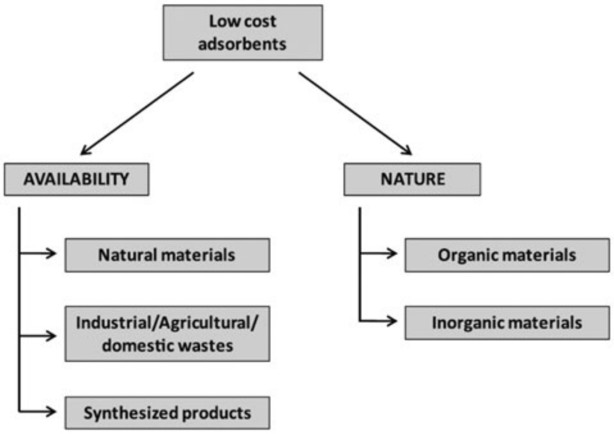

Fig. 2. Possible classification of low-cost adsorbents [159].

Some of the biomass waste materials and natural adsorbents used for dye removal are reviewed under the following headings.

6. Lignocellulosic biomass

6.1. Peanut hulls

Gong and coworkers [164] investigated the roles played by three major functional groups (amino, carboxyl, and hydroxyl groups) in the biomass of peanut hull in the adsorption of six dyes. These functional groups in peanut hull were chemically modified individually to determine their contribution to the adsorption of ionic dyes. The dyes used were methylene blue (MB), brilliant cresyl blue, neutral red, amaranth, sunset yellow, and fast green. It was found that carboxyl group inhibited the adsorption of anionic dyes but it was a major functional group in the adsorption of cationic dyes; hydroxyl group was one important functional group in the adsorption of the six dyes studied. The effect of methylation of amino groups was not significant on the adsorption of six dyes. Studies on the effects of chemical modification on dye adsorption on sorbent derived from peanut hull suggested that the effect of methylation of amino group on ionic dye adsorption was not significant. The possible reason was that at ambient temperature, the methylation could not be carried out completely due to the tough cell wall of peanut hull. The carboxyl group inhibited the adsorption of anionic dyes because of its negative charge thereby removing the negative charge of carboxyl group by esterification; anionic dye uptake capacity was obviously increased. The carboxyl group was the major functional group in the adsorption of cationic dyes. The hydroxyl group was an important functional group in the adsorption of cationic and anionic dyes [165].

6.2. Coconut husk

Hameed and coworkers determined from batch tests the adsorption isotherm and kinetics of MB on activated carbon prepared from coconut husk [166–168].The effects of contact time (1–30 h), initial dye concentration (50–500 mg L^{-1}), and solution temperature (30–50˚C) were investigated. Equilibrium data were fitted to Langmuir, Freundlich, Temkin and Dubinin–Radushkevich isotherm models. The equilibrium data were best represented by Langmuir isotherm model, showing maximum monolayer adsorption capacity of 434.78 mg g^{-1}. The kinetic data were fitted to pseudo-first-order, pseudo-second-order, and intraparticle diffusion models and were found to follow closely the pseudo-second-order kinetic model. The adsorption interaction was found to be exothermic in nature. Coconut husk-based activated carbon was

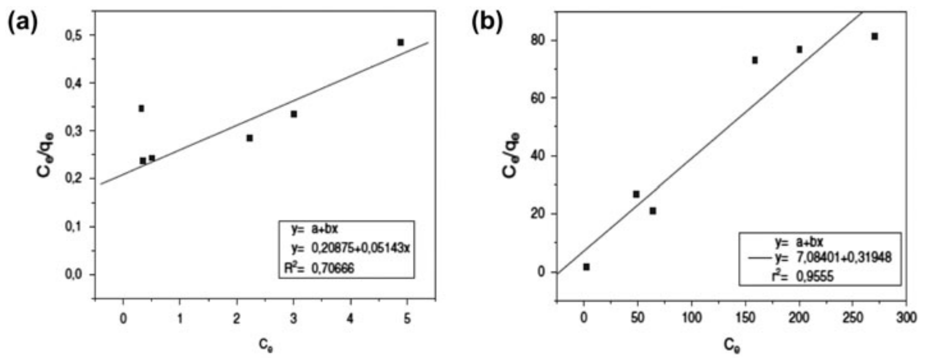

Fig. 3. Langmuir isotherms for adsorption of the dyes (a) RO 16 and (b) RR 120 by humin; at different concentrations (10, 20, 30, and 50 mg L^{-1}), pH 1.0, and using a humin mass of 0.2 g [191].

shown to be a promising adsorbent for removal of MB from aqueous solution. The surface area of the prepared activated carbon was relatively high with large pore volume and was found to be mesoporous in nature. MB was found to adsorb strongly on the surface of the activated carbon. The maximum monolayer adsorption capacity decreased with increasing temperature. The negative ΔH° value confirmed the exothermic nature of the adsorption interaction whereas the positive ΔS° value showed the increased randomness at the solid–solution interface during the adsorption process. The negative value of ΔG° indicated the feasibility and the spontaneous nature of the adsorption of MB onto the prepared activated carbon. The adsorption performance of the coconut husk-based activated carbon was comparable to those from the commercially available activated carbon [166–168].

6.3. Palm ash

The removal of acid green 25 (AG25) dye onto activated palm ash from aqueous solutions was carried out by Hameed and coworker [169]. Experiments were carried out as a function of contact time, initial dye concentration (50–600 mg L^{-1}), pH (2–12), and temperature (30–50 °C). The equilibrium adsorption data of AG 25 dye on activated palm ash were analyzed by Langmuir and Freundlich models. Results indicated that the Freundlich model provides the best correlation for the experimental data. The adsorption capacities of activated palm ash for removal of AG 25 dye was determined using Langmuir equation and found to be 123.4, 156.3, and 181.8 mg g^{-1} at 30, 40, and 50 °C, respectively. Adsorption data were modeled using the pseudo-first-order, pseudo-second-order, and intra-particle diffusion kinetics equations. It was shown that pseudo-second-order kinetic equation best described the adsorption kinetics. The positive value of the enthalpy change (26.64 kJ mol^{-1}) indicates that the adsorption is an endothermic process. Their results indicated that activated palm ash is suitable as adsorbent material for adsorption of AG25 dye from aqueous solutions [122,123,140,141,169].

6.4. Cashew nut shell

Cashew nut shell (CNS)—a novel, low-cost adsorbent prepared from agricultural waste [170–178] was used as an adsorbent for the removal of congo red (CR) dye from aqueous solution. The effect of pH, adsorbent dose, initial dye concentration, time, and temperature on the adsorption was studied. Results indicated that CNS can be employed as a low-cost alternative compared to other commercial adsorbents in the removal of dyes from wastewater. Thermodynamic parameters such as ΔG°, ΔH°, and ΔS° were evaluated and it was found that the sorption process was feasible, spontaneous, and exothermic in nature. Pseudo-first-order, pseudo-second-order, and intraparticle diffusion models were used to fit the experimental data. It was shown that the adsorption of CR dye could be described by the pseudo-second-order equation, suggesting that the adsorption process is presumably governed by chemisorption. The equilibrium data were analyzed using Langmuir, Freundlich, Redlich–Peterson, Koble–Corrigan, Sips, Toth, Temkin, and Dubinin–Radushkevich isotherms. The characteristic parameters for each isotherm and related correlation coefficients were determined. The experimental data yielded excellent fits with the isotherms studies in this order: Redlich–Peterson > Toth, Koble–Corrigan, Sips, Freundlich, Langmuir, Temkin, Dubinin–Radushkevich, based on its correlation coefficient values. Thermodynamic analysis indicates that the system is spontaneous and exothermic [170].

7. Synthetic adsorbents

7.1. Chitosan beads

Chiou and coworkers [179] used ionically and chemically crossed-linked chitosan beads to prepare adsorbent with high adsorption capacity for anionic dyes. A batch system was applied to study the adsorption of four reactive dyes (RB2, RR2, RY2 and RY86), three acid dyes (AO12, AR14, AO7) and one direct dye (DR81) from aqueous solutions by the cross-linked chitosan beads. The adsorption capacities had very large values of 1,911–2,498 (g kg^{-1}) at pH 3–4, at a temperature of 30 °C which were 3.4–15.0 and 2.7–27.4 times those of the commercial activated carbon and chitin, respectively. The Langmuir and Freundlich adsorption models were applied to describe the equilibrium isotherms. The Langmuir model agreed very well with experimental data (R^2 > 0.9893). The kinetics of adsorption, the ADMI color value, and decolorization effciency for different initial dye concentrations were evaluated by the pseudo-first-order and pseudo-second-order models. The data agreed very well with the pseudo-second-order kinetic model. The adsorption capacity increased largely with decreasing solution pH and adsorbent dosage. The free energy changes ΔG° for adsorption of anionic dyes in acidic solutions at 30 °C were evaluated. The negative values of ΔG° revealed that the overall adsorption processes are spontaneous. The cross-linked chitosan beads had very high adsorption affinity to remove the anionic dyes, whose maximum monolayer adsorption capacity ranges from 1,911 to 2,498 (g kg^{-1}) at 30˚C. The adsorption capacities are affected significantly by the dye initial concentration, pH, and adsorbent dosage. The uptake increases with increase in dye initial concentration, with decrease in pH, and adsorbent dosage. To compare with commercial activated carbon, the cross-linked chitosan exhibits excellent performance for adsorption of anionic dyes. The adsorption capacities of the cross-linked chitosan beads are much higher than those of chitin for anionic dyes. It shows that the major adsorption site of chitosan is an amine group, $–NH_2$, which is easily protonated to form $-NH_3^+$ ion in acidic solutions. The strong electrostatic interaction between the $-NH_3^+$ of chitosan and dye anions can be used to explain the high adsorption capacity of anionic dyes onto chemically cross-linked chitosan beads. The Langmuir equation agrees very well with the equilibrium isotherm. The pseudo-second-order kinetic model fitted the adsorption data most [179–181].

7.2. Cross-linked amphoteric starch

Xua and coworkers [182] investigated cross-linked amphoteric starch with carboxymethyl and quaternary ammonium groups as an adsorbent for removal of both acid and basic dyes in solution. Acid light yellow 2G, acid red G, methyl green, and methyl violet were used to study the adsorption behaviors under various operational parameters such as pH, dose of amphoteric starches, initial dye concentration, adsorption time, and adsorption temperature. At the preset pH value, the relationship between the amount adsorbed and the initial dye concentration was given. They found out that this adsorbent can be applied on both acid and basic dyes. For acid dyes, the active site are the quaternary ammonium groups, while for basic dyes, they are carboxymethyl groups. Adsorption processes are exothermic for acid dyes, which shows that low temperature will facilitate their adsorption, while the basic dyes have the highest adsorption capacity at 303 K. Both Langmuir isotherm and Freundlich isotherm models fitted well at all concentrations studied. Kinetic study shows that the pseudo-second-order model fits the experimental data better [182].

8. Minerals adsorbents

8.1. Clay

The adsorption kinetics of a cationic dye, MB, onto clay from aqueous solution with respect to the initial dye concentration, temperature, pH, mixing rate, and adsorbent dosage were investigated. In order to understand the adsorption mechanism in detail, zeta potentials and the conductivities of clay suspensions at various pH (1–11) and cation exchange capacity (CEC) porosity and BET surface area of clay were measured. It was found that the amount of MB adsorbed increases with decreasing temperature, increasing sorbent dosage, and increasing initial dye concentration. Adsorption capacity decreases with increasing pH, except for the natural pH (5.6) of clay suspensions. The adsorption kinetics of MB was studied in terms of pseudo-first-order sorption, pseudo-second-order sorption, and intraparticle diffusion processes thus comparing chemical sorption and diffusion sorption processes. It was found that the pseudo-second-order mechanism is predominant and the overall rate of the dye adsorption process appears to be controlled by more than one step. Clay mineral crystals carry a charge arising from isomorphous substitutions of certain atoms in their structure for other atoms of different valence. In the tetrahedral sheet, Si (IV) may be replaced by trivalent cations, or divalent cations may replace Al (III) in the octahedral sheet. When this is the case, charged efficiency results and negative potential at the surface of the clay is created. The negative potential is compensated by the adsorption of cations on the surface. The total number of cations adsorbed on the clay is called the CEC [183,184]. In their findings, they found that the sorption tends to attain the equilibrium in about 60 min. Also, a comparison of the kinetic models on the overall adsorption rate showed that dye/clay system was best described by the pseudo-second-order rate mode. It was observed that the clay particles moved to the negative electrode when the adsorption of the organic cations was in excess of the CEC. Some researchers have discussed the sorption of organic cations on a negatively charged site that is already neutralized [185]. Organic cation sorption on neutral sites and neutral complexes results in a charge reversal of the surfaces, which in turn alters the sorption characteristics, and colloid behavior of the clays [186,187].

8.2. Kaolin

The adsorption of toxic brilliant green dye from aqueous medium using kaolin as an adsorbent has also been investigated [188]. Characterization of kaolin was done by measuring the particle size distribution using particle size analyzer, BET surface area using BET surface analyzer, and structural analysis using X-ray diffractometer. The effects of initial dye concentration, contact time, kaolin dose, stirring speed, pH, and temperature were studied for the adsorption of brilliant green in batch mode. Results from adsorption experiments revealed that the extent of adsorption is strongly dependent on pH of solution. Free energy of adsorption (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) changes were calculated to know the nature of adsorption. The calculated values of ΔG° at 299 and 323 K revealed that the adsorption process is spontaneous [188]. The estimated values of ΔH° and ΔS° both gave negative value, indicating that the adsorption process is exothermic and the dye molecules are organized on the kaolin surface in less randomly fashion than in solution. Adsorption kinetic was tested using pseudo-first-order, pseudo-second-order, and intraparticle-diffusion models. It was observed that the rate of dye adsorption follows pseudo-second-order model for the dye concentration range studied. Standard adsorption isotherms were used to fit the experimental equilibrium data. It was found that the adsorption of brilliant green on kaolin follows the Langmuir adsorption isotherm. Kaolin is identified to be an effective adsorbent for the removal of brilliant green dye from aqueous medium [188–190].

8.3. Humin

Jesus and coworkers [191] studied the optimum conditions for adsorption/desorption of reactive dyes using both batch experiments and fixed bed columns with humin immobilized on silica. Results obtained from batch experiments showed that adsorption of reactive orange 16 and Reactive Red 120 by humin was most effective at pH 1.0. Adsorption equilibrium was achieved rapidly for all initial dye concentrations employed. Temperature measurements indicated that the process was exothermic, and was best described by a pseudo-second order kinetic model for both dyes while Langmuir and Freundlich isotherms best described the adsorptions of RO 16 and RR 120 (Figs. 3 and 4), respectively.

Column experiments also showed adsorption efficiencies of 62.1–39.6% and 96.2–74.8%, and desorption efficiencies of 81.4–51.3% and 66.8–36.8% for RO 16 and RR 120, respectively, over three cycles (Table 3) [191]. The biosorbent gave excellent performance in the removal of reactive dyes, with the advantage that it can be regenerated and reused [191–193].

8.4. Sepiolite

Eren and coworkers [194] studied the temperature, pH, and ionic strength effects on the adsorption of crystal violet (CV) by raw, base–activated, and Al-saturated sepiolite samples. Al-saturated sepiolite has relatively weak ability for CV adsorption compared to the raw clay, whereas the highest adsorption observed for the base-activated sepiolite may be due to the Na^+ ions replacing part of Mg^{2+} located at the edges of the channels. Different thermodynamic parameters for raw and pre-treated sepiolite samples indicate the adsorption of the dye component on a nonuniform surface. The adsorption of dye in sepiolite suspensions is highly affected by the sepiolite surface feature. It was noticed that the amount of adsorbed CV cations on the base-activated sepiolite is 2.6-fold higher than raw sepiolite. Base activation leads to the replacement of part of Mg_2^{2+} located at the edges of the channels by Na^+; the reactivity of basal and edge surface groups is enhanced and consequently the adsorption capacity of base activated sample is increased. It was also observed that lowest adsorption capacity on the Al-saturated samples arising from the

Table 3
Efficiencies of retention and removal of the dyes RO 16 and RR 120 over three cycles, using columns of humin immobilized on silica [191]

	Reactive orange 16		Reactive red 120	
	Adsorption		Adsorption	
Cycle	Mass retained (mg)	Retention efficency (%)	Mass retained (mg)	Retention efficency (%)
1	31.3	62.1	45.2	96.2
2	25.1 + 5.8 = 30.9	46.3	32.0 + 15 = 47	75.6
3	20.2 + 13.1 = 33.1	39.6	41.7 + 21.3 = 63	74.8
	Desorption		Desorption	
	Mass removed (mg)	Removal efficency (%)	Mass removed (mg)	Removal efficency (%)
1	25.5	81.4	30.2	66.8
2	17.8	57.6	25.7	54.6
3	17.0	51.3	23.2	36.8

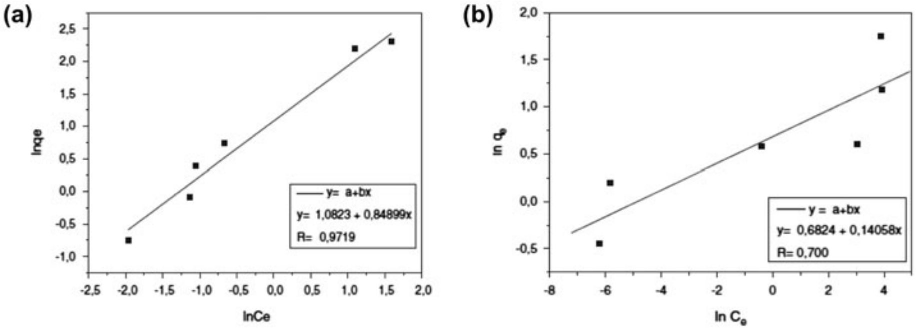

Fig. 4. Freundlich isotherms for adsorption of the dyes (a) RO 16 and (b) RR 120 by humin, at different concentrations (10, 20, 30, and 50 mg L^{-1}). Conditions: 0.2 g of humin; pH 1.0; $T = 25 \pm 0.2$℃ [191].

aluminium cation exchange process leads to pore blocking and the reactivity of basal or edge oxygen atoms of the silicate. Adsorption of the CV molecule on these surfaces is controlled by several mechanisms. Firstly, pH-independent adsorption, part of the adsorption is by an exchange mechanism releasing exchangeable cations in the interlayer and on basal plane surface and part of the adsorption is via noncoulombic interactions to one adsorbed cation and neutralized site. Secondly, amphoteric sites SiOH groups of sepiolite are responsible for a pH-dependent adsorption on the clay edges. The calculated adsorption enthalpy and entropies showed that clay surfaces are not uniform in nature towards the adsorption of CV. Conclusively, adsorption of cationic dyes on sepiolite can be considered as a simple, fast, and economic method for their removal from water and wastewater [194].

8.5. Clinoptilolite

Qiu and coworkers [195] studied the adsorption of dyes into Clinoptilolite by series of experiments. The influence of sorbent concentration, adsorption time, initial dye concentration, and pH was analyzed in detail with Amido Black 10B and Safranine T, respectively. Results indicated that clinoptilolite has a limited adsorption capacity for Amido Black 10B and has a good adsorption capacity for Safranine T. The adsorption isotherms were also determined using the adsorption data. It was found that the adsorption isotherms for Safranine T dye-natural zeolite system and Amido Black 10B dye-natural zeolite system at 80 mg L^{-1} of solid concentration, pH 7.0, and 2EC both fitted to the Langmuir isotherm. For the Safranine T dye natural zeolite system, the fitted parameters q_m and b_L are 0.05513 mg g^{-1} and 1.9623 mg L^{-1}, respectively, and for the Amido Black 10B dye-natural zeolite system, the fitted parameters quality matter, q_m and black light, b_L are 0.0112 mg g^{-1} and 0.5962 mg L^{-1}, respectively. The adsorption contact times are 80 and 360 min for the Safranine T dye-natural zeolite and Amido Black 10B dye-natural zeolite systems. Amido black 10B and Safranine T are adsorbed into natural zeolite with a removal efficiency of 81.2 and 16.3%, respectively, for 360 min of contact time at 4 mg L^{-1} initial dye concentration, 80 mg L^{-1} of solid concentration, pH 7.0, and 2EC. It was concluded that clinoptilolite has a limited adsorption capacity for Amido Black 10B. The amount of adsorption for Safranine T increases as pH increases, contrary to the trend observed with Amido Black 10B [195–198].

9. Activated carbon from lignocellulosic biomass

9.1. Durian peel

Another research group evaluated durian peel as raw material for the production of activated carbon used in adsorption of dyes [199]. Post-treatment of synthesized activated carbon with either acid or base solution was performed. Activated carbon synthesized under vacuum pyrolysis had better properties (BET surface area, pore volume, and adsorption capacities) than that under nitrogen atmospheric pyrolysis. Improvement in such properties was also observed when activated carbon was post-treated with HCl solution. The HCl-treated activated carbon best adsorbed Basic Green 4 dye compared to acid dyes. Adsorption of Basic Green 4 dye was well explained with pseudo-second-order kinetics. The experimental adsorption equilibrium data were fitted to Langmuir and Freundlich models, with the former providing the best fit. The maximum adsorption capacities of activated carbons synthesized under nitrogen atmosphere and vacuum pyrolysis were very high (303.0 and 312.5 mg g^{-1}, respectively). Vacuum pyrolysis gave activated carbon with greater BET surface area, pore volume, and adsorption capacities for Basic Green 4, acid blue 11, and direct blue 80 dyes than the one obtained from nitrogen atmospheric pyrolysis [199]. Post-treatment of activated carbon with HCl solution increased both BET surface area and pore volume of activated carbon in comparison to NaOH and H_2O treatment. This is due to the removal of impurities rather than the modification of activated carbon surface. BET surface area and pore volume differs from those synthesized from chemical activation with KOH. The effect of solution pH on the adsorption of dyes was observed, and such influence depended on type of dyes. The adsorption kinetic of Basic Green 4 was well described by the pseudo-second-order model. Adsorption isotherm was described by a monolayer Langmuir model. Durian peel is judged as a promising raw material in the production of activated carbon for effective removal of harmful basic dyes [199].

9.2. *Sawdust*

Sawdust [200] is one of the most appealing materials among agricultural waste materials, used for removing pollutants such as, dyes, salts, and heavy metals from water and wastewater. The material consists of lignin, cellulose, and hemicellulose, with polyphenolic groups playing important role for binding dyes through different mechanisms. Generally, the adsorption takes place by complexation, ion exchange, and hydrogen bonding. The effect of pH on the batch kinetics of MB adsorption on beech sawdust was studied, in order to evaluate the potential use of sawdust as low-cost adsorbent for dye removal. The zero point charge pH_{pzc} of the sawdust was studied, in order to explain the effect of pH in terms of $pH_{pzc,}$ It was measured by the mass titration and the automatic titration methods. The adsorption capacity, estimated according to Freundlich's model, indicated that increase of the pH enhances the adsorption behavior of the adsorbent material. The lower adsorption of MB at acidic pH is due to the presence of excess H^+ ions that compete with the dye cation for the available adsorption sites. As the pH of the system increases, the number of positively charged sites decreases while the number of the negatively charged sites increases. The negatively charged sites favor the adsorption of dye cation due to electrostatic attraction. The increase in initial pH from 8.0 to11.5 increase the amount of dye adsorbed. Above the point of zero charge (pH_{pzc} 5.2), the negatively charge density on the surface of the adsorbent favors the sorption of basic dyes such as MB. The significantly lower adsorption of MB at acidic pH can be attributed to the presence of excess H^+ ions that compete with the dye cation for adsorption sites. As the pH of the system increases, the number of positively charged sites decreases while the number of the negatively charged sites increases. The negatively charged sites favor the adsorption of dye cation due to electrostatic attraction. This amount decreases at higher pH values, due to MB stepwise demethylation. The adsorbent was found to be a suitable alternative to the commercial activated carbons for the removal to basic dyes from nonacidic water or wastewater effluents [201].

Hamdaoui [202] studied the removal of MB, from aqueous solution (40 mg L^{-1}) onto cedar sawdust in order to explore their potential use as low-cost adsorbents for wastewater dye removal. Adsorption isotherms were determined at 20°C and the experimental data obtained were modeled using the Langmuir, Freundlich, Elovich, and Temkin isotherm equations. Considering the experimental results and adsorption models applied in this study, it was concluded that equilibrium data were represented well by Langmuir isotherm equation with maximum adsorption capacity of 142.36 mg g^{-1} for cedar sawdust. The extent of the dye removal decreased with increase in the solution temperature and optimum pH value for dye adsorption was observed at pH 7 for both adsorbents. Ahmad et al. [203] investigated the scavenging behavior of meranti sawdust in the removal of MB from aqueous solution. Batch studies were performed to evaluate and optimize the effects of various parameters such as contact time, pH, initial dye concentrations, and adsorbent dosage. Langmuir, Freundlich, and Temkin isotherms were used to analyze the equilibrium data at different temperatures. Equilibrium data fitted very well in the Langmuir isotherm equation, confirming monolayer adsorption (120.48 mg g^{-1}) of MB onto meranti sawdust. Most of the studies showed that sawdust in natural form or modified form is highly efficient for the removal of MB [73,204–208]. Chemical pretreatment of sawdust has been shown to improve the adsorption capacity and to enhance the efficiency of sawdust adsorption [201,208,209]. Batzias and Sidiras [201] also studied beech saw dust as low-cost adsorbent for the removal of MB.

10. Others adsorbents

10.1. *Green coconut fiber*

Raquel and coworkers [210] immobilized commercial laccase for the first time on an agro industrial residue, green coconut fiber, by physical adsorption. The effect of the immobilization conditions (enzyme concentration, contact time and pH value) on the properties of the biocatalyst was determined. Then, the immobilized enzyme characterization was performed and kinetic parameters were obtained. Thermal and operational stabilities were improved compared with free commercial laccase showing its potential for continuous applications. Finally, the performance of immobilized laccase for the continuous degradation of various reactive textile dyes [163] and their mixture in batch reactors was evaluated. Decolorization of the solutions due to the adsorption of dyes on the support and due to the enzyme action was observed. A high decolorization percentage of practically all dyes in the first two cycles and an effective decolorization of the dye mixture were obtained, showing the suitability of the immobilized commercial laccase for continuous color removal from textile industrial effluents [210].

10.2. Fly ash

Suna and coworker [211] studied the adsorption behavior of two reactive dyes (Reactive Red 23 and Reactive Blue 171) and two acid dyes (acid black 1 and acid blue193) from aqueous solution on fly ash in order to identify the ability of this waste material to remove colored textile dyes from wastewater. For this purpose, a series of batch tests were carried out as a function of solution pH value, contact time, dye concentration, and adsorption temperature. Experimental findings showed that the removal of four dyes on fly ash was a pH-dependent process with the maximum adsorption capacity at the initial solution pH of 7.5–8.5 for reactive dyes and 5–6 for acid dyes. Adsorption equilibrium of each anionic dye on fly ash was reached within 60 min at optimum pH at 293 K. An increase in the initial dye concentration enhanced the adsorption capacity, but failed to increase the dye removal efficiency. The adsorption capacity for Reactive Red 23, Reactive Blue 171, acid blue193, and acid black 1 was found to be 2.102, 1.860, 10.937, and 10.331 mg g^{-1}, respectively. Kinetic studies of four dyes followed the pseudo-second-order model. Freundlich isotherm described the equilibrium data of acid dyes on fly ash better than Langmuir isotherm, but Langmuir isotherm gave better fit to the equilibrium data of reactive dyes. The values of Langmuir and Freundlich constants were found to be 18.939 and 0.989 mg g^{-1} for AB1, 22.075 and 1.936 mg g^{-1} for AB193, 5.041 and 0.980 mg g^{-1} for RR23, and 3.754 and 0.879 mg g^{-1} for RB171, respectively. Different thermodynamic parameters such as the free energy, enthalpy, and entropy of adsorption of the dye–fly ash systems were evaluated and it was found that the reaction was spontaneous and endothermic in nature [211]. Bello and coworkers [212] carried out batch experiment on the adsorption of eosin dye onto fly ash. Operational parameters such as contact time, initial dye concentration, pH, and temperature were varied. Langmuir and Freundlich isotherm models were used to fit the sorption data. Freundlich model fitted better. The maximum adsorption capacity of fly ash obtained was 48.48 mg g^{-1} at 30°C at 10 mg L^{-1} eosin dye. Two kinetic models, pseudo-first-order and pseudo-second-order, were used to fit the data. Pseudo-second-order kinetic model gave the best description of the adsorption of eosin dye unto fly ash. Thermodynamic parameters, ΔH°, ΔS°, and ΔG°, revealed the physical nature, spontaneity, and the endothermic nature of the adsorption process. The study showed that fly ash could be used efficiently as low-cost adsorbent for removing dyes from aqueous solution [212–215].

10.3. Bamboo

Char was prepared from waste bamboo scaffolding for wastewater treatment [216,217]. Carbonization parameters such as temperature, holding time, heating rate, and particle size were investigated. When the material was heated to 1,173 K for 2 h, surface area (SBET-N_2) and total pore volume (V_{total}) were 327 m^2 g^{-1} and 0.185 cm^3 g^{-1}, respectively. Particle size and heating rate appear less influential to the yield and textural characteristics of the resultant chars. Bamboo char obtained in this study did not show very high adsorption capacities for the two acid dyes (acid blue 25 and acid yellow 117) but exhibited significant uptake of MB. The predicted data were compared using optimized parameters from each model based on the SSE error function. The Redlich–Peterson isotherm was the best model that correlated the experimental data. Bamboo char obtained in this study did not show very high adsorption capacities for two acid dyes (acid blue 25 and acid yellow 117) but exhibited a significant uptake of MB over 0.99 mmol g^{-1}. The effect of temperature, holding time, heating rate, and particle size on the characteristics of chars prepared from bamboo was investigated. It was found that, temperature and holding time are the most important factors affecting the yield of chars and surface areas as most volatiles were evolved at temperature of 773 K; increasing temperature raised the surface area at the expense of yield. Heating rate and particle size are not influential but they may also affect the textural characteristics of the resultant chars [216].

10.4. Tyre rubber waste

Converting waste tyre rubber to activated carbon for liquid phase application has received substantial attention in the last decade [218]. Two types of activated carbons from tyre char (with or without sulfuric acid treatment) were produced via carbon dioxide activation with BET surface areas in the range 59–1,118 m^2 g^{-1}. Other characterization tests include micropore and mesopore surface areas and volumes, pH, and elemental compositions, particularly heteroatoms such as nitrogen and sulfur. The adsorption capacities were in the range of 0.45–0.71 mmol g^{-1} (untreated) and 0.62–0.84 mmol g^{-1} (acid-treated) for acid blue 25. In the case of larger sized molecules like acid yellow 117, the capacities were in the range of 0.23–0.42 mmol g^{-1} (untreated) and 0.29– 0.40 mmol g^{-1} (acid-treated). Some tyre carbons exhibit a more superior performance than the microporous commercial activated carbon [218]. By modeling the dye adsorption equilibrium data, the Redlich–Peterson isotherm [218] was adopted as it has the lowest SSE. Based on the surface coverage analysis, a novel molecular orientation modeling of adsorbed dyes has been proposed and correlated with surface area and surface charge. For the acid dyes used in this study, molecules were likely to be absorbed by the mesopore areas [218–220].

10.5. Pearl millet husk

The potential of pearl millet husk (PMHC) as an adsorbent was explored by Inbaraj et al. [221]. An adsorption capacity of 82.37 mg g^{-1} of this adsorbent for MB at pH 6.0 was reported. They further reported that MB adsorption on PMHC was a chemisorption process and formic acid could be used to remove the adsorbed dye. The potential of cotton plant wastes—stalk (CS) and hull (CH)—as sorbents for the removal of Remazol Black B (RB5) was investigated [221]. The results indicated that adsorption was strongly pH dependent but slightly temperature-dependent for each sorbent–dye system. CS and CH sorbents exhibited the highest RB5 dye uptake capacities of 35.7 and 50.9 mg g^{-1}, respectively, at an initial pH value of 1.0. It was found that both external mass transfer and intraparticle diffusion played an important role in the adsorption mechanisms of dye, and adsorption kinetics followed the pseudo-second-order kinetic model for each sorbent.

10.6. Adsorbents from seed, seed coat, stem, and stalk of different agricultural products

Many researchers also examined the suitability of seed, seed coat, stem, and stalk of various agricultural products as inexpensive adsorbents for the removal of toxic pollutants from water [222]. The feasibility of papaya seeds (PS) for the MB adsorption has been investigated [223]. Batch adsorption studies were conducted to study the effects of contact time, initial concentration (50–360 mg L^{-1}), pH (3–10), and adsorbent dose (0.05–1.00 g) on the removal of MB (MB) at 30°C. The data fitted well with the Langmuir model with a maximum adsorption capacity of 555.55 mg g^{-1}. The pseudo-second-order kinetics fitted well for the adsorption of MB by PS with good correlation.

Sunflower stalks as adsorbents were also reported to be used for the removal of two basic dyes (MB and basic red 9) and two direct dyes (CR and direct blue 71) from aqueous solutions by Sun and Xu [224]. The maximum adsorption of two basic dyes on sunflower stalks was found to be 205 and 317 mg g^{-1} for MB and basic red 9, respectively. They also reported relatively lower adsorption for two direct dyes on sunflower stalks. Table 4 reveals the comparison of adsorption capacities for activated carbons derived from different agricultural wastes for the removal of MB dye while Table 5 compares the Freundlich (K_F, n) and the Langmuir isotherm parameters (q_m, K_L) for various adsorbents for the most common adsorbate—MB dye.

Shi et al. [271] improved the adsorption capacity of sunflower stalks by chemically grafting quaternary ammonium groups on them. The modified sunflower stalks exhibited increased adsorption capacity for anionic dyes, due to the existence of quaternary ammonium ions on the surface of the residues. The maximum adsorption capacities on modified sunflower stalks were found to be 191.0 and 216.0 mg g^{-1} for CR and direct blue, respectively, which were at least four times higher than that observed on unmodified sunflower stalks. Further, the same authors observed that adsorption rates of two direct dyestuffs were much higher on the modified residues than on unmodified ones. A comparison of various low-cost adsorbents derived from different agricultural wastes for the removal of diverse types of aquatic pollutants is summarized in Tables 1 and 2. The percentage removal corresponding to 1 mg/L dose of activated carbon CC-AC, 100 mg/L dose of Z1 and Z2 and three weeks contact time, 5 mg/L dose of PAC and 4 h contact timeis also shown (Fig. 5).

10.7. Adsorbent from *C.* elegans

Russo and coworker reported on dye adsorption on granular particles of lyophilized *C. elegans* which was characterized in terms of adsorption isotherm and

Table 4
Comparison of adsorption capacities for activated carbons derived from agricultural wastes for MB dye removal

Adsorbent	Adsorption capacity (mg g^{-1})	Refs.
Periwinkle shells	500.00	[225]
Palm kernel fiber	217.95	[226]
Modified rice straw	208.33	[227]
Broad bean peels	192.72	[228]
Gulmohar (*Delonix regia*) plant leaf powder	186.22	[229]
Castor seed shell	158.73	[230]
Cedar sawdust	142.36	[231]
Pumpkin seed hull	141.92	[232]
Saw dust	133.87	[233]
Chemically treated guava leaves	133.33	[232]
Meranti sawdust	120.48	[234]
Dehydrated peanut hull	108.6	[235]
Coconut	99	[236]
Coffee husks	90.1	[237]
Phosphoric acid treated *Parthenium hysterophorus*	88.49	[238]
Teak wood bark	84	[239]
Garlic peel	82.64	[240]
Rubber seed shell	82.64	[241]
Fallen phoenix tree's leaves	80.9	[242]
Raw date pits	80.3	[243]
Coconut bunch waste	70.92	[244]
Peanut hull	68.03	[245]
Luffa cylindrica fibers	47	[246]
Yellow passion fruit waste	44.70	[247]
Rice husk	40.59	[248]
Cherry sawdust	39.84	[73]
Sulfuric acid treated *Parthenium hysterophorus*	39.68	[238]
Hazelnut shell	38.22	[249]
Mansonia wood sawdust	33.44	[250]
Paspalum notatum (garden grass)	30.4	[251]
Oak sawdust	29.94	[73]
Rice husk	28	[239]
Pitch-pine sawdust	27.78	[73]
Cotton waste	24	[239]
Salsola vermiculata leaves	23	[252]
Jute processing waste	22.47	[253]
Banana peel	20.8	[254]
Cereal chaff	20.3	[255]
Orange peel	18.6	[254]
Spruce wood shavings from *Picea abies*	17.91	[256]
Wheat shells	16.56	[257]
Indian rosewood sawdust	11.8	[258]
Neem (*Azadirachta indica*) leaf powder	3.67	[259]
Fine grinded wheat straw	2.23	[260]
Grass waste	457.64	[261]
Pomelo (*Citrus grandis*) peel	324.83	[262]
Untreated guava leaves	295	[263]
Jackfruit peel	1285.71	[264]
Cotton waste	277.77	[266]

kinetics [273]. The study refers to dyes of an acid bath for wool: acid blue 62, acid red 266, and acid yellow 49. The dye concentration in model solutions—containing a single dye or the three dyes all together in order to mimic the wastewater—was increased up to about 500 mg L^{-1}. Tests showed that the maximum adsorption capacity of the biomass ranges between 300 and 600 mg dye g^{-1} DM. Mutual interferences among dyes caused the reduction of the adsorption capacity of the biomass towards the model wastewater. An experimental procedure for the assessment of biosorption kinetics was developed in order to control the effects of the interphase mass transfer on the biosorption rate (Fig. 6) [273]. The biosorption kinetics were described by both pseudo-first-order and pseudo-second-order models, depending on the saturation level of the sorbent, it was characterized by a time scale of 1–10 min. The role of the molecular structures of the dyes were discussed. In particular, both kinetics and equilibrium tests confirm that the biomass is more selective towards AR266, probably for the high negative charge density of the—CF3 functional group that can interact with—NHx active sites of the biomass. The reported scenario suggests that the development of novel biomasses having high adsorption capacity towards specific pollutants asks for the characterization of biosorption equilibrium and kinetics. Recent studies have shown that inactivated biomass of the zygomycetes *C. elegans* has proven to be a reliable and cost-effective alternative to activated carbon to remove dyes and chromium from water [274–276]. It was patented for dye biosorption [277] and deposited at the Deutsche Sammlung von Mikroorganismen und Zellkulturen (DSMZ-Braunschweig, Germany).

11. Major challenges and future prospects

In this review, we made an attempt to focus on the recent developments related to the dye adsorption from aqueous solution, water, and wastewater by low-cost adsorbents using biomass wastes and natural adsorbents. It is important to note here that the maximum adsorption capacities reported in this paper

Table 5
Langmuir and Freundlich isotherm constants for adsorption of MB onto some selected adsorbent [265–270]

Adsorbents	Isotherms			
	Langmuir		Freundlich	
	q_m (mg g^{-1})	K_L (dm^3 mg^{-1})	K_F (mg g^{-1})	n
Pineapple stem	119.05	0.00009	25.42	2.97
Cotton fiber	56.40	14.780	91.87	1.972
Wool fiber	84.90	0.0036	0.984	6.127
Orange peel	10,000	0.00502	0.775	1.50
Neem leaves	3,333.33	0.00903	0.327	0.612
Banana peel	10,000	0.00411	0.635	1.273
Sawdust	19.41	1.94	16.3	3.2
Sawdust coated with polypyrrole	34.36	38.2	4.8	4.0
Langsat (*Lansium domesticum*) peel	45.454	0.039	4.899	2.409
Ground palm kernel coat	277.77	0.273	55.219	1.870
Acid modified *Calotropis procera* leaf powder	192.31	2.73×10^{-3}	1.16	0.76

provide some idea of adsorbent's effectiveness for each type of pollutant, and mainly depends on experimental conditions. The use of waste materials as low-cost adsorbents for removing various pollutants from aqueous solution, water, and wastewater presents many attractive features; especially, their contribution in the reduction of costs for waste disposal, therefore, contributing to environmental protection.

Although the amount of available literature data on the use of low-cost adsorbents for the adsorption of dyes from aqueous solution, water, and wastewater is increasing at a tremendous pace, there are still several gaps which need to be filled in the aspect of the selection and identification of an appropriate low-cost adsorbent which is one of the key issues to achieve the maximum removal/adsorption of specific type of pollutant depending upon the adsorbent–adsorbate characteristics. Therefore, the conditions for the production of low-cost adsorbents after surface modification for higher uptake of pollutants need to be optimized. The world is currently facing the worst environmental crisis in its entire history. Within the space of last few decades, the enthusiasm of huge waste production and environmental preservation has been one of the most challenging topics which have been of public concern and critical considerations towards the recovery of contamination resources. Since industrial effluents are always contaminated

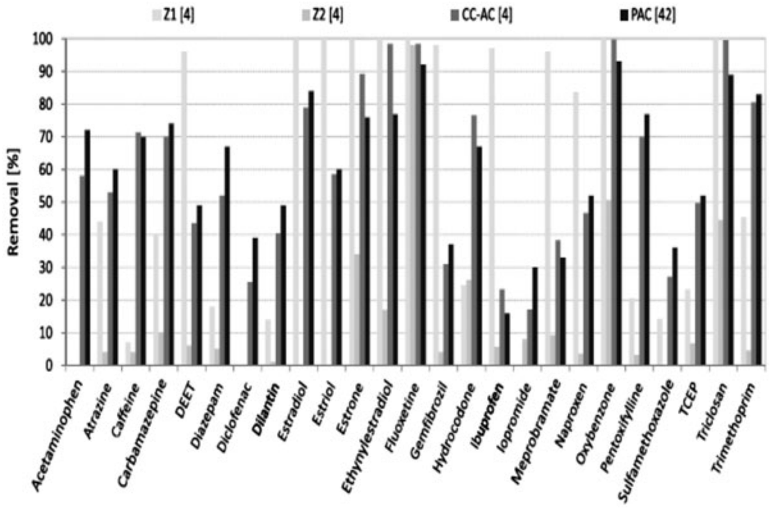

Fig. 5. Removal percentages correspond to 1 mg L^{-1} dose of activated carbon CC-AC, 100 mg L^{-1} dose of Z1 and Z2 and three weeks contact time, 5 mg L^{-1} dose of PAC and 4 h contact time [272].

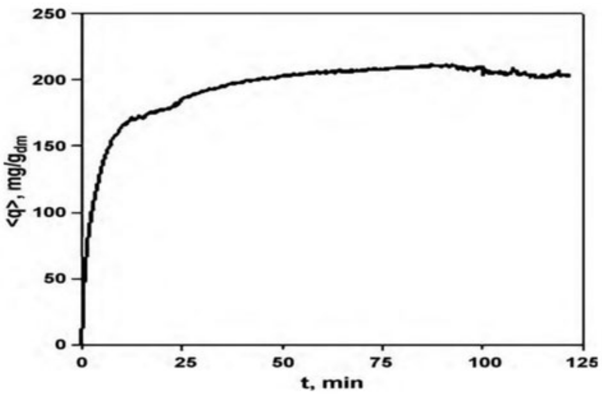

Fig. 6. Time-resolved concentration of adsorbed AR266 on *C. elegans*. Liquid volume: 0.2 L; biomass: 0.03 g; $C_0 = 250$ mg L^{-1} [273].

with various additives such as inorganic salts, it is therefore important to study the effect of these ions on the adsorption property of dye solutions. The adsorption of dye in the presence of anions (added as sodium salts) and cations (added in the nitrate form) should be carried out.

The carcinogenic nature of dye creates numerous environmental problems in waste products and aqueous solutions. Dye waste products are inevitably formed during the numerous industrial processes using dyes. The textile industries use high volumes of dyes in their processes. Waste products from these processes normally contain compounds that are not environmental friendly. Management of waste products from industrial processes is always problematic and is associated with poor housekeeping. Therefore, dye is commonly spilled or leaked into the environment as a contaminant, frequently with other associated wastes and ultimately moving into the aqueous solutions, soil, and groundwater. The characteristics of dyes in the sub-surface make assessment and remediation of these dyes-impacted sites challenging. Competent environmental professionals should be employed to assess and remediate dyes that contaminate soil and groundwater resources. The environmental consulting industry in conjunction with the regulatory agencies, including the USEPA, must continue to establish reliable, accurate analytical methods for dye adsorption. The environmental industry must develop and optimize effective *ex situ* treatment systems for dyes in aqueous solutions, especially in areas where trace levels exist. Environmental regulatory agencies must evaluate health risks posed by dye concentrations and establish background levels for aqueous solutions (accompanied by health effect studies) before implementing new regulations.

With the arising concern for the protection of the environment in recent time, the necessity for protection of the environment from huge waste production has been a major challenging task for the general public; several studies have been carried out on the use of activated carbon as adsorbent in the adsorption of these hazardous wastes. Due to the fact that activated carbon is expensive and not always available, this has lead to the use of low-cost activated carbon obtained from agricultural waste. Despite the numerous work done on the use of agricultural wastes and other adsorbents, there are certain limitations; first, less emphasis has been placed on the regeneration of the activated carbon for possible reuse, this will help in reducing cost. Secondly, surface charge of both the activated carbon and the dyes should be thoroughly investigated; as surface charge enhances the attraction of the dye to the adsorbents. Also, these adsorbent should be tested with real industrial effluents to demonstrate its ability on large scale and its performance on the field so that the studies will not be limited to the laboratory alone. The search for alternative adsorbents to activated carbon in dye removal should equally be geared towards getting adsorbents with shorter contact time, as these will enhance their industrial uses.

Another crucial challenge is that there is scarce data available for the competitive adsorption of pollutants (dyes adsorption and other contaminants). Therefore, more research should be focused in this direction. Research should also be conducted on pilot-plant studies with the utilization of low-cost adsorbents to check their feasibility on commercial scale and not limited only to laboratory scale batch studies. It is, therefore, also necessary to embark on environmentally safe disposal of pollutants-laden adsorbents which is another important topic of concern which should not be overlooked. Furthermore, for any work on chemically modified plant/biomass wastes, characterization studies involving surface area, pore size, porosity, pH_{ZPC}, etc. should be carried out because the effectiveness of the treatment depends not only on the properties of the adsorbent and adsorbate but also on various environmental conditions and variables used for the adsorption process, e.g. pH, ionic strength, temperature, existence of competing organic or inorganic compounds in solution, initial adsorbate/adsorbent concentration, contact time and speed of rotation, particle size of adsorbent, etc. These parameters should also be taken into account while examining the potential of low-cost adsorbents. Spectroscopic analyses involving Fourier transform infrared, energy dispersive spectroscopy, X-ray absorption near edge structure spectroscopy, and extended X-ray absorption fine structure spectroscopy are also important in order to have a better understanding on the mechanism of dye adsorption on modified plant wastes.

12. Conclusion

The increase in number of modern industries that uses dyes for their production has affected the rate of environmental pollution and by extension it has affected both human and aquatic lives. In this literature, the use of waste biomass as adsorbent for adsorption of dyes in dye-contaminated water has been discussed. To date, limited success has been recorded in field application and has raised apprehension over the use of adsorption capacity as a measure of the extent of adsorption in real industrial effluents and in waste water treatments because these wastes contain mixture of dyes. A wide and an encompassing study in this area are expected in the nearest future. The sorption capacity is dependent on the type of the adsorbent investigated and the nature of the aqueous solution treated. The use of commercially available activated carbon for the removal of the dyes can be replaced by the utilization of inexpensive, effective, and readily available biomass wastes and natural by-products as adsorbents. More studies should be carried out to better understand the process of low-cost adsorption and to demonstrate the technology effectively.

Literature studies showed above highlighted that adsorption process can be considered an efficient treatment for the removal of emerging compounds from water. It gave good removal percentage and, being a physical process, does not imply by-products formation, which could be more toxic than parent compounds. It is obvious that adsorption process is encompassed in an integrated treatment system which involves many factors, such as available space for the construction of treatment facilities, waste disposal constraints, desired finished water quality, and capital and operating costs. All these factors imply the achievement of the optimal operating conditions for low-cost high efficiencies.

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