Desalination and Water Treatment
www.deswater.com

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doi: 10.5004/dwt.2010.1214

19 (2010) 255–274
July

An overview of dye removal via activated carbon adsorption process

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Received 20 May 2009; Accepted in revised form 29 December 2009

ABSTRACT

Water scarcity and pollution rank equal to climate change as the most intricate environmental turmoil for the 21st century. Today, the percolation of textile effluents into the waterways and ecosystems remain a fastidious conundrum towards the public health and food chain interference. In such circumstances, it becomes mandatory for each dye emitting industry or facility to opt for proper dye control measures. With the renaissance of activated carbon, there has been a steadily growing interest in this research field. This paper attempts to postulate a state of the art review of dye processing industry, its fundamental characteristics and environmental implications. Moreover, the key advance of proposed precursors, activated agents, together with the effects of adsorbent dosage, concentration, contact time, pH, particle size, temperature competition, and isotherms, kinetic and thermodynamic studies for adsorption of dyes onto activated carbons are summarized and discussed. Conclusively, the expanding of adsorption science in dye treatment represents a plausible tool for accruing the worldwide environmental benefit and shaping the national economy.

Keywords: Activated carbon; Adsorption; Dye; Isotherm; Kinetics; Thermodynamics

1. Introduction

1.1. Textile wastewater

Concern about environmental protection has increased over the years from a global viewpoint. Today, rapidly changing technologies, industrial products and practices generate waste that, if improperly managed, could threaten public health and the environment. Various industries such as dye manufacturing, pulp and paper, tanneries, cosmetic, coffee pulping, pharmaceuticals, food processing, electroplating, and distilleries spew out colored and toxic effluents to water bodies rendering them murky, dirty, and unusable for further use [1]. Among all industrial sectors, textile industries are rated first seat among the most important sources of contamination responsible for continuous pollution of the environment. With the escalating demand for textile products, textile industry and its wastewaters have risen proportionally [2]. In particular, textile industries consume large volumes of water and chemicals for wet processing of textiles. In the dyeing section of a textile industry, approximately 1000 L of water is used for every 1000 kg clothes processed [3].

Accordingly, Environmental Protection Agency (USEPA) has classified textile wastes into four groups, namely dispersible, hard to-treat, high-volume, and hazardous and toxic wastes [4]. These textile wastewaters are usually a mixture of large complex, particulates, processing assistants, salts, surfactants, acids and alkalis, which diverse widely in chemical composition, ranging from inorganic compounds to polymers and organic products. Among different pollutants of aquatic ecosystems, dyes are a large

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and important group of industrial chemicals with over 700,000 tons of waste produced annually [5]. Considering both volume discharged and effluent composition, wastewater from textile industry was declared as one of the major sources of wastewater in the Association of Southeast Asian Nations (ASEAN) countries [6]. Statistically, the total dye consumption of the textile industry worldwide is in excess of 107 kg/y, and an estimated 90% of this ends up on fabrics. Consequently, 1,000 t/y or more of dyes are discharged into waste streams by the textile industry [7].

Today, synthetic dyes are used extensively for textile dyeing and as additives in petroleum products. There are more than 10,000 different types of dyes and pigments available commercially and about 10–15% of dyes are lost in the effluent during the dyeing process [8]. Among them, more than 9,000 types of dyes have been incorporated in the color index and environmental regulations in most countries have made it mandatory to decolorize dye wastewater prior to discharge [9]. Lately, the discharge of textile effluents in the environment has received great attention, because it is inherently highly visible to human eye, which captures the attention of both public and the authorities, due to the physiological impact upon the population [10]. Visual pollution, aggravated or not by toxicity questions is by itself a serious problem in water quality. By design, dyes are highly stable molecules, specifically resist fading upon exposure to sweat, light, water, heat and oxidizing agents. Inefficiencies in dyeing of textiles result in large amounts of the non-biodegradable, toxic and inhibitory nature of spent dye baths being lost directly into wastewater and consequently having a considerable detrimental effect on flora, fauna, and environmental matrix (water and soil) [11].

Numerous studies have been conducted to assess the harm impacts of colorants on the ecosystem. The real hazard setting aside aesthetic considerations is caused when colored agents interfere with the transmission of light through water, retard photosynthetic activities in aquatic life, inhibit the growth of biota, affect the symbiotic process, damage the quality of the receiving streams and toxic to food chain organisms, thus upsetting biological processes within streams, resulting in ecological imbalance [12]. Recent studies indicated that such colored effluents in wastewater may undergo chemical and biological changes, consume dissolved oxygen from the stream, prevent reoxygenation in receiving waters and have a tendency to sequester metal ions producing microtoxicity to fish and other organisms [13]. In some cases, less than 1 ppm of dye concentration may produce obvious water coloration [14]. Additionally, acute exposure to colored effluents can cause increased heart rate, vomiting, shock, Heinz body formation, cyanosis, jaundice, quadriplegia, tissue necrosis, allergy, dermatitis, eye or skin infections, irritation and have carcinogenic and mutagenic effects [15].

1.2. Dye removal technologies

In recent decades, a wide range of conventional treatment technologies for dye removal has been investigated extensively. The conventional methods for treating dye-containing wastewater include coagulation–flocculation, sedimentation, membrane technologies (reverse osmosis, nanofiltration and dialysis), cloud point extraction, microbiological decomposition, aerobic and anaerobic degradation, chemical oxidation technologies (Fenton's reagent with hydrogen peroxide, photo-catalysis with UV radiation and ozonation), electrochemical treatment and adsorbents utilization (activated carbon, inorganic adsorbents such as silica or clays, synthetic ion, exchange resins and chitin-based adsorbents) [16–45]. Table 1 illustrates critical study of the most widely used methods of dye removal from dye containing industrial effluents. Among these processes, adsorption has been found to be more versatile and superior to other techniques for wastewater treatment in terms of initial cost, simplicity of design, ease of operation, insensitivity to toxic substances and complete removal of dyes even from dilute solutions [48].

1.3. Adsorption process using activated carbon

In general, adsorption is a separation process by which certain components of a fluid phase are attracted to the surface of a solid adsorbent and form attachments via physical or chemical bonds, thus removing the component from the fluid phase [49]. More precisely, adsorption processes usually occur in interfacial layers, which are regarded as two regions: the surface layer of the adsorbent (often simply called the adsorbent surface) and the adsorption space, in which the enrichment of the adsorptive can occur. It may be classified as physical or chemical depending on the nature of forces involved [50]. Table 2 exhibits the general features which distinguishing physical and chemisorption. The quality of the liquid-phase adsorption process can be influenced by various physico-chemical factors and these include properties of the adsorbed material (molecular size, boiling point, molecular mass and polarity), properties of the surface of the adsorbent (polarity, pore size and spacing), effect of other ions, particle size, solution pH, temperature and contact time [52].

For this purpose, activated carbon (AC) has been proven to be an effective adsorbent for the removal of a wide variety of organic and inorganic pollutants dissolved in aqueous media, or from gaseous environment [53]. In fact, activated carbon is a crude form of graphite with a random or amorphous structure, which is highly porous, exhibiting a broad range of pore sizes, from visible cracks, crevices and slits of molecular dimensions. The wide usefulness of activated carbon is a result of its chemical and mechanical stability, high adsorption capacity and high degree of surface reactivity [54]. These unique characteristics are usually related to its internal

Table 1
Advantages and drawbacks of current methods of dye removal from industrial effluents [46,47]

Physical/chemical methods	Method description	Advantages	Disadvantages
Fenton reagents	Oxidation reaction using mainly H_2O_2-Fe(II)	Effective decolorization of both soluble and insoluble dyes	Sludge generation
Ozonation	Oxidation reaction using ozone gas	Application in gaseous state: no alteration of volume	Short half-life (20 min)
Photochemical	Oxidation reaction using mainly H_2O_2-UV	No sludge production	Formation of by-products
NaOCl	Oxidation reaction using Cl^+ to attack the amino group	Initial and acceleration of azo-bond cleavage	Release of aromatic amines
Electrochemical destruction	Oxidation reaction using electricity	Breakdown compounds are non-hazardous	High cost of electricity
Activated carbon	Dye removal by adsorption	Good removal of a wide variety of dyes	Regeneration difficulties
Membrane filtration	Physical separation	Removal of all dye types	Concentrated sludge production
Ion exchange	Ion exchange resin	Regeneration: no adsorbent loss	Not effective for all dyes
Electrokinetic coagulation	Addition of ferrous sulphate and ferric chloride	Economically feasible	High sludge production

Table 2
Typical characteristics of physical adsorption and chemisorption processes [51]

Parameter	Physical adsorption	Chemisorption
Adsorption enthalphy	Low (<2 or 3 times latent heat of evaporation)	High (>2 or 3 times latent heat of evaporation)
Specificity	Non specific	Highly specific
Adsorption site	Monolayer or multilayer	Monolayer only
Nature of adsorption	No dissociation of adsorption species	May involve dissociation
Temperature range	Only significant at relatively low temperature	Possible over a wide range range of temperature
Kinetics of adsorption	Rapid, non activated, reversible	Activated, may slow and reversible
Electron transfer	No electron transfer although polarization of sorbate may occur	Electron transfer leading to bond formation between sorbate and surface

pore structure, surface characteristics, pore volume, porosity, chemical nature of the source materials, presence of functional group on pore surface and the activation process. Surface structures of carbon oxygen (functional groups) are by far the most important structures influencing surface characteristics of activated carbon, and functional groups suggested most often are the carboxyl (I), phenolic hydroxyl (II), carbonyl (e.g., quinone-type) (III), and lactone groups (fluorescein-type) (IV) [55].

1.4. Preparation of activated carbon

Basically, there are two different processes for the preparation of activated carbon: physical and chemical activation. Physical activation involves carbonization of the carbonaceous precursor followed by activation of the resulting char in the presence of activating agents such as carbon dioxide or steam. Chemical activation, on the other hand, involves the carbonization of the precursor in the presence of chemical agents [56]. In physical activation, the elimination of a large amount of internal carbon mass is necessary to obtain a well developed porous structure, whereas in chemical activation process, chemical agents used are dehydrating agents that influence pyrolytic decomposition and inhibit the formation of tar, thus enhancing yield of carbon [57]. Table 3 lists technical advantages and disadvantages of the existing modification techniques.

Despite its prolific use in water and wastewater treatment, the biggest barrier of its application by industries is

Table 3
Technical advantages and disadvantages of existing modification techniques [58]

Modification	Treatment	Advantages	Disadvantages
Chemical characteristics	Acidic	Increases acidic functional groups on AC surface. Enhances chelation ability with metal species surface.	May decrease BET surface area and pore volume Has adverse effect on uptake of organics May give off undesired SO_2 (treatment with H_2SO_4) or NO_2 (treatment with HNO_3) gases
	Basic	Enhances uptake of organics	May, in some cases, decrease the uptake of metal ions
	Impregnation of foreign material	Enhances in-built catalytic oxidation capability	May decrease BET surface area and pore volume
Physical characteristics	Heat	Increases BET surface area and pore volume	Decreases oxygen surface functional groups
Biological characteristics	Bioadsorption	Prolongs AC bed life by rapid oxidation of organics by bacteria before the material can occupy adsorption sites	Thick biofilm encapsulating AC may impede diffusion of adsorbate species

the cost-prohibitive adsorbent and difficulties associated with regeneration [59]. This has prompted a growing research interest in utilizing of natural as well as renewable and cheaper precursor (waste materials) as alternative adsorbents for the production of activated carbons. The aim behind using waste materials as adsorbents is that it will provide a twofold advantage with respect to environmental pollution. First, the volume of waste materials could be partly reduced, and second, the low-cost adsorbent, if developed, can reduce the pollution of wastewaters at a reasonable cost [60]. In view of the aforementioned, this bibliographic review surveys recent publications on the adsorption of various dyes from aqueous solution by commercial and synthesized activated carbon. The present work is aimed at providing a concise and up to date picture of the dye removal technologies, physical adsorption, chemisorption processes and modification techniques. The effects of parameters, kinetic, isotherm and thermodynamic studies have been highlighted and outlined, to familiarize the knowledge deficiencies regarding dye removal technologies.

2. Precursors and activation agents for preparation of activated carbon

Over the last few years, the selection of activation agents for preparation of activated carbons has been one the most challenging tasks, which is gaining extensive attention and stern consideration around the world. In fact, the nature of an activated carbon surface is usually related to its modifications and treatments, whereby a thorough knowledge of the surface chemistry represents a convenient means of guideline, enabling the preparation of ACs with appropriate characteristics for specific applications [61]. Table 4 lists the effect of activations towards adsorption capacities for numerous dye molecules. Analysing the research data, steam activation appears to be most effective, signifying the highest adsorption performance of 1176 mg/mg for anionic (acidic, direct and reactive) and 1250 mg/mg for cationic (basic) dye molecules. In the treatment process, oxygen releasing groups (carboxylic, anhydrides and lactones) are selectively removed from the carbon surface, resulting in carbonyls, pyrone and chromene type structures, with reactive sites capable of readsorbing oxygen, leading to the formation of previously removed groups [114]. At higher temperature, pyrone and chromene structures are presumed to be the only oxygen containing groups remain on the carbon surface, thus originating electron rich oxygen-free sites located on the carbon basal planes with the highest basic character.

Meanwhile, oxidative treatments with hydrochloric (HCl), phosphoric (H_3PO_4) and nitric (HNO_3) acids are associated with a substantial enhancement in volatiles content, which is corroborated with large amounts of surface acidic groups, mainly the carboxylic group and in less extent, the lactones, anhydrides, phenol and carbonyl groups [115]. In the perspective, the abundant presence of oxygen-containing groups displays an electron-withdrawing characteristic, which corresponding to the reduction of electron density on the carbon surfaces, simultaneously induce a repulsive force for anionic species and good anchoring sites for cationic dyes, indicating a preferable adsorption for basic dye molecules (maximum adsorption capacity of 450.5 mg/mg).

On the contrary, base treatments (sodium hydroxide, potassium hydroxide or ammonia) at low temperatures tend to produce basic surfaces with relatively large ion exchange capacities, characterized by high content of electron rich sites and low concentration of electron withdrawing groups [116]. Accordingly, the interaction is expected to occur between the delocalised π electrons

Table 4
Activation agents proposed for preparation of activated carbons in dye removal processes

Activation agents	Maximum monolayer adsorption capacity qe (mg/g)							
	Basic dye	Ref.	Acid dye	Ref.	Direct dye	Ref.	Reactive dye	Ref.
Steam	1250.00 (Bismark brown R)	[62]	1176.00 (Acid blue 264)	[65]	518.00 (Direct blue 2B)	[67]	509.00 (Orange 16)	[69]
	710.10 (Basic red 22)	[63]	1060.00 (Acid blue 25)	[66]	64.00 (Direct Orange 34)	[68]		
	588.00 (Methylene blue)	[64]	904.4 (Acid blue 25)	[63]				
H_2SO_4	221.23 (Astrazon yellow 7)	[70]	204.00 (Acid red 114)	[73]	118.93 (Disperse orange 25)	[76]	250.20 (Saphranine)	[79]
	85.84 (Crystal violet)	[71]	116.27 (Acid violet 17)	[74]	49.20 (Direct red 89)	[77]		
	57.30 (Astrazon blue)	[72]	53.00 (Acid red 18)	[75]	37.33 (Direct blue 86)	[78]		
HCl	303.03 (Methylene blue)	[80]	—	—	—	—	—	—
	238.50 (Methylene blue)	[81]						
H_3PO_4	412.00 (Methylene blue)	[82]	62.18 (Acid red 27)	[85]	—	—	50.00 (Reactive orange)	[88]
	388.50 (Methylene blue)	[83]	55.40 (Acid blue)	[86]			3.92 (Reactive orange)	[89]
	355.72 (Malachite green)	[84]	8.33 (Acid blue)	[87]				[90]
							208.30 (Reactive red 23)	
HNO_3	450.50 (Methylene blue)	[81]	189.00 (Congo red)	[91]	—	—	166 (Rifafix red 3BN)	[93]
							157 (Reactive red 241)	[61]
KOH	289.26 (Methylene blue)	[94]	388.00 (Acid blue 74)	[97]	—	—	—	—
	243.90 (Malachite green)	[95]	1058.00 (Acid blue 74)	[98]				
	241.00 (Methylene blue)	[96]						
NaOH	1845.00 (Basic brown 1)	[99]	912.00 (Acid blue 74)	[100]	—	—	—	—
	1165.00 (Methylene blue)	[100]	567.00 (Acid blue 74)	[99]				
			391.00 (Acid blue 80)	[101]				
H_2O_2/thermal	91.10 (Rhodamine B)	[102]	219.00 (Acid blue 113)	[103]	—	—	11.57 (Vertigo blue 49	[9]
KOH/CO_2	441.10 (Methylene blue)	[104]	—	—	—	—	—	—
	413.52 (Methylene blue)	[96]						
	294.86 (Methylene blue)	[105]						
H_3PO_4/steam	355.72 (Malachite green)	[84]	—	—	—	—	—	—
CO_2/steam	360.00 (Methylene blue)	[106]	—	—	—	—	235.4 (Reactive black 5)	[108]
	90.9 (Methylene blue)	[107]						
$ZnCl_2$	519.00 (Methylene blue)	[109]	438 (Acid orange 7)	[92]	374 (Direct yellow 12)	[92]	176.10 (Remazol yellow)	[111]
	222.22 (Malachite green)	[54]	5.78 (Acid orange 10)	[110]			37.31 (Reactive brilliant red K-2BP)	[112]
							62.40 (Composite reactive)	[113]

(the oxygen free Lewis basic sites) and free electrons present in the aromatic rings and multiple bonds (dye molecules), thus conferring a positively charged surface to the carbon. More recently, the existence of two parallel adsorption mechanisms (the adsorption of aniline on activated carbons) has been reported by Radovic and Rodríguez-Reinoso [117], which involves the electrostatic interactions between the anilinium cations and negatively charged carbon surface groups, and the dispersive interactions between the aniline molecules and graphene layers. The observation has successfully explained the high adsorption performance of cationic dyes on basic activated carbons (even though the adsorption of cationic species on activated carbons is enhanced by the presence of surface acid groups), which attributed to the dominant role of dispersive interactions in the adsorption mechanism.

Hitherto, a notable trend in the growing research to investigate the feasibility and suitability of natural, renewable and low cost materials as alternative precursors in the production of activated carbon has been witnessed. Table 5 illustrates the most successive precursors utilized for preparation of activated carbons in dye removal processes. Of major interest, rubber wood and oil palm waste, in the forms of palm kernels, palm leaves, palm fronds, palm trunks, empty fruit bunches, palm shells, palm fibers and palm stones is rated top seat, recording a maximum adsorption capacity of 1176 mg/g for anionic dye (acid blue 264) and 1845 mg/g for cationic dye (basic blue 1), followed by rubber wood sawdust and sugar cane bagasse, which individually achieves a maximum capacity of 1456 mg/g (Bismark Brown) and 1060 mg/g (acid blue 25). The results are very promising, attributed to the relatively high fixed carbon and volatile matters content, low ash content, and high density or mechanical strengths of the precursors, associated with the inherent highly porous structures within the carbon matrix [122].

Recognizing the above phenomenon, the successful integration of activated carbons has reflected the diversification of agricultural waste, contributed effectively towards the stability and sustainable development of the agricultural-based industry. In particular, with the price of crude petroleum oil in the world market escalates to unprecedented height due to political instabilities in many oil-exporting countries and diminishing oil reserves in the world, the amount of biomass produced by an oil palm tree, inclusive of the oil and lignocellulosic materials is on the average of 231.5 kg dry wt/y [123], allowing continuous operation of the process. Its potentiality is further facilitated by the increase environmental consideration and public concern, which striving towards judicious utilization of the heavily polluted agricultural waste. Thus, a widespread and great progress of in this area can be expected in the future.

Table 5
Precursors proposed for preparation of activated carbons in dye removal processes

Precursor	Maximum monolayer adsorption capacity q_e (mg/g)		Precursor	Maximum monolayer adsorption capacity q_e (mg/g)	
	Anionic dye	Reference		Cationic dye	Reference
Pinewood	1176.00	[65]	Plum kernels	1845.00 710.10 333.33	[99] [63] [118]
Sugarcane bagasse	1060.00 391.00	[66] [101]	Rubber wood sawdust	1456.00 1250.00	[119] [62]
Fir wood	912.00	[100]	Fir wood	1165.00	[100]
Plum kernels	904.40 567.00 388.00	[63] [99] [97]	Bituminous coal	588.00	[64]
Mahogany sawdust	518.00	[67]	Cattle manure compost	519.00	[109]
Coconut-shell	509.00	[69]	Coconut-shell	450.50	[81]
Polyacrylonitrile	438.00 374.00	[92] [92]	Bamboo	441.10	[104]
Peat bog	369.80	[120]	Coconut husk	413.52	[96]
Peach stones	412.00	[82]	Waste tyre	360.00	[106]
Pistachio shells	401.30	[98]	Rubber wood sawdust	355.72	[84]
Peanut hulls	388.50	[83]	Oil palm shell	303.03	[80]
Oil palm fiber	384.62	[121]			

3. Process parameters

3.1. Effect of adsorbent dosage

Effect of adsorbent dosage is particularly the most important parameter to be considered, due to the reason that it determines the extent of decolorization and predicts the cost of adsorbent per unit of solution to be treated. Table 6 summarizes the effect of adsorbent dosage on dye adsorption capacity. Generally, at fixed dye concentrations, degree (% removal) of adsorption increases with adsorbent dose, mainly hinges on the availability of adsorption sites and the surface area, resulting in higher amount of adsorbed dye [130]. Bhatnagar and Jain [102] also indicated a dramatic positive impact of activated carbon dosage on the colour removal, with approximately a linear relationship. However, with the rises of the dosage, adsorption density deceases significantly, consequence of the unsaturation of adsorption sites during the adsorption process [131]. Similar behavior was shown for adsorption capacity (mg/g), which decreases proportional to the increase in adsorbent dosage, induced by the overlapping or aggregation of adsorption sites, leading to the depression of total adsorbent surface area and elevation in diffusion path length. At higher adsorbent doses, equilibrium time required decreases accordingly [60].

3.2. Effect of initial concentration

Initial concentration provides an important driving force for alleviating mass transfer resistances of dye molecules between the aqueous solution and solid phases. Hence, a higher initial dye concentration enhances the sorption process. This phenomenon will take relatively longer contact time (equilibrium time) [105]. The time profile of dye uptake is a single, smooth and continuous curve leading to saturation, suggesting possible monolayer coverage of dye onto surface of the adsorbent [132]. The effect of initial dye concentration on adsorption capacity is illustrated in Table 7. In most cases, at lower concentration, the ratio of the initial number of dye molecules to the available surface area is low, subsequently, the fractional adsorption become independent on the initial concentration [138]. However, at higher concentration, the available sites of adsorption becomes fewer (saturation of the sorption sites) and hence, the percentage removal of dye is dependent upon the initial concentration [134]. At fixed adsorbent doses, adsorption capacity (mg/g) increases proportional to the dye concentration, but the percentage of removal decreases, indicating residual dye concentration is higher than the initial concentration [139].

For column adsorption systems, breakpoint time decreases with the inlet concentration as binding sites are rapidly attained its saturation, evidenced that increase in concentration could modify continuous adsorption system [140]. Decreases in initial concentration (lower concentration gradient) demonstrate an extended breakthrough curve, implying that higher volume of pollutants could be treated, owing to the reduction of diffusion or mass transfer coefficient [141]. In batch adsorption systems, formation of monolayer adsorbate on the adsorbent surface is usually taking place, whereby the adsorption rate is controlled primarily by the transportation of adsorbate species from exterior (outer) sites to the interior site of the adsorbent particles [11]. Accordingly, Karaca et al. [142] reported that longer equilibrium reaching time for higher initial concentrations, could be ascribed to a significant dimerization or aggregation of monomers, resulting in lower intraparticle diffusion rate after the fast initial sorption (dye monomers and dimmers) on the external surfaces of activated carbons.

Table 6
Effect of adsorbent dosage on adsorption capacity

AC type/precursor	Dye	Type	Range (mg/L)	q_e (mg/g)	Reference
Rubberwood sawdust	Bismark brown dye	Basic	100–1200	1456.00	[119]
Parthenium biomass	Rhodamine-B	Basic	4000–20,000	7.10	[124]
Coconut husk	Methylene blue(MB)	Basic	50–500	405.00	[96]
Pongam seed shells	Acid violet 17	Acid	500–4,500	130.00	[74]
Cone biomass	Acid blue 40	Acid	400–1,000	71.63	[125]
Egg shell	Acid orange 51	Acid	500–1,500	71.94	[126]
Jute fiber carbon (JFC)	Reactive red 120	Reactive	400–4,000	11.96	[8]
PAC (Merck)	Bomaplex bed CR-L	Reactive	4,000–16,000	108.75	[127]
Silk cotton hull waste	Reactive blue MR	Reactive	250–300	12.90	[128]
Wheat shell	Direct blue 71	Direct	2,500–20,000	36.67	[129]
Orange peel	Direct blue 86	Direct	2,000–6,000	37.33	[78]
E. rigida (plant)	Direct orange 25	Direct	200–2,000	134.53	[76]

Table 7
Effect of dye concentration on adsorption capacity

AC type/precursor	Dye	Type	Range (mg/L)	Maximum monolayer adsorption capacity (mg/g)	Reference
Bamboo	Methylene blue (MB)	Basic	100–500	420.00	[104]
Parthenium biomass	Rhodamine-B	Basic	50–250	10.80	[124]
Palm kernel shell	Basic blue 9	Basic	100–700	333.33	[118]
Peat bog	Acid blue 161	Acid	50–500	369.80	[120]
Coconut coir pith	Acid brilliant blue	Acid	10–70	13.68	[133]
Cotton	Acid violet 17	Acid	60–100	47.62	[74]
Carbon slurry	Vertigo blue 49	Reactive	20–300	7.30	[7]
Coconut tree flower	Reactive red	Reactive	10–40	6.70	[8]
PAC (Merck)	Reactive black 5	Reactive	5–100	49.90	[134]
Coconut coir pith	Direct red 12B	Direct	20–100	66.00	[135]
Almond shell	Direct red 80	Direct	50–150	26.04	[136]
Wheat shell	Direct blue 71	Direct	20–250	24.50	[129]
AC (Fine Chemical)	Congo red	Direct	50–545	183.10	[137]

3.3. *Effect of contact time*

Generally, adsorption capacity and dye removal efficiency increase with prolonging the contact time [143]. Initially the amount of dye adsorbed onto the carbon surface increases rapidly, and at some point of time, the process slows down and reaches a plateau (constant value) [104]. At this point, the amount of the dye desorbing from the adsorbent is in a state of dynamic equilibrium with the amount of the dye being adsorbed onto the activated carbon. The time required to attain this state of equilibrium is termed as equilibrium time [144]. This phenomenon is attributed to reduction in immediate solute adsorption due to the lack available open sites for dye adsorption (saturation), which in turn supported film diffusion [145]. Three consecutive mass transport steps are associated with the adsorption of solute from solution by porous adsorbent. Initially the dye molecules have to first encounter the boundary layer effect, migrates through the solution (film diffusion), followed by solute movement from particle surface into interior site by pore diffusion and finally the adsorbate is adsorped into the active sites (porous structure) at the interior of the adsorbent particle [141]. The amount of dye adsorbed at the equilibrium time reflects the maximum adsorption capacity of the adsorbent under those operating conditions [80].

3.4. *Effect of solution pH*

Solution pH affects adsorption by regulating the adsorbents surface charge as well as the degree of ionization of constituents (acidic and basic compounds) present in the solution. In general, initial pH value may enhance or depress the dyes uptake rate, inter-correlated to the changes of adsorbent surface and dye chemistry [64]. Hydrogen and hydroxyl ions are adsorbed quite strongly, thus affecting adsorptive process through dissociation of functional groups on the active sites of adsorbent, subsequently leads to a shift in reaction kinetics and equilibrium characteristics [133]. Accordingly, Juang and Swei [146] have linked dye adsorption process to the existence of conjugated-bonds on the carbon surface, which are non-localized and highly active in nature. During the activation step, a substantial formation of oxygen-containing functional groups in the crystal lattice was noted, mainly the electron acceptor (carbonyl and carboxyl), thus verified the electrostatic interaction between the electron-acceptor groups with dye molecules [142]. Similarly, Ko et al. [147] has proposed the expressions for adsorbent surface acidity and basicity as Eq. (1) and (2), respectively.

$$\text{Adsorbent-OH} + \text{H}^+ \rightarrow \text{Adsorbent-OH}_2^+ \tag{1}$$

$$\text{Adsorbent-OH} \rightarrow \text{Adsorbent-O} + \text{H}^+ \tag{2}$$

In most cases, it is commonly observed that adsorbent adsorbs anions favorably at lower pH due to the presence of H^+ ions, where at higher pH, it is active for adsorption of cations due to the deposition of OH^- ions [148]. This illustrates that alkalinity enhances the adsorption of electropositive substances where acidity reduces the adsorption of positively charged dye cations due to electrostatic repulsion [149]. Lower adsorption of cationic dyes at acidic pH is associated with the presence of excess H^+ ions, competing with dye cations for the adsorption surfaces [150]. However, in the basic medium, the formation of electric double layer changes its polarity and consequently dye uptake increases. Table 8 displays the effect of solution pH on dye adsorption capacity.

Table 8
Effect of solution pH on adsorption capacity

AC type/precursor	Dye	Type	Range	Adsorption uptake of equilibrium q_e (mg/g)	Reference
Bituminous coal	Methylene blue	Basic	4–11	Increase	[10]
Apricot stone	Astrazon yellow 7GL	Basic	4–10	Increase	[70]
Bamboo	Malachite green	Basic	2–8	Increase	[151]
Peat bog	Acid blue 161	Acid	1–9	Decrease	[120]
Rice husk	Acid yellow	Acid	2–10	Decrease	[86]
PAC (Merck)	Acid orange 7	Acid	2–11	Decrease	[152]
Carbon slurry	Vertigo blue 49	Reactive	3–10	Increase	[9]
Norit ROX 0.8	Reactive red 241	Reactive	2–12	Increase	[93]
AC (Commercial)	Carmine	Reactive	3–11	Increase	[153]
Pitch fiber	Direct black 19	Direct	3–11	Decrease	[154]
Almond shell	Direct red 80	Direct	2–12	Decrease	[136]
Orange peel	Direct red 23	Direct	2–10	Decrease	[155]

Besides, the effect of pH can be described on the basis of zero point of charge (pH_{ZPC}), the point which the net charge of adsorbent is zero [156]. According to Al-Degs et al. [157], pH_{ZPC} is an index of the ability of surface (either positively or negatively charged), controlled by the pH of the surrounding solution. When solution pH < pH_{ZPC}, activated carbon adsorbent will react as a positive surface and as a negative surface when solution pH > pH_{ZPC}. Therefore, as pH is increased, surface functional groups on the carbon are deprotonated, which results in reduction of surface charge density. Meanwhile, Al-Degs [158] has revealed that activated carbon generally carries a net negative surface charge and exhibits relatively negative zeta potential values in solution for all ranges of pH (show gradually more negative values as the pH is increased), thus significantly affects adsorption processes.

3.5. *Effect of adsorbent particle size*

Adsorbent particle size plays an important role in manipulating the degree of surface adsorption. As particle size decreases, the surface area values increase proportionally [64], and as a consequence, the saturation capacity per unit mass of adsorbent increases. In particular, the large external surface area for small particles removes more dye in the initial stage than large particles, thus confirming the hypothesis [108]. Further, Hartono et al. [159] have recorded a longer equilibrium time by expanding the adsorbent particles size, indicating that intraparticle diffusion greatly influences the accessibility of dye molecules to the internal sites.

Meanwhile, it can be noted that with decreasing particle size, adsorption increases to some extent, beyond which decrease in particle size is negligible, which could not be due to any substantial increase in surface area. In such condition, it appears that dye molecules do not completely penetrate the adsorbent particle (large molecules of dyes are not able to penetrate into some of the interior pores of the particles when their size is large), or partly that dye molecules are preferentially adsorbed near the outer surface of adsorbent due to steric hindrance of large dye molecules [160]. Effect of adsorbent particle size on dye adsorption capacity is presented in Table 9. Apparently access to all pores is facilitated as particle size becomes smaller, resulting in higher adsorption. Similar trend was reported by Matthews [166]. The authors correlated the phenomenon to the inclusion of the surface area associated with pores inside the particles, thus limiting the adsorption to the external surface area and a narrow layer just below the surface.

Accordingly, Daifullah et al. [160] commented that adsorption is a surface phenomenon and as such, the extent of adsorption is proportional to specific surface area, the portion of the total surface area that is available for adsorption. Adsorbent particles size influences both adsorption kinetics and equilibrium due to the resistance to intraparticle diffusion, where the greater the particle size, the greater the contribution of intraparticle diffusion resistance to the control of adsorption kinetics [167]. As shown by Guibal and his co-workers [168], the relationship of adsorption capacity to particle size is principally depended on two criteria: (i) the chemical structure of the dye molecule (its ionic charge) and its chemistry (its ability to form hydrolyzed species) and (ii) the intrinsic characteristic of the adsorbent (its crystallinity and porosity, the rigidity of the polymeric chains, the degree of crosslinking). Moreover, in some cases, small particle sizes are not compatible with large-scale applications. In fixed bed columns for instance, small particles are inappropriate since they induce head loss and column blocking and cause serious hydrodynamic limitations.

Table 9
Effect of adsorbent particle size on adsorption capacity

AC type/precursor	Dye	Type	Range (μm)	Maximum q_e (mg/g)	Reference
Sago waste	Rhodamine-B	Basic	125–750	7.10	[161]
Bituminous coal	Methylene blue	Basic	106–500	325.00	[64]
Filtrasorb 400	Remazol golden yellow	Reactive	300–710	1111.00	[158]
Filtrasorb 400	Remazol black B	Reactive	300–710	434.00	[157]
Filtrasorb 400	Cibacron navy F-G	Reactive	412–1,050	27.00	[152]
Medium density fiberboard (MDF) sawdust	Levafix brilliant red E-4B	Acid	100–900	15.60	[153]
GAC F400	Tectilon orange 3G	Acid	355–1,400	39.60	[154]
Colemanite ore waste	Acid blue 062	Acid	30–500	34.50	[155]

3.6. Effect of temperature

Temperature has two major effects on the adsorption process. Firstly, increasing temperature is known to enhance the diffusion rate of adsorbate molecules across the external boundary layer and in the internal pores of adsorbent particle, owing to the decrease in viscosity of the solution [142]. In addition, temperature alteration will differ the equilibrium capacity of the adsorbent for a particular adsorbate (increase the tendency of deaggregation) [169]. Table 10 illustrates the effect of temperature on dye adsorption capacity. Adsorption equilibrium usually decreases with the rises in temperature, indicating exothermic nature of the adsorption reaction. Herby, reduction in adsorption capacity is contributed primarily by the enhancement of desorption step in the sorption mechanism and weakening of sorptive forces (physical bonding) between active sites on the activated carbon and the dye species, or between adjacent dye molecules on the sorbed phase [172].

With the elevation of temperature, solubility of dye molecules increases proportionally, therefore strengthens the interaction forces between solute and solvent stronger than solute and adsorbent, and hampers the adsorption processes [173]. Increasingly, rises in temperature also lead to the increase in kinetic energy between dye molecules and AC particles, consequence of the higher collision frequency between adsorbent and the dye molecules [127]. In short, temperature could affect not only the chemical potential of the material (its potential increases), the potential being a controlling factor in adsorption, but also the desorption step and consequently reversibility of the adsorption equilibrium.

3.7. Effect of multisolute competition

In wastewater purification processes, the pollutants will normally be a mixture of numerous compounds rather than a single solute. The interactions of these constituents may mutually induce or inhibit the adsorption capacity [174]. Thus, variation of adsorption capacity in a multisolute solution can be evaluated by comparing the adsorption profile from single solute solutions to multisolute solutions for each particular dye. However,

Table 10
Effect of temperature on adsorption capacity

AC type/precursor	Dye	Type	Range (K)	Maximum monolayer adsorption capacity (mg/g)	Adsorption capacity changes	Reference
Plum kernel	Methylene blue	Basic	973–1173	334.00	Increase	[170]
PAC (Merck)	Momaplex red CR-L	Basic	291–313	83.00	Increase	[127]
Apricot shell	Basic blue 9	Basic	303–323	201.61	Increase	[72]
Peat bog	Acid blue 161	Acid	298–318	370.00	Increase	[120]
Sludge	Orange II	Acid	283–333	270.00	Increase	[92]
AC(Commercial)	Eosin	Acid	303–323	99.00	Decrease	[171]
Carbon slurry	Vertigo blue 49	Reactive	298–318	7.10	Decrease	[9]
Loofa	Reactive orange	Reactive	298–333	38.31	Decrease	[88]
E. rigida (plant)	Direct orange 25	Direct	283–293	88.50	Increase	[76]
Sludge	Chrysophenine	Direct	283–333	270.00	Increase	[92]
Pandanus carbon	Congo red	Direct	303–333	8.42	Increase	[138]

such comparisons should only be made if the initial conditions are identical in single and multisolute systems [175]. Nevertheless, only very limited information is available on the competitive adsorption of dye molecules with the carbon based materials

In an effort conducted by Faust and Aly [141], adsorption capacities of each individual dye were found proportionally less with another solute, indicating the existing of competition against each other for available sites, with non-equal competition attributed to the heterogeneity of the carbon surface. Other factors may include electrical repulsion of dyes from carbon surface by other adsorbed dyes of similar charge, and attraction of dyes to each other on the surface. Hereby, dyes with the greatest ionic potential saturation value may effectively occupy most of the active sites hindering further adsorption of other dyes with possessing a lower affinity and if the sites were still unsaturated, then those having lower ionic potential would be removed in sequence [176].

Meanwhile, Allen et al. [177] in another study recorded that adsorption capacity of an individual dye decreased in the presence of a second or a third dye. They reported that reduced capacities could be the consequence of a combination of factors, including (i) interactions between dyes in solution; (ii) changes of adsorbent surface charge due to adsorption and (iii) competitive adsorption between dyes for active sites on carbon surface where displacement effects replace other dyes from the adsorption sites. In the case of adsorption isotherm, curve for single component system rises steeply at low liquid phase concentration, indicating complete adsorption with a plateau formed when the maximum adsorption capacity is achieved. However, in multicomponent adsorption (particularly multisolute adsorption systems), a less regular isotherm shape is found, possibly showing that concentration of the dye being adsorbed increased and then decreased, due to a displacement effect, which effectively displaced and replaced other dyes with lower affinity to the adsorption sites [158].

4. Adsorption isotherms

Adsorption isotherms are viable for describing the relationship between adsorbate molecules and adsorbent surface, highlighting the distribution of adsorbate molecules between the liquid and solid phases when an adsorption process reaches its equilibrium state [96]. The analysis of isotherm data by fitting different isotherm models is a significant step to determine the suitable model which can be used for design purposes. Hence, the correlation of equilibrium data using either a theoretical or empirical equation is essential for interpretation and prediction of the extent of adsorption, in deciding the maximum adsorption capacity of adsorbate for a given adsorbent [178].

In the perspective, adsorption equilibrium is a dynamic concept achieved when the rate of molecules adsorb to a surface is equal to the rate which they desorb [179]. An accurate mathematical description of equilibrium adsorption capacity is indispensable for reliable prediction of adsorption parameters and quantitative comparison of adsorption behavior for different adsorbent (or for varied experimental conditions) within any given system. Meanwhile, the distribution of dye molecules between liquid phase and adsorbent is a measure of the position of equilibrium in adsorption process and can generally be expressed by one or more series of isotherm models. Isotherm shape can also provide qualitative information on the nature of solute–surface interaction, which acts as a convenient tool to discriminate among different materials and thereby select the most appropriate one for particular applications [180].

There are several isotherm models available for analyzing experimental data and for describing the adsorption equilibrium, including Langmuir, Freundlich, BET, Toth, Temkin, Redlich–Peterson, Sips, Frumkin, Harkins-Jura, Halsey, Henderson and Dubinin–Radushkevich isotherms. The applicability of the isotherm equation is judged by correlation coefficients, R^2 values generated by the linear regression of each model [181]. Various researchers have adopted these isotherms to examine the importance of different factors on dye adsorption. However, the two most frequently used equations applied in solid-liquid adsorption systems are the Freundlich [182] and Langmuir models [183] (Table 11). Table 12 demonstrates isotherms study of adsorption of dyes on activated carbon.

Table 11
Linear and nonlinear forms of Freundlich and Langmuir isotherms

Isotherm	Equation	
	Nonlinear form	Linear form
Freundlich	$q_e = K_F C_e^{\frac{1}{n}}$	$\log q_e = \log K_F + \frac{1}{n} \log C_e$
Langmuir	$q_e = \frac{Q_o b C_e}{1 + bC_e}$	$\frac{C_e}{q_e} = \frac{1}{bQ_o} + \frac{C_e}{Q_o}$
		$\frac{1}{q_e} = \frac{1}{Q_o} + \frac{1}{bQ_o C_e}$
		$q_e = Q_o - \frac{q_e}{bC_e}$
		$\frac{q_e}{C_e} = bQ_o - bq_e$

Table 12
Isotherms study of adsorption of dyes on activated carbon

AC type/precursor	Dye	Type	Langmuir			Freundlich			Ref.
			q_e (mg/g)	K_L (dm^3/g)	R^2	K_f (mg/g)(dm^3/g^n)	n	R^2	
Waste apricot	Crystal violet	Basic	94.74	0.227	0.993	34.292	4.68	0.987	[184]
Palm fruit bunch	Basic yellow 21	Basic	327.57	0.073	0.880	30.750	1.59	0.996	[185]
Palm shell	Methylene blue	Basic	243.90	0.930	0.999	69.470	6.25	0.965	[80]
Plum kernel	Acid blue 74	Acid	388.00	0.112	0.996	162.900	6.25	0.996	[97]
Sugarcane bagasse	Acid blue 80	Acid	384.60	0.329	1.000	179.000	6.67	0.990	[101]
Sunflower seed hull	Acid violet 17	Acid	107.52	0.134	0.998	22.510	2.43	0.984	[74]
Sludge	Chrysophenine	Direct	0.19	0.024	0.900	15.000	2.05	0.940	[92]
Orange peel	Direct blue 86	Direct	33.78	0.120	0.990	7.130	2.62	0.983	[78]
Almond shell	Direct red 80	Direct	28.50	1.821	0.962	1.560	7.35	0.881	[136]
Filtrasorb 400	Remazol reactive red	Reactive	278.00	16.750	0.994	55.200	0.29	0.937	[175]
Sugarcane bagasse pith	Reactive orange	Reactive	3.48	0.538	0.964	5.105	1.96	0.973	[89]
PAC (Merck)	Reactive black 5	Reactive	58.82	0.109	0.940	5.754	1.70	0.940	[139]

4.1. Freundlich isotherm

Freundlich isotherm is the earliest known relationship describing an adsorption isotherm. It endorses the heterogeneity of surface (when more than monolayer coverage of the surface) and assumes that adsorption occurs at sites with different binding energy [50]. Freundlich equation predicts that dye concentrations on adsorbent will increase as long as there is an increase in dye concentration, where the amount of adsorbed material is the summation of adsorption on all sites [186]. Adsorption energy varies as a function of surface coverage and an unlimited supply of unreacted sites is assumed to be available.

The empirical model is generally suitable for high and middle-concentration environments and is not suitable for low concentrations because it does not meet the requirements of Henry's law [187]. However, Wong et al. [188] reported that if the whole concentration range was divided into three different regions, excellent fits to the experimental data could be observed with the Freundlich model, especially at lower concentrations. Additionally, this fairly satisfactory empirical isotherm can be used for non-ideal sorption, which the non-identical adsorption sites are not always available [164]. Based on this model, the slope $1/n$ ranging between 0 and 1 is a measure of adsorption intensity or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero. A value for $1/n$ below unity indicates a normal Langmuir isotherm while $1/n$ above one is an indicative of cooperative adsorption [189].

4.2. Langmuir isotherm

The Langmuir model is the most frequently used model for the sorption of pollutants from a liquid solution, which the Langmuir equation is typically intended for a homogenous surface [142]. It is based on a theoretical model, suggesting that monolayer adsorption occurs on a surface containing finite number of adsorption sites with no transmigration of adsorbate, that are equally available and energetically equivalent with each site carries equal numbers of molecules and no interaction between adsorbate molecules in the plane of surface [190]. Langmuir's model depends on the assumption that intermolecular forces decrease rapidly with distance and consequently predicts the existence of monolayer coverage of adsorbate at the outer surface of adsorbent. It also presumes that adsorption takes place at specific sites within the adsorbent and believes that once the adsorbate occupies a site, no further adsorption takes place at that site [104].

Theoretically, sorbent has a finite capacity for sorbate, where as saturation value is reached no further sorption can take place [191]. Basic forms of the Langmuir adsorption isotherm were found having reasonably good agreement with a number of experimental isotherms, including those having different interfaces between the two phases. Conformation of the experimental data into the Langmuir isotherm equation is analyzed according to the linear form of the Langmuir isotherm. The essential characteristics of the Langmuir equation can be expressed in terms of dimensionless separation factor R_L, defined by Hall et al. [192]:

$$R_L = \frac{1}{1+bC_o} \tag{3}$$

with b is the Langmuir constant and Co is devoted to the highest initial dye concentration (mg/L). If:

$R_L > 1$ Unfavorable

$R_L = 1$	Linear
$0 < R_L < 1$	Favorable
$R_L = 0$	Irreversible

5. Adsorption kinetics

Adsorption kinetics describes the solute uptake rate (chemical reaction), which in turn governs residence time of the adsorption process. Throughout recent years, three kinetic models have been widely used in the literature for adsorption processes, namely the pseudo-first-order kinetic model (Lagergren model) [193], pseudo-second-order kinetic model (Ho and McKay model) [194], and intraparticle diffusion model (Weber and Morris model) [195]. Parameters of the kinetic constants are identified by linearization followed by both linear and non-linear regression analysis. Table 13 concludes kinetics study of the dyes adsorption onto activated carbon.

5.1. Pseudo-first-order adsorption kinetics

When adsorption is preceded by diffusion through a boundary, the kinetics in most systems follow the pseudo-first-order rate equation. Lagergren proposed pseudo-first-order kinetic equation [193] in the form of:

$$\log\left(\frac{q_e}{q_e - q_t}\right) = \frac{k_1}{2.303}t \tag{4}$$

Even though it is proven to be effective in representing the experimental kinetic data, in many cases the first order equation does not fit well with the whole range of contact time and is generally applicable over the initial stage of the adsorption processes [196]. In some cases, despite the Lagergren model provides an excellent fit with the experimental kinetic data, it fails to predict the amount of dyes adsorbed theoretically thereby deviating from its theory.

5.2. Pseudo-second-order adsorption kinetis

Contrary to the other model, the pseudo-second-order equation predicts the behavior over the whole time of adsorption, with the adsorption (chemisorption) mechanism being the rate controlling step [134], which involves valency forces through electrons sharing or exchange (between adsorbate and adsorbent). The model has the advantage that without knowing any parameters beforehand, adsorption capacity, pseudo-second-order rate constant, and initial adsorption rate can be determined from the equation [197].

$$\frac{1}{\left(q_e - q_t\right)} = \frac{1}{q_e} + k_2 t \tag{5}$$

5.3. Intraparticle diffusion adsorption kinetics

Within the last few years, physical significance problem associated closely with the interpretation of adsorption kinetics by the pseudo-first and pseudo-second-order equations has stimulated a developing diffusion mechanisms utilizing the Weber and Morris equation, Avrami model and Elovich equation [198]. Accordingly, an empirically found functional relationship, common to most adsorption processes, is the Weber–Morris plot, which is derived from the simple diffusion and convective diffusion mathematical expressions.

Based on the model [195], the fractional approach to equilibrium changes according to a function of $(D_t/R^2)^{1/2}$, where R is the particle radius, D_t is the diffusivity of sol-

Table 13
Kinetics study of adsorption of dyes on activated carbon

AC type/Precursor	Dye	Type	Applicable model	Reference
Apricot stone	Astrazon yellow 7GL	Basic	Pseudo-second order model, Intraparticle diffusion model	[70]
Tuncbilek lignite	Malachite green	Basic	Pseudo-second order model	[95]
Palm shell	Methylene blue	Basic	Pseudo-second order model	[80]
Gingelly (sesame)	Acid red 114	Acid	Pseudo-second order model	[73]
Colemanite ore waste	Acid blue 062	Acid	Pseudo-second order model, Intraparticle diffusion model	[165]
Orange peel	Direct blue 86	Direct	Pseudo-second order model, Intraparticle diffusion model	[78]
Almond shell	Direct red 80	Direct	Pseudo-second order model	[129]
Laboratory grade AC	Congo red	Direct	Pseudo-second order model	[156]
Sugarcane bagasse pith	Reactive orange	Reactive	Pseudo-second order model, Intraparticle diffusion model	[89]
Coir dust	Remazol yellow	Reactive	Pseudo-second order model, Intraparticle diffusion model	[111]

ute within the particle at time, t, and value of I gives an idea about the thickness of the boundary layer, which the larger the intercept the greater is the boundary layer effect [11]. The initial rate of intraparticle diffusion is obtained by the linearization of curve $t^{1/2}$ vs. q_t given by [199]:

$$q_t = k_i t^{\frac{1}{2}} + I \tag{6}$$

When intraparticle diffusion is applied in the adsorption process, the plot of the square root of time vs. the uptake rate would result in a linear relationship, and intraparticle diffusion would be the rate-limiting step if the line passed through the origin.

However, deviation of the straight line from the origin exhibits the presence of multilinearity, indicating that transportation of adsorbate from solution phase to the surface of adsorbent particles is controlled either by a single or more steps (film or external diffusion, pore diffusion, surface diffusion and adsorption on the pore surface, or a combination of more than one step), illustrating that it is not the only rate controlling step [200]. Under such conditions, the first, sharper portion is attributed to the diffusion of adsorbate through the solution to the external surface of the adsorbent or the boundary layer diffusion of the solute molecules (instantaneous adsorption or external surface adsorption) [133]. The second portion describes the gradual layer adsorption stage, where intraparticle diffusion is rate limiting and the third portion is the final equilibrium stage where intraparticle diffusion starts to slow down due to extremely low adsorbate concentrations left in the solution [201]. The uptake rate might be limited by the size of adsorbate molecule, concentration of the adsorbate and its affinity to the adsorbent, diffusion coefficient of adsorbate in the bulk phase, pore size distribution of the adsorbent, and degree of mixing [202]. Usually, intraparticle diffusion is the rate-limiting step for the systems with high concentration of adsorbate, good mixing, large particle size and low affinity towards the adsorbent [201].

6. Thermodynamic parameters

Thermodynamic parameters such as ΔG (Gibbs free energy change), ΔH (enthalpy change) and ΔS (entropy change) are among the most important features involved in establishing an adsorption system. Its original concepts assume that energy cannot be gained or lost, which entropy change is the driving force [142] in an isolated system. ΔG (Gibbs free energy change), ΔH (enthalpy change) and ΔS (entropy change) for a particular system can be computed according to the thermodynamic equations in Table 14 [102].

Generally, ΔG value is the fundamental criterion of thermodynamic study, which is estimated under a reversible adsorption batch system. Negative ΔG indicated the feasibility (highly favorable) of a process and spontaneous nature of adsorption with high preference of dyes onto activated carbon [127]. With the rises in temperature, ΔG decreases accordingly, illustrating less driving force and thereby resulting in lower adsorption capacity. Whereas, ΔH (experimentally measured) is a measure of the interaction force between the adsorbate and adsorbent, giving an indication of the bonding strength [203].

Table 14
Thermodynamic expressions for an adsorption system

Expression	Equation	Parameters
Arrhenius	$\ln K_c = \ln A - \frac{E_a}{RT}$	Apparent activation energy
Gibbs	$\Delta G = -RT \ln K_c$	Free energy change
Van't Hoff	$\ln K_d = \frac{\Delta S}{R} - \frac{\Delta H}{RT}$ with $K_d = \frac{q_e}{C_e}$	Enthalpy change

Typically, ΔH for physical adsorption ranges from 4 to 40 kJ/mole, compared to chemical adsorption which varies from 40 to 800 kJ/mole [204]. In this context, positive ΔH represents endothermic nature of an adsorption and possibility of physical adsorption. In physical adsorption processes, an increase of temperature leads to the elevation of dye adsorption, thus rules out the possibility of chemisorptions. However, low ΔH shows that dyes are physisorbed onto the adsorbent while negative ΔH indicates the exothermic nature of the adsorption [9], resulting in extra energy release (interaction of dye with its adsorbent surface). As a matter of fact, ΔH values reflect a combined effect of both endothermic hydrogen bond breaking and the exothermic adsorption processes [205].

Entropy of activation is a measure of the "saddle point of energy" over which reactant molecules must pass as activated complexes. Thus, ΔS conveys whether a particular reaction proceeds faster or slower than another individual reaction. The sign of ΔS would denote the direction, for adsorption (+ΔS), and for desorption (–ΔS) [206]. Positive value of ΔS shows the affinity of activated carbon and increasing randomness at the solid–solution interface during the fixation of both dyes on active sites of the adsorbent [133]. This may be interrelated to the extent of hydration of cationic dye molecules and deaggregation of the formed dimers or aggregates. When dye molecules get absorbed on the adsorbent surface, water molecules previously bonded to the dye molecules are displaced by the adsorbate species [127], receiving more translational entropy than it is lost by the adsorbate molecules [138], get release and disperse in the solution, thus allowing the prevalence of randomness in the system. The reorientation or restructuring of water around dye molecules is very unfavorable in terms of entropy, since it disturbs the existing water structure and imposes a new and more ordered structure on the surrounding water molecules. Therefore, positive entropy change is usually linked to the increasing disorder due to reduction of water molecules surrounding dye molecules or formation of monomer upon the deaggregation processes, which increase their degree of freedom [142]. Thermodynamic parameters study of adsorption of dyes on activated carbon is presented in Table 15.

7. Conclusion

In line towards achieving the status of industrial order economy, the past ten years have seen a rapid developing of giant dyeing factories and processing industries. To date, the limited success of dye treatment technologies in field applications has raised apprehensions over the use of low-cost materials as alternatives precursors for the preparation of activated carbons as a measure in water and wastewater treatment processes. The evolution has turned from an interesting alternative approach into a powerful standard technique by offering a number of advantages. The review presented the utilization of activated carbon for removal of various kinds of dyes from aqueous solution. Process parameters such as adsorbent dosage, concentration, contact time, adsorbent particle size, pH, temperature and competition effect play a key role in determining the adsorption rate and adsorption capacity of dyes onto activated carbons. Most adsorption works adopted either the Langmuir isotherm or the Freundlich isotherm for equilibrium adsorption data correlation. In addition, the data is found fitted well with either the pseudo-first order, pseudo-second order or intraparticle diffusion models. Conclusively, the expanding of activated carbon in the adsorption science represents a potentially viable and powerful tool, leading to the superior improvement of dye pollution control and environmental conservation.

Acknowledgement

The authors acknowledge the research grant provided by the University of Science Malaysia under the Research University (RU) Scheme (Project No.1001/PJKIMIA/814005).

Symbols

A — Arrhenius factor
b — Langmuir isotherm constant, dm^3/mg
C_e — Equilibrium dye concentration, mg/L
C_o — Highest initial dye concentration, mg/L
k_1 — Pseudo first order kinetic model rate constant, min^{-1}
k_2 — Pseudo second order kinetic model rate constant, g/g min
k_i — Intraparticle diffusion rate constant, mg/g $min^{1/2}$
K_c — Reaction rate constant
K_D — Distribution coefficient
K_F — Freundlich isotherm constant, $(mg/g)(dm^3/g)^n$, related to adsorption capacity
n — Adsorption intensity
q_e — Amount of adsorbate in the adsorbent at equilibrium, mg/g
q_t — Amount of adsorbate in the adsorbent at time t, mg/g
Q_o — Maximum monolayer coverage capacities, mg/g
R — Universal gas constant, 8.314 J/mol K
R^2 — Correlation coefficient
R_L — Separation factor

Table 15
Thermodynamic parameters study of adsorption of dyes on activated carbon

AC type/precursor	Dye	Type	$-\Delta G$ (kJ/mol)	ΔH (kJ/mol)	ΔS (J/K.mol)	Reference
Tuncbilek lignite	Malachite green	Basic	5.69	43.06	167.00	[95]
Fertilizer waste	Bismark brown R	Basic	28.20	86.20	86.20	[102]
Coconut husk	Methylene blue(MB)	Basic	5.09	20.81	51.64	[96]
Coir pith	Congo red	Acid	0.15	7.71	23.55	[207]
Sunflower seed hull	Acid violet 17	Acid	7.85	152.45	0.50	[74]
Coconut coir pith	Acid brilliant blue	Acid	13.57	48.02	185.45	[133]
Peat bog	Acid blue 161	Acid	6.89	34.67	0.14	[120]
Rice husks	Safranin-T	Reactive	5.96	32.74	86.06	[208]
Loofa	Reactive orange	Reactive	3.27	–14.20	8.59	[88]
Carbon slurry	Vertigo blue 49	Reactive	20.96	–4.34	45.78	[9]
Coconut coir pith	Direct red 12B	Direct	33.94	21.85	167.85	[135]
Wheat shell	Direct blue 71	Direct	32.99	42.76	242.00	[129]
E. rigida (plant)	Direct orange 25	Direct	26.49	44.31	242.17	[76]

t — Time, min
T — Temperature, K
ΔG — Gibbs energy change, kJ/mol
ΔH — Enthalpy change, kJ/mol
ΔS — Entropy change, kJ/mol

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