Review Article

# Cationic dyes removal from wastewater by adsorptive method: A systematic in-depth review

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**ABSTRACT**

One of the most pressing environmental problems is the existence of synthetic dyes in industrial wastewaters. The presence of dyes in wastewater can cause severe problems to human beings and aquatic life, and that warrants removal from aqueous medium. Amongst the multiple strategies to treat dye-contaminated water, adsorption is considered superior because of low cost, flexibility, ease of operation and lower energy consumption. Many adsorbents, including carbon-based materials, biomaterials, nanomaterials, Schiff bases, metal-organic frameworks as well as natural and synthetic polymers, have been successfully applied in cationic dyes remediation. The current review article provides literature information about cationic dyes, various treatment strategies, principles of adsorption and recent trends in adsorbents application were discussed at length. The applicability of various isotherms and kinetic models for cationic dyes removal by diversified adsorbents is also highlighted here. Conclusions have been drawn from the literature reviewed and recommendations for future research perspectives are proposed.

## 1. Introduction

The quest for potable water has been a daunting challenge to the global community, especially in developing and underdeveloped countries. It was reported that about 800 million people still lack access to potable water. For example, only 39% of the Sub-Sahara Africa population and 46% of the Oceania population have access to potable water [1,2]. With the rapid industrialization and urbanization, the amount of wastewater bearing anthropogenic pollutants such as synthetic dyes and heavy metals emanating from the textile and other industries are doubled. The polluted water enter into aquatic systems such as rivers, lakes and other water sources accompanied by various chemical changes and eventually become one of the major sources of pollutants in water bodies [3]. These pollutants cause health complications when they exceed the tolerance limits in water. A report from the World Health Organisation (WHO) revealed that 1.8 million people die from polluted water-related illnesses [4]. The alarming

trend in water pollution has triggered scientist to proffer an effective and sustainable solution to this menace affecting the survival of human lives and the aquatic environment [5].

The demand of synthetic dyes has witnessed tremendous growth in the past decades and it was reported that approximately 1.6 million tons of various dyes are synthesized yearly [6]. Due to their good solubility, synthetic dyes are prominent pollutants of aquatic ecosystem and they are usually found in appreciable amount in industrial wastewater. An indication of the gravity of the problem is given by the fact that 2-20% of dyes that are produced are disposed directly in wastewaters [7,8]. Different industries with products such as ink, leather, cosmetics, textiles, paper and pulp, plastics, printing, paint etc., use numerous synthetic dyes to color their products and thus, effluents emanating from these industries contain dyes in sufficient quantities [9,10]. The main sources of synthetic dyes pollution are textile, paper

and pulp, tannery, paints, dye manufacturers, and dyeing industries as displayed in Fig 1.

The disposal of colored effluent is highly objectionable on both aesthetical and toxicological grounds. Dyes are resistant to biotransformation, stable to light and strongly absorb sunlight which impedes the intensity of light absorbed by the phytoplankton and aquatic plants, increasing COD and reducing photosynthesis and dissolved oxygen of the aquatic ecosystem [11,12]. Some dyes exhibit carcinogenic effects and others after degradation or transformations produce compounds such as aromatic amines, which may be mutagenic, carcinogenic or otherwise toxic towards human beings, animals and microbial population [13]. Due to increasingly strict restrictions on the content of industrial effluents, it is highly desirable and imperative to effectively eliminate dyes from water and wastewater.

In order to eliminate dyes from wastewater, a number of chemical, biological and physical techniques have been proposed over the years [14]. However, the adsorption technique has occupied a prominent place in recent time for wastewater treatment owing to its efficiency in the elimination of pollutants too stable for biological methods. Several works have been reported with the primary purpose being the investigation of adsorptive removal of different pollutants including heavy metals, pesticides, dyes, pharmaceuticals, and others (inorganic anions, hydrocarbons, etc.) [15,16]. Adsorption is influenced by many operating variables such as contact time, particle size, adsorbents dose, temperature, pH and agitation speed. The major advantage of adsorption recently became the use of low-cost materials, which reduces the operational cost [7].

Although there are a number of review articles focusing on the use of different adsorbents in the removal of dyes from aqueous medium [17-31], but most are not specifically presented with up-to-date developments on the adsorptive removal of cationic dyes from water phase. Moreover, some of the reviews are relatively old and, in the meantime numerous high performing adsorbents have been developed. Therefore, the current review was undertaken in order to provide comprehensive up-to-date information on the application of adsorption in the removal of cationic dyes from aqueous medium. Besides, the primary purpose of this review is to assemble the recent available information on the removal of cationic dyes using a wide range of potentially effective adsorbent materials. A compilation of the optimum process condition (contact time and pH) and maximum adsorption capacities along with adsorption isotherms and kinetics of the adsorbents was also presented. It is imperative to note here that the maximum adsorption capacities reported in this article provide some insight of adsorbent's effectiveness for each type of dye, and have relative meaning due to some factors,

such as different experimental conditions and method (batch versus dynamic) used in each referenced study. The reader is greatly encouraged to consult the original research articles for information on specific set of conditions.
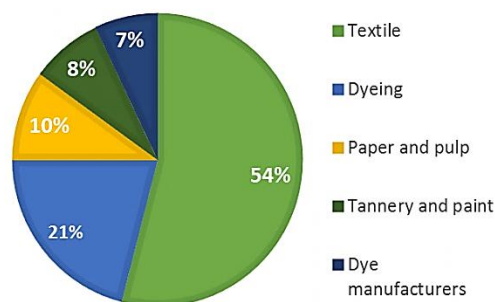


Fig. 1. Major industrial sources of synthetic dyes pollution in aquatic environment [33].

### 1.1. Categorization of Dyes

Dyes exhibit substantial structural diversity and can be categorized in several ways such as on the basis of their sources (e.g natural, synthetic), chemical structure (e.g azo, triphenylmethane, anthraquinone, indigo) and domain of usage or application to the fiber type (e.g reactive, disperse, direct, mordant, vat, sulphur, metal complexes, pigments, chromic) [27,29,32]. The classification on the basis of application is depicted in Table 2. Dyes can also be classified on the basis of their solubility: soluble dyes which comprise of basic, acid, reactive, direct, metal complex and mordant dyes; and insoluble dyes which include sulfur, vat, disperse and azo dyes [8,33,34]. Apart from the above, dyes are also grouped based on their particle charge upon dissolution in aqueous solution such as anionic (direct, acid, and reactive dyes), cationic (all basic dyes), and non-ionic (vat and dispersed dyes) [35-37]. The general classification of dyes is depicted in Fig 2.

### 1.2. Cationic Dyes

Cationic dyes (also called basic dyes) are a group of water soluble dyes that carry localized or delocalized positive charge in their molecules and produce colored cation in solution [38]. They include varying chemical structures based on substituted aromatic groups [39]. The major chemical classes of cationic dyes are: triarylmethane, acridine, diazahemicyanine, cyanine, hemicyanine, oxazine and thiazine [8,18,32]. Cationic dyes are stable due to the presence of aromatic and various functional groups which in turn render recalcitrant effect of the dyes [40]. Among all commercially available synthetic dyes, cationic dyes are the brightest class of soluble dyes used by the textile industry [41]. They are extensively employed in wool, acrylic, silk and nylon dyeing. This group of dyes is considered as toxic colorants and can cause harmful effects such as allergic dermatitis, skin irritation, mutations and

cancer [42]. Cationic dyes have great tinctorial strength where obvious coloration can be observed at concentration as low as 1ppm [43]. They adsorb readily and effectively on the negatively charged surface of adsorbents by electrostatic attraction. However, adsorption efficiency will differ depending on the surface area and functional groups present on the adsorbent surface [44]. Common cationic dyes that have been widely used as model pollutants in dye adsorption studies include methylene blue, crystal violet and malachite green. Their general properties is displayed in Table 1. The aforementioned cationic dyes are the focus of the present review.



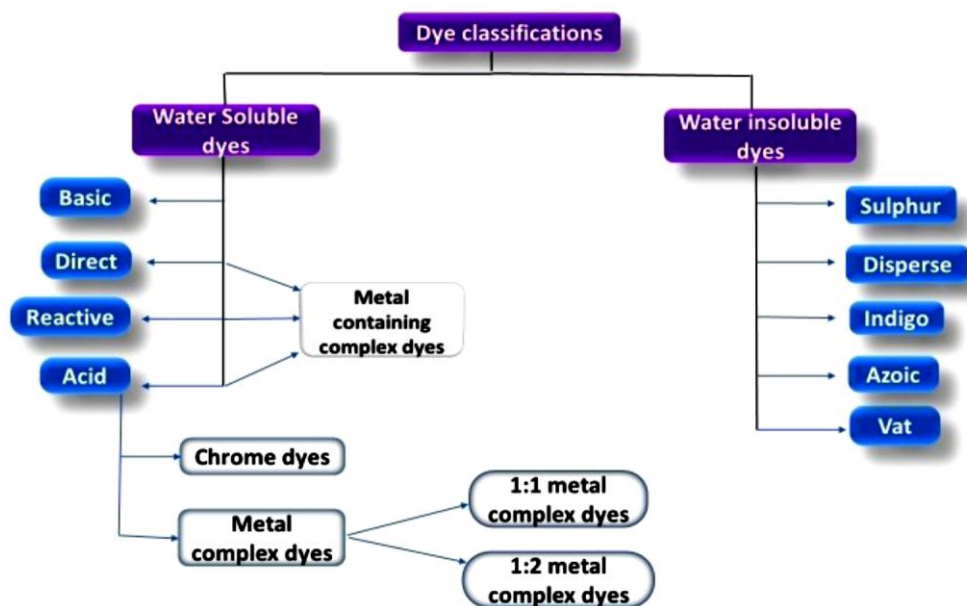Fig. 2. Categorization of dyes [34].

Table 1. General properties of the target cationic dyes
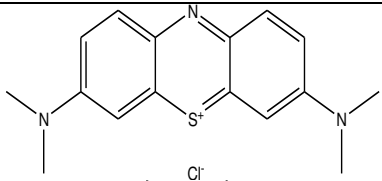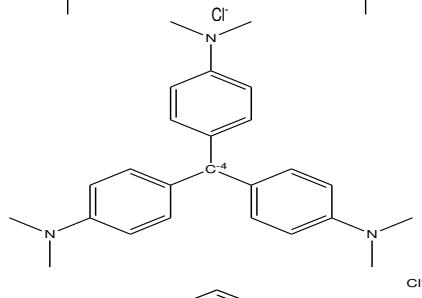
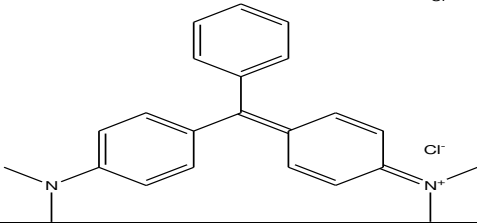| Cationic dyes | Chemical structure | Properties |
|---|---|---|
| Methylene Blue |  | $C_{16}H_{18}N_3SCl$ 319.85 g/mol $\lambda_{max} = 662$ nm |
| Crystal violet |  | $C_{25}H_{30}N_3Cl$ 407.98 g/mol $\lambda_{max} = 590$ nm |
| Malachite green |  | $C_{23}H_{25}N_2Cl$ 364.92 g/mol $\lambda_{max} = 618$ nm |

Table 2. Various classes, solubility, applications and toxicity of dyes [18,21]

| Dye class | Solubility | Applications | Toxicity | Example |
|---|---|---|---|---|
| Acid | Water soluble | Printing ink, cosmetics, fibres, polyurethane | Skin irritations and burns | Acid Black 1 |
| Basic | Water soluble | Medicine, inks, silk, modified nylon, cotton and wool | Cancer, allergic dermatitis, mutations | Basic Violet 10 |
| Direct | Water soluble | Leather, nylon, cotton, paper and silk | Bladder cancer | Direct Blue |
| Disperse | Water soluble | Cellulose, polyester, cellulose acetate, acrylic fibres and plastic | Splenic sarcomas, bladder cancer | Disperse Red 4 |
| Reactive | Water soluble | Cotton, nylon, cellulose, silk and wool | Asthma, rhinitis | Reactive Red 120 |
| Vat | Water insoluble | Polyrster-cotton, rayon and silk | Skin irritation, severe burns | Vat Blue 35 |
| Sulphur | Water insoluble | Polyamide fibres, rayon, silk and cotton | Cancer | Sulphur Black 1 |
| Solvent | | Waxes, lubricants, oil, fats, gasoline, ink and plastics | breathlessness | Solvent Red 26 |

### 1.2.1. Methylene Blue

Methylene blue (MB), (also referred to as swiss blue) is a basic cationic dye that belongs to the thiazine group and has the chemical formula $C_{16}H_{18}N_3SCl$. It was first prepared in 1876 by Heinrich Caro of Badische Anilin and Soda Fabrik as an aniline-based synthetic dye for staining cotton to be used in the textile industry [22,45]. Due to its high intense color even at low concentration, MB is used in paper coloring, cottons and wool dyeing, solar cells and as temporary hair color [44,46-48]. It is also used to stain and inactivate microbial species. Furthermore, MB was reported to be antidote for cyanide and carbon dioxide poisoning [49]. It also find application as an antiseptic and for other medical purposes [50]. However, MB has drawn attention due to it deleterious effect on humans and environment. Acute exposure to it may cause high blood pressure, vomiting, increased heart rate, shock, cyanosis, faecal discoloration, discoloration of urine, gastrointestinal pain, headache, diarrhea, jaundice, quadriplegia, fever, heinz body formation, diziness and tissue necrosis in humans [9,50]. It may cause eye burns which may lead to permanent injury of the eyes of human and animals. On inhalation, it may induce brief periods of rapid or difficult breathing, while ingestion through the mouth produces a burning sensation and may cause nausea, methemoglobinemia, profuse sweating and mental confusion [7].The negative consequences from MB pollution to human underscores the need to remove it from wastewater before releasing in to the receiving waters.

### 1.2.2. Crystal Violet

Crystal violet (CV), also known as gentian violet belongs to a category of intensely colored compounds called triarylmethane dyes and has the chemical formula $C_{25}H_{30}N_3Cl$. CV was first prepared by German chemists Kern and Caro and it involved the reaction of dimethylaniline with phosghene to give 4,4'-bis(dimethylamino)benzophenone as an intermediate. This was then reacted with additional dimethylaniline in the presence of phosphorus oxychloride and hydrochloric acid [51]. The dye can also be prepared by the condensation of formaldehyde and dimethylaniline to give a leuco dye. Then, this colorless product is oxidized to the colored cationic form using a suitable oxidizing agent [52]. CV is one of the most widely used coloring agents in different industrial treatments [53]. It is commonly used to colorize diverse products such as detergents, paper, fertilizer, and leather [54]. CV is extensively employed in textile dyeing, dermatology and veterinary fields [55]. It also finds application in the manufacture of paints and printing inks [56]. It is used as an additive to poultry feed to inhibit propagation of mold, intestinal parasites and fungus [57,58]. CV is a protein dye and hence it is used as an enhancer for bloody finger prints. In the medical community, it is used as a histological stain in Gram staining for classifying bacteria [59]. It also has antifungal and antibacterial properties [60]. However, CV is very poisonous in nature and extremely harmful for ecosystem if it reaches [61]. It is carcinogenic and mutagenic dye that

pose great danger to human health [59]. It is highly toxic to mammalian cells and if absorbed in harmful amounts through skin, it can cause skin irritation and digestive track irritation [62,63]. It can cause vomiting, eye irritation, jaundice, heart disease, quadriplegia and cyanosis in humans [44]. In extreme cases it may lead to respiratory and kidney failure [56,64]. For this reasons, CV dye even at the level of 1 ppb is not desirable in the environment and should be removed [65].

### 1.2.3. Malachite Green

Malachite green (MG), (also known as Basic Green 4) is a water-soluble cationic dye belonging to the triphenylmethane family and has the chemical formula $C_{23}H_{25}N_2Cl$. MG is prepared via a two-step reaction. The first step involve the preparation of leuco malachite green which is achieved by the condensation of benzaldehyde and dimethylamine. In the second step, malachite green dye is obtained by the oxidation of the colourless leuco compound into the corresponding malachite green cation [66]. Among other dyes, MG is the most widely used raw material as a colorant in several industrial applications [67]. MG has been extensively used in dyeing of wool, leather, silk, paper, jute, cotton, pharmaceuticals and also in distilleries [22,44]. It is also broadly used as a bactericide, fungicide and parasiticide in several aquaculture industries worldwide [68]. It is also find application as a food coloring agent, food additive, a medical disinfectant, and anthelminthic [30]. However, the hazardous effects of MG have been reported by numerous studies. It is extremely hazardous to mammalian cells and has been identified as a liver tumour promoter [67]. MG causes toxicity to respiratory system and damages fertility systems in humans; and is a well-known carcinogenic, mutagenic, and teratogenic substance [22]. Moreover, MG's resistivity to light and reduction due to oxidizing agents are very high [69]. It inflicts lesions on the lungs, skin, eyes, and bones. MG also causes damage to the liver, brain, spleen, kidney and heart [30]. Despite a large amount of available toxicity data, the dye is still used widely in aquaculture and other allied industries. Therefore, the removal of MG from effluent before been disposed into the waterways is a management serious concern [70].

### 1.3. Removal Technologies for Dyes in Aqueous Environments

The approaches generally employed to treat dye effluent can be grouped into three categories: (i) chemical (ii) physical and (iii) biological processes [32,71]. Some of the dye remediation technologies lying in the aforementioned categories include membrane filtration [72], photo-Fenton processes [73], coagulation [74], biodegradation [75], sono-chemical degradation [76], ultrafiltration [77], advanced oxidation process [78], ion exchange [79], precipitation [80], ozonation [81], reverse osmosis [82],

microbial reduction [83], electrolytic methods [84], photocatalytic degradation [85] and adsorption. However, the selection of wastewater treatment technique is a difficult task involving the consideration of several factors which include, reliability of process equipment, available space for the construction of treatment facilities, desired finished water quality, waste disposal constraints, and capital and operating costs [71,86]. The merits and limitations of various techniques employed for the treatment of dye-laden effluent are summarized in Table 3. Among various available water treatment technologies, adsorption process is considered superior because of excellent efficiency, flexibility, low cost, ease of operation, insensitivity to toxic pollutants, lower energy consumption, convenience, and simplicity of design. Moreover, this technique does not result in the generation of any toxic material [9,21,71].

## 2.    Principles of Adsorption Process

Although certain phenomenon related to adsorption were known since antiquity, the earliest known quantitative studies were reported by C.W. Scheele in 1773 on the uptake of gases by charcoal. This was followed by Lowitz' observations in 1785, who employed charcoal for decolorization of tartaric acid solutions. Larvitz in 1792 and Kehl in 1793 observed similar phenomenon with vegetable and animal charcoals, respectively [87,88]. However, the term 'adsorption' as distinct from 'absorption' was proposed by Emil du Bois-Reymond in 1881, but introduced into the literature by Heinrich Kayser [89,90]. Ever since then, the adsorption process has been extensively used for the removal of solutes from solutions and gases from air atmosphere.

Adsorption, strictly defined, is a surface phenomenon that involves the adhesion of atoms, ions, or molecules from a gas, liquid, or dissolved solid onto a surface. The substance that accumulates at the surface is called an adsorbate while the material upon whose surface the adsorption occur is called an adsorbent. Adsorption, similar to surface tension, is a consequence of surface energy that is commonly used to remove substances from liquid phases and can be grouped into two types: physical adsorption (physisorption) and chemical adsorption (chemisorption). Physical adsorption is characterized by weak attractive forces between adsorbate molecules and adsorbent and thus reversible in most cases [91]. In physisorption, adsorbates may attach on the adsorbent through van der Waals forces, electrostatic attraction, ion exchange, hydrogen bonds, Π–Π interaction, dipole–dipole interaction, and hydrophobic hydration, etc. [27,92].

Table 3. Merits and demerits of various techniques used for the removal of dyes [9,14,20,27]

| Techniques | Merits | Demerits |
|---|---|---|
| Photocatalysis | No sludge production, economically favorable | Long durational time, toxic by-products |
| Ultrafiltration | Low energy consumption | Need for chemicals to induce aggregation |
| Biodegradation | Low operational cost, publicly acceptable treatments | Relatively slow process, maintenance and nutrition requirements |
| Coagulation-flocculation | Simple and cost effective, complete decolorization | Handling and disposal issues, bulk sludge generation |
| Ozonation | No sludge formation, fast | Formation of oxidative by-products, extremely short half-life |
| Electrochemical process | Rapid and selective process, high efficiency | Energy expensiveness, economically unattractive |
| Ion exchange | Adsorbent regeneration, no sludge production | Highly pH sensitive, high operational cost |
| Advanced oxidation process | Rapid process, high efficiency | High operation and maintenance cost, undesirable by-products formation |
| Membrane separation | High efficiency | Economically unattractive, short lifetime |
| Adsorption | Simple and highly effective, wide applicability | High cost of some adsorbents |

Chemical adsorption on the other hand proceed through the formation of strong chemical bond between adsorbate species and adsorbent, which is generally due to the sharing or transfer of electrons and thus substances chemisorbed on solid surface are hardly removed because of stronger forces at stake [93]. Under favorable conditions, both processes can occur concurrently or alternatively [90]. As a standard, an isosteric heat of adsorption with magnitude between 5 and 40 kJ/mol signifies physisorption as the dominant adsorption mode, while one between 40 and 125 kJ/mol indicates chemisorption [94]. The type and nature of adsorbent and its functional groups determine the mechanism of dyes abstraction. However, some studies have postulated that adsorption of cationic dyes by different adsorbents proceed majorly through electrostatic attractions [95].

In practice, adsorption is performed as an operation, either in batch or continuous mode, in a column packed with porous adsorbents [91]. Batch adsorption studies provide vital information on the application of adsorption to the removal of specific pollutant constituents. In contrast, continuous column (dynamic) studies provide the most practical application of the adsorption process in wastewater treatments. As highlighted in some studies, the dynamic adsorption process is more applicable and suitable in real water treatment industries due to its low operating cost and the ability of columns to adapt to versatile processes. While batch reactors are easy for researchers to use in laboratory studies, they definitely are less suitable for industrial applications [96]. Most of the reported studies on the adsorption of cationic dyes by various adsorbents were performed in batch mode techniques. Fig. 3 depicts the batch adsorption treatment in a pictorial form. Readers interested in detailed information about the historical aspects, theory and application of adsorption should consult critical reviews by Swenson et al. [89] and Dabrowski [90].
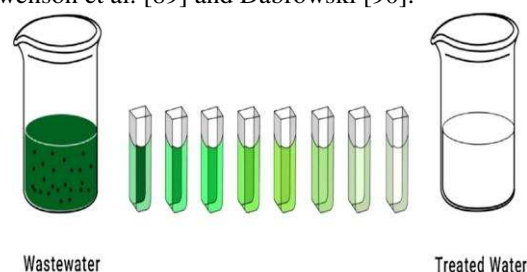


Fig. 3. Pictorial representation of adsorption treatment [97]

## 2.1. Influence of Adsorption Factors on Cationic Dyes Removal

There are many operational parameters that significantly influence the capacity of potential adsorbents and overall efficiency of the adsorption process including solution pH, contact time, adsorbent dose, coexisting ions, initial dye concentration, and temperature. The optimization of these experimental parameters is one of the key objective of adsorption studies as it can help in the development of large-scale treatment process for dye-bearing effluent [9,19]. The subsequent sections will briefly discuss some of the common parameters.

### 2.1.1. Influence of Contact Time

Equilibrium time is a very crucial element to investigate when considering an economical wastewater treatment application. The faster the equilibrium time, the lesser are the operational costs that favor the adsorbent for industrial-scale application [98]. Generally, there are three phases for the adsorption process as follows: in the initial stage, removal efficiency increases at a rapid rate due to the accessibility of a large number of vacant surface-active adsorbing sites and after the lapse of time, the adsorption rate gradually slows down due to a reduction in the number of free-active adsorbing sites and the molecule have to traverse farther and deeper into the pores and thereafter adsorption process attains plateau state (equilibrium phase) [96]. Gupta and Lataye [50] have reported that the findings of the influence of contact time on CV and MB removal using sawdust activated carbon revealed a rapid rate of adsorption in the first 15 min, and the equilibrium state was reached after 60 min of exposure time. Other comparable results have been reported for the adsorption of MG onto coconut shell activated carbon [67] and biochar [99].

### 2.1.2. Influence of Solution pH

The solution pH is a critical factor that govern the adsorption process as it affects not only the surface charge of the adsorbent, but also the degree of speciation and ionization of the dyes during adsorption [100,101]. In fact, pH can also influence the structural stability of dyes and therefore the intensity of their color (e,g transforming a dye to a leuco-structure under basic conditions) [102]. It is instructive to note that cationic dyes exist in the form of positively charged ions in aqueous environment. Therefore, as charged species, the rate of their adsorption onto the surface of the adsorbent is primarily controlled by the surface charge on the adsorbent, which in turn is influenced by the solution pH. In general, when the medium has high pH, the adsorption of cationic dyes on most adsorbents will increase because positive charges on the dye ensured that they are attracted to anionic surface of adsorbent through electrostatic attraction [18,29,32].

Besides, the electrokinetic properties of the adsorbent surface can be quantified by an important parameter, known as pH of zero-point charge ($pH_{zpc}$) [27]. It is defined as a point at which the adsorbent surface has net electrical neutrality [103]. $pH_{zpc}$ plays an important role in determining the adsorption ability of a surface as it predicts how easily an adsorbent can bind potentially harmful ions [104]. If the pH of the medium is greater than $pH_{zpc}$, the adsorbent surface functional groups are deprotonated and acquire a negative charge and hence favor the uptake of cationic dyes. Conversely, for solution pH less than $pH_{zpc}$ value, the adsorbent surface bears a positive charge due to the protonation of the surface functional groups and thus capable of repelling cationic dyes [60]. Abbas et al. [52] investigated the adsorption of CV onto commercial activated carbon and they found that pH of zero point charge of the activated carbon was 5.23, while the maximum adsorption capacity was observed at pH 11, in other word $pH > pH_{zpc}$. In another study, Fabryanty et al. [65] investigated the influence of pH on the adsorption of CV onto bentonite, aliginate and composites. The findings showed that maximum uptake of CV were achieved at pH 4.1, 8.9 and 8.0 respectively. The reason for this outcome was attributed to the optimum pH values been greater than the $pH_{zpc}$ values of each of the respective adsorbent.

### 2.1.3. Influence of Solution Temperature

The effect of temperature on adsorption is another noteworthy factor because it is an indicator of the thermal character of the adsorption, i.e, whether it is an endothermic or exothermic process [29]. If the amount of dye adsorbed per unit mass of adsorbent increases with increasing temperature then the adsorption is an endothermic process. This may be ascribed to the increase in kinetic energy of the dye molecules which enhance their collision frequency and mobility within the pores of the adsorbent [105]. On the other hand, the decrease in the amount of dye adsorbed per unit mass of adsorbent with increasing temperature is a signifier of an exothermic process. This is attributed to the weakening of the adsorptive forces between the active sites on the adsorbent and dye species, and also between adjacent dye molecules on the adsorbed phase [106]. Collins et al. [107] studied the adsorption of CV on an acid modified clay. They observed that the amount of CV adsorbed on modified clay increases with temperature which is valid for an endothermic process. Ahmad et al. [108] investigated the adsorption of MG by activated carbon prepared from rambutan seed and they concluded that the adsorption capacity increased with increase in solution temperature. Umoren et al.

[109] studied the adsorption of MB from industrial effluent using polyvinyl alcohol (PVA) and they found that the adsorption of MB decreased with increase in temperature, indicating an exothermic process.

### 2.1.4. Influence of Initial Dye Concentration

The wastewater from various industrial operations may have different concentration of dyes. The initial concentration is one of the key parameter that influence the adsorption kinetics and provides driving force for alleviating mass transfer resistances of dye molecules between the aqueous solution and solid phases [110]. Therefore, a lower initial dye concentration will take a relatively shorter contact time. In an experiment performed for the investigation of kinetics of MG adsorption from aqueous solution using desert date seed shell, Yunusa et al. [111] have proved that the contact time required for the attainment of equilibrium was dependent on initial dye concentration. For the initial concentrations of 20-40 mg $L^{-1}$, equilibrium was attained around 30 min. However, for dye solutions of higher concentration (60-100 mg $L^{-1}$), equilibrium time of 60 min was required. The variation in the contact time required to attain equilibrium was attributed to the fact that the dye molecules have to first overcome the boundary layer effect and then diffuse into the pores of the adsorbent. Therefore, MG solutions with higher initial concentration would take relatively longer time to attain equilibrium due to the higher amount of adsorbate molecules that will diffuse from the bulk liquid into the pores of the adsorbent. Similar behavior was shown for the removal of MG using walnut shell [112] and teak leaf litter [113], adsorption of CV from aqueous solution using spent tea leaves [63], and removal of MB from aqueous solution using groundnut shell activated carbon [114].

### 2.1.5. Influence of Adsorbent Dose

The effect of adsorbent dosage gives a clue about the effectiveness of an adsorbent and predicts the price of adsorbent per unit adsorbate solution to be treated. It has been generally observed that the percentage of adsorption increased with increase in the adsorbent dose but, the amount of molecules adsorbed per unit mass of adsorbent decreased [96]. The increase in the percentage adsorption with increase in adsorbent amount was ascribed to the increase in the number of adsorption sites and thus enhances the overall surface area of the adsorbent. The reverse trend with adsorption capacity was attributed to the fact that many adsorbent sites remain unoccupied, and ultimately, the adsorption could not reach saturation [115]. A similar observation was reported in the literature dealing with the removal of MB by apple peels [116], adsorption of CV using different materials [117,118]

and the removal of MG from aqueous solution using pulverized teak leaf litter [113].

### 2.1.6. Influence of Ionic Strength

The industrial effluents are normally associated with higher salt concentration, and influence of ionic strength are of some importance in the study of dye adsorption onto adsorbents [119]. Theoretically, when the electrostatic forces between the adsorbent surface and adsorbate ions were attractive, an increase in ionic strength will decrease the adsorption capacity. Conversely, when the electrostatic attraction was repulsive, an increase in ionic strength will increase adsorption [120]. Chieng et al. [121] reported that the extent of MG adsorption using chemically modified breadnut peel was sensitive to changes in salt concentration ($KNO_3$). The findings showed that the presence of 0.001 M $KNO_3$ was able to cause a reduction in the removal of MG by 65%. In another study, Shee et al. [122] investigated the influence of ionic strength on MB adsorption using husk/polyactide blended films. They observed that the amount of MB adsorbed decreased with increase in salt concentration (NaCl). They attributed this to competition for available active sites between the cationic dye ions and the positively charged ions being introduced. Furthermore, $Na^+/K^+$ (aq) ions being smaller in size would easily access anionic sites on surface of adsorbent compared to the larger cationic dye ions. These observations are consistent with those reported previously by other researchers for the adsorption of cationic dyes [123,124].

### 2.2. Multi-Component Adsorption

Within the literature, numerous research articles have focused on single component adsorption of dyes onto various adsorbents. However, in real industrial effluent, like textile industries, the wastewater usually contain a mixture of several dyes rather than a single one [14]. The presence of other pollutants in the real effluent may influence the adsorption behavior of the targeted dye on the adsorbent. In general, three different types of interactions are possible between the adsorbate molecules in a multicomponent solution such as if the adsorption of the adsorbate decrease when there are additional adsorbates present in the mixture (antagonism), the adsorption of the adsorbate increase when there are other adsorbates present in the mixture (synergism), and if the multicomponent mixture has no effect on the adsorption of each adsorbate (non-interaction) [125]. It is, therefore, imperative to study the interference in terms of antagonistic and synergetic effect of other pollutants on the adsorption of a target dye in order to better understand how they behave in the natural aquatic environment. Unfortunately, up to date, only a limited data is available on multicomponent dye adsorption [22].

Recently, the adsorption of MG and MB by Thiourea-modified polymer adsorbent in the binary system has been reported by Adeyi et al. [126]. The comparison delineates antagonistic interaction between the MG and MB dyes, which mutually hindered the adsorption of both dyes on the polymeric adsorbent. They also observed that MG is adsorbed preferentially over MB in the binary system, viz. 150 mg g$^{-1}$ against 124 mg g$^{-1}$ at equilibrium. The factors that influence the preference of adsorbent for different types of adsorbates may be linked to the characteristics of the active sites (functional groups and surface properties), the solution chemistry (pH and ionic strength) and the properties of the adsorbates (molecular structure, molecular weight, solubility and ionic nature) [127]. In another study, Abd-Elhamid et al. [128] investigated the removal of MB and CV dyes from an aqueous binary system using local soil clay as affordable adsorbent. It was found that maximum removal of MB and CV up to 47.82 and 35.71 mg g$^{-1}$, respectively was achieved by the adsorbent. Meanwhile, Olajire et al. [129] investigated the adsorption of MB in single dye system (SDS) and in ternery dye system (TDS) comprising of MB, Congo red and methyl orange onto formaldehyde-treated melon husk (FMH). The results showed that the presence of other components in TDS reduced the amount of MB adsorbed notably due to competitive effect.

# 3.  Application of Various Adsorbents for Cationic Dyes Removal

Some studies have revealed that many factors influence the adsorption process but worth to note is that the physical and chemical characteristics of the adsorbent can have significant impact on the adsorption capacity, rate and mechanism responsible in the removal of the pollutant from the media [130]. For instance, low surface area for some adsorbents leads to low adsorption capacity. A variety of adsorbents with wide-ranging characteristics including activated carbon, polymers, resins, mesoporous silica, clay, graphene oxide, nanomaterials, biomaterials and metal-organic frameworks have been investigated for pollutants removal [23,92]. Among these adsorbents, activated carbon has been widely applied. However, high production and regeneration cost are the major limitations of this adsorbent [131]. Therefore, the attention has been shifted towards the development and utilization of new generation of adsorbents with high efficiency and capacity.

In recent years, a large amount of newly modified adsorbents, composite adsorbents or multi-functional materials have been prepared and

examined as potential wastewater adsorbents. However, the selection for the most appropriate adsorbent would be based on some desirable properties such as: (i) the low-cost coupled with the ease of operation (ii) faster binding kinetics while using the minimum dosage concentration (iii) appreciable properties such as porosity, large surface area, high adsorption capacity, easy availability, reuse potential, likelihood of regeneration, high selectivity, mechanical stability, industrial-scale use etc. and (iv) the environmentally-friendly nature of each adsorbent [7,19,27]. Therefore, careful consideration of these properties is imperative for appraising adsorptive performance of adsorbents.

## 3.1. Carbon-based Adsorbents

The use of carbon-based adsorbents for the removal of pollutants from wastewater have received significant interests over the past decade. This is attributed to their unique properties and adjustable surface chemistry. Carbon-based materials include activated carbon, biochar, graphene and its derivaties, carbon nanotubes and carbon aerogels. Fig. 4 displays the different forms of carbon materials employed as adsorbents for wastewater treatment. This review will however be limited to only the use of activated carbon, biochar and graphene as adsorbents for cationic dyes removal from water systems.
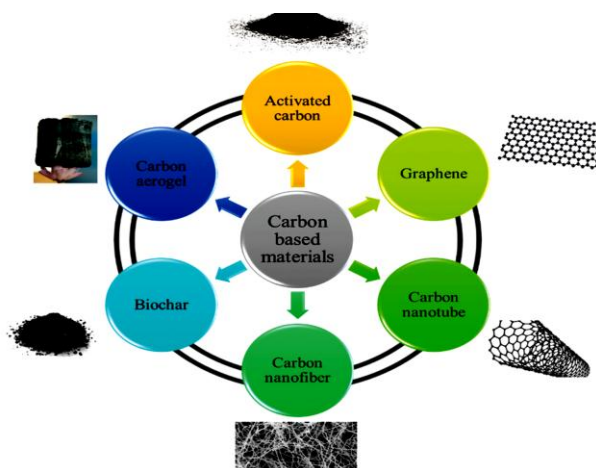


Fig. 4. Different forms of carbon materials employed as adsorbents for water treatment [131]

### 3.1.1. Activated carbon

Activated carbons (ACs) are known as very efficacious adsorbents due to their high surface area, controllable pore structure, high adsorption capacity, high thermal stability and a high degree of surface reactivity, which can be appropriately modified by physical and chemical treatments to enhance the extent of a given adsorption process [28]. AC substances can be available in the form of either

granular (GAC), powder (PAC), fibrous (FAC) and cloth (CAC). Among the various forms of AC, the powdered form is the one conventionally used in dye removal due to its larger surface area than the other forms. AC can be produced from any carbonaceous materials and are commonly classified as; (i) commercial (coal-based) activated carbons (CAC) and (ii) waste-based activated carbons (WAC) [21,30]. Coal-based ACs have been used by various researchers to successfully remove cationic dyes from wastewater. For example, Niu et al. [132] prepared CACs from three long flame coals with different ash and volatile matter composition. They concluded that all the three CACs showed good adsorption efficiency towards MB. However, the major drawback with CAC is that coal itself is a fossil fuel and therefore an expensive non-renewable resource which might eventually cease to exist. To overcome this, AC nowadays is prepared from renewable, low-cost and locally available resources like waste materials. The preparation of the AC is normally performed by the carbonization of the raw material in the absence of oxygen (pyrolysis), followed by the activation of the carbonized product, which is done either chemically or physically as highlighted in Fig. 5.



Fig. 5. Scheme of the process of preparing activated carbon

The application of waste-based ACs as dyes abstractors has been a subject of intense research. Indeed, this can be judged by the publications discussed in the present review. Tuli et al. [133] successfully synthesized activated carbon from tea-waste by chemical activation method using three activating agents $H_3PO_4$ (H-AC), KOH (K-AC), and $ZnCl_2$ (Z-AC). The synthesized materials were employed for the removal of MB from aqueous solution. The adsorption capacity of H-AC was found to be higher than those of K-AC and Z-AC, and MB removal efficiency was around 98% for the adsorbent dose of 0.15 g $L^{-1}$. Yunusa et al. [134] fabricated three different activated carbons from desert date seed shell using NaOH, $H_3PO_4$, and $ZnCl_2$ as activating agent. The prepared ACs were tested for the removal of CV and MG dyes from simulated wastewater. It was found that the AC prepared using NaOH offered the best adsorption performance with removal efficiencies of 91.10 and 99.15% for CV and MG, respectively. Han et al. [135] prepared activated carbon with large surface area (1542 $m^2$ $g^{-1}$) and total acid density (2.47 mmol $g^{-1}$) by thermal activation of phosphoric acid treated eucalyptus residue, and was successfully utilized for simulated dye wastewater treatment where it

exhibited high MB adsorption capacity (977 mg $g^{-1}$). Jayganesh et al. [136] utilized eco-friendly carbon prepared from Sargassm wightii seaweeds for the efficient removal of CV from aqueous solution. Results revealed optimum values for dye removal to be; pH value of 7, dose of adsorbent as 0.2 g, agitation time of 60 min, temperature 30$^{\circ}$C and concentration to be 80 mg $L^{-1}$. Li et al. [137] demonstrated the ability of activated carbons derived from ashitaba waste and a walnut shell to remove MB and congo red dyes from aqueous solution. Results of the experimental adsorption data revealed that both adsorbents were effective in the removal of the dyes. Table 4 summarizes previous studies on the adsorption of cationic dyes by ACs prepared from waste materials.

In addition to using the as-prepared AC, research efforts have been recently geared toward improving and diversifying the AC surface functionality via re-activation, modification, and functionalization [92]. Generally, van der Waals forces dominate the adsorption processes of non-modified carbon for the removal of organic compounds from aqueous solution [138]. Surface modification or functionalization can introduce other dominating adsorption forces such as electrostatic interaction, H-bonding and hydrophobic bonding. In recent studies, native ACs were modified to improve their adsorption capacity toward some pollutants. For example, El-Shafey et al. [139] prepared acidic, basic and hydrophobic AC and utilized them for MB removal. Acidic AC was prepared from date palm leaflets using KOH activation followed by nitric acid oxidation to produce oxidized activated carbon (OAC) which possesses acidic surface. Basic activated carbons (BACs) were synthesized via surface functionalization of OAC using ethylene diamine (EDA) and propylene diamine (PDA) producing basic activated carbons BAC-EDA and BAC-PDA, respectively. Hydrophobic activated carbons (HACs) were also synthesized via OAC surface functionalization using ethylamine (EA) and aniline (AN) producing hydrophobic activated carbons HAC-EA and HAC-AN, respectively. It was found that AC surface area was high (823 $m^2$ $g^{-1}$) with microporosity domination, however, after oxidation and surface functionalization, both the surface area and surface microporosity decrease tremendously. It was observed that despite possessing a very low surface area, HAC-EA exhibited the best performance in MB removal in terms of equilibrium and kinetics.

Elsewhere, the improved adsorption of MB on AC modified by three surfactants was investigated by Kuang et al. [140]. Anionic surfactants; sodium lauryl sulfate (SLS) and sodium dodecyl sulfonate (SDS), and cationic surfactant; hexadecyl trimethyl ammonium bromide (CTAB) were employed for the

modification of AC. The findings revealed that the methylene blue adsorption performance by AC modified by anionic surfactant (SLS) was significantly enhanced, whereas the methylene blue adsorption performance by AC modified by cationic surfactant (CTAB) was reduced.

Activated carbon fibers (ACFs) are emerging form of AC variant that have broad application prospects in treatment of dye-bearing wastewater owing to their high specific surface area, abundant pore structure and a variety of active groups [31]. Accordingly, tremendous efforts have been devoted to the preparation of ACFs from different sources especially biomass resources owing to their low cost, low technical barriers and sustainable advantages [141]. Wang et al. [142] developed a facile carbonization strategy to obtain a novel biomass ACF from the seed hair fibers of metaplexis japonica (MACFs). It was observed that MAFCs possess well-developed pore distribution, with a maximum specific surface area and total pore volume of 1882.003 $m^2 g^{-1}$ and 1.613 $cm^3 g^{-1}$, respectively. The prepared MACFs was applied in the removal of MB and achieved a maximum adsorption capacity of 685.25 mg $g^{-1}$.

### 3.1.2. Biochar

Besides AC, biochar, is another carbon variant derived from pyrolysis of biomass and biomass waste in a limited oxygen environment. Biochar often possess similar structural properties as AC and has been investigated for cationic dyes adsorption [143]. Recently, cassava stem biochar (CSB) produced by pyrolyzing CS at 500°C for 2 hours was experimented for MG adsorption. It was found that the optimum pH, contact time and maximum sorption capacity of CSB were 7.4, 10 minutes and 40.5 mg $g^{-1}$, respectively [99]. Kyi et al. [144] explored the adsorption potential of biochar derived from palm kernel shell (BC-PKS) as an adsorbent for the removal of CV from wastewater. They concluded that the increase in solution pH changed the surface charge of BC-PKS, which served favorable adsorption sites for cationic CV dye via electrostatic attraction. In another study, sludge biochar (SB) was prepared through slow pyrolysis at 500°C and utilized for the adsorption of synthetic aqueous MB dye. The removal efficiencies of MB by SB was found to be in the range of 73.0-98.9% at varied experimental conditions [145]. Chahinez et al. [146] developed a biochar (DPP-biochar) material through direct pyrolysis of date palm petioles (DPP) at 700°C and probe its feasibility for the removal of CV dye from aqueous solution. Results demonstrated that the adsorption process was strongly pH dependent and fast equilibrium was attained at approximately 15 min of contact.

In addition to using the native biochar, magnetic biochars (MBC) have attracted much attention recently and exhibited potential for removal of cationic dyes from aqueous medium. Luyen et al. [147] synthesized MBC from rice husk and iron oxide nanoparticles (IONPs), which was employed for removal of CV from aqueous solution. Results indicated the high adsorption capacity of rice husk-based MBC (185.6 mg $g^{-1}$) compared to other MBCs. Ruthiraan et al. [148] demonstrated the adsorption of MB dye from aqueous solution onto nickel oxide attached MBC derived from mangosteen peel. The MBC displays adsorption capacity as high as 22.883 mg $g^{-1}$ for MB. Eltaweil et al. [149] prepared a corn straw-derived biochar supported nZVI magnetic composite and evaluated its efficiency in adsorption of MG dye. It was observed that the maximum adsorption capacity of the composite (515.77 mg $g^{-1}$) was much higher than most of reported biochar adsorbents.

### 3.1.3. Graphene and its Derivatives

Graphene, is a porous carbonaceous material with planar structure and theoretical surface area values up to 2630 $m^2 g^{-1}$. However, despite the large surface area, graphene is a relatively poor adsorbent due to inferior boundary layer needed for diffusion of dyes into pores. To address this limitation, many researchers have focused on the oxidation of graphite to graphene oxide (GO) and reduced graphene oxide (RGO) using Hummers' method or its modification [92]. GO, chemically-functionalized graphene and RGO have attracted the attention of the scientific community and enjoyed more application in adsorption due to their large specific surface area, superior mechanical properties and presence of negatively charged oxygen functionalities which are favorable for adsorption of cationic dyes [150]. Sabna et al. [151] prepared and evaluated GO for the removal of CV and methyl orange (MO) from aqueous solution. It was found that the maximum uptake by GO was higher for CV (207.4 mg $g^{-1}$) than that for MO (37.2 mg $g^{-1}$). Robati et al. [152] studied and elucidated the adsorption of MG onto GO and RGO surfaces. The obtained optimized values of pH and contact time was 3.0 and 100 min, respectively. Siong et al. [153] fabricated RGO at different temperatures via a solvothermal approach. It was demonstrated that the RGO could achieve a higher adsorptive removal (87.39%) of MB dye when 50 mg $L^{-1}$ of dye at pH 11 was used. Elsewhere, Arias et al. [154] prepared RGO by a scalable and eco-friendly method, and its potential for the removal of MB from water systems was explored. The maximum adsorption capacity of RGO for MB was found to be 121.95 mg $g^{-1}$ at pH 9.0.

Although, GO has been applied as adsorbent, the major limitation in exploiting it is the agglomeration of the graphene sheets. The agglomeration

diminishes the accessibility and thus reduces the active binding sites for the contaminants present in wastewater. Moreover, after the adsorption, the separation of GO from aqueous solution is difficult due to its hydrophilic property and nano-sized particle. This limitation has been remedied by decorating the graphene nanosheets with metal and metal oxide nanoparticles to develop high performing graphene-based nanocomposites [155]. Aboelfetoh et al. [156] immobilized silver nanoparticles (AgNPs) onto nanosheets of GO through in situ reduction process using green tea aqueous extract as a reductant and stabilizer. The adsorption behavior of the GO/Ag nanocomposites towards MB dye was investigated under diverse adsorption factors. The results indicate that GO/Ag nanocomposites displayed high MB adsorption potential of up to 633.30 mg g$^{-1}$.

### 3.2. Biomass-based Adsorbents

Biomasses are renewable organic materials, such as plant-based materials, dead microbes, and materials of animal origin. Biomasses can be divided majorly into three classes, namely lignocellulosic, animal, and microbial. The lignocellulosic biomasses are normally plant-based, such as agricultural solid wastes (e.g. bark, leaves, fibers, fruit, peel and seed), but the microbial biomasses could be dead or live microbes [181].

### 3.2.1. Agricultural Solid Wastes

Agricultural wastes are materials that consist of polysaccharides of hemicellulose, cellulose, and lignin within their cellular constituents as main structural components [29]. The structures of these biochemical substances in some parts are associated with a number of functional groups such as hydroxyl and carbonyl groups. When such materials are ready as adsorbents for wastewater treatment, these functional groups become responsible for adsorption of dye molecules. Recently, special attention has been paid for the utilization of the available-in-nature, abundant, and eco-friendly adsorbents to replace the conventional and expensive sorbents based on economic and environmental point of views [96]. The abundance and widespread availability of agricultural wastes/by-products make them good source of raw materials for dye removal. Several agricultural waste products like coconut shell, groundnut shell, orange peel, rice husks, corncob, sugarcane bagasse, nut shells, fruit peels, leaf powders, fruit shells, seed husk, saw dust, etc were tested in their natural form as alternative low cost adsorbent by various researchers in recent past decades [11,182]. Agricultural wastes are better than other adsorbents because they are commonly used without or with a minimum of processing (washing, drying, grinding) and therefore minimize production costs by using a cheap raw material and eliminating energy costs associated with thermal treatment

[183]. Table 5 shows previous studies on the adsorption of cationic dyes using adsorbents based on agricultural solid wastes.

Odoemelam et al. [184] studied the adsorption of MG and MB dyes from aqueous solution by neem leaves using experimental and computational chemistry approaches. Results of their study revealed that the adsorption of the dyes on neem leaves was influenced by concentration, biomass dose, pH and by contact time. Quantum chemical analysis supported better adsorption of MB dye on neem leaves surface than MG dye. Duran et al. [185] utilized almond shell as adsorbent for the removal of MB from water systems. It was observed that equilibrium was rapidly attained after 30 min of contact time with maximum adsorption capacity of 76.34 mg g$^{-1}$. Moussavi and Khosravi [186] evaluated the efficacy of pistachio hull powder as adsorbent for the elimination of MB from contaminated streams. The experimental results indicated that maximum adsorption could be attained at a solution pH of 8.0.

Despite the breakthroughs made in using raw agricultural wastes as adsorbents, low surface area, low adsorption capacity and poor separability from aqueous media, remain the main drawbacks of applying them in their native form. Hence, research on the improvement of adsorption performances through various modifications have been conducted to address these shortcomings. The techniques of modification can be categorized into three broad groups: (i) modification of chemical characteristics; (ii) modification of physical characteristics and; (iii) modification of biological characteristics. Among these three methods, modification with chemical compounds like mineral acids, bases and oxidizing agents have been more frequently employed to enhance and reinforce the functional group potential and, consequently, increase the removal capacity of adsorbents [86]. In addition to the above, cross-linking, grafting, converting to activated carbon or fabricating composite materials are usually the secondary treatments undertaken.

Recently, Ahmed et al. [187] evaluated the adsorption performance of raw orange peel (OP) and magnetically modified orange peel (MOP) as adsorbents for the confiscation of CV from simulated wastewater. It was observed that the equilibrium adsorption capacity was significantly higher with MOP (46.94 mg g$^{-1}$) than with OP (22.73 mg g$^{-1}$). They concluded that magnetization results in improvement of adsorption performance of OP for the removal of CV from aqueous media. In another study, Shoukat et al. [188] prepared mango stone biocomposite (MSBC) and used it for the elimination of CV from wastewater. Results showed that maximum adsorption of 352.79 mg g$^{-1}$ was achieved at pH 8.0, 30 min contact time, 0.05 g adsorbent dose and temperature of 33°C.

Table 4. Reported studies on the use of waste-based ACs for adsorption of cationic dyes

| AC source | Dye | $q_{max}$ (mg g$^{-1}$) | pH | Time (min) | Reference |
|---|---|---|---|---|---|
| Apricot stones material | Malachite green | 88.50 | 10.0 | 40 | [157] |
| Pomegranate peels | Crystal violet | 76.92 | 11.0 | 45 | [158] |
| Acorn shell | Methylene blue | 303.00 | 4.0 | 30 | [159] |
| Kendu fruit peel | Methylene blue | 144.90 | 6.0 | 100 | [160] |
| Rubber seed shell | Crystal violet | NR | 10.0 | 30 | [161] |
| Fir bark | Methylene blue | 330.00 | NR | 40 | [162] |
| Date stones | Methylene blue | 163.67 | 10.0 | NR | [163] |
| Mangosteen peel | Methylene blue | 1193.00 | 9.0 | 600 | [164] |
| Biological sludge | Crystal violet | 640.70 | 8.0 | 120 | [165] |
| Cork waste | Methylene blue | 350.00 | NR | 5 | [166] |
| Tobacco stalks | Methylene blue | 123.45 | 6.5 | 100 | [167] |
| Citrus limetta peel | Methylene blue | 500.00 | 7.0 | 180 | [168] |
| Waste active sludge | Crystal violet | 62.11 | 6.0 | 150 | [169] |
| Oil palm fruit fibre | Malachite green | 356.27 | 6.0 | 45 | [170] |
| Spent tea leaves | Malachite green | 256.4 | 4.0 | 180 | [171] |
| Corncob | Crystal violet | 2.498 | 7.0 | NR | [172] |
| Date palm dead leaflets | Crystal violet | 36.63 | 6.5 | 30 | [173] |
| Corncob | Crystal violet | 320.00 | 5.0 | 240 | [174] |
| Wood apple shell | Crystal violet | 142.85 | 8.0 | 160 | [175] |
| Rattan waste | Methylene blue | 359.00 | 7.0 | 480 | [176] |
| Pea shells | Methylene blue | 246.91 | 11.5 | 180 | [177] |
| Coconut husk | Crystal violet | 315.86 | 9.0 | NR | [178] |
| Eichhornia | Crystal violet | 58.13 | 8.0 | NR | [179] |
| Durian seed | Malachite green | 476.19 | 8.0 | NR | [180] |

NR: Not Reported

Table 5. Previous studies on the removal of cationic dyes using agricultural wastes as adsorbents.

| Adsorbent | Dye | $q_{max}$ (mg g$^{-1}$) | pH | Time (min) | Reference |
|---|---|---|---|---|---|
| Fugas sawdust | Crystal violet | 78.69 | 10.0 | NR | [55] |
| Sawdust | Crystal violet | 131.58 | 7.5 | 180 | [60] |
| Oat hull | Malachite green | 83.00 | 8.0 | 80 | [68] |
| Malt bagasse | Malachite green | 117.65 | 8.0 | 60 | [69] |
| Napier grass stem | Malachite green | 32.27 | NR | 90 | [70] |
| Teak leaf powder | Malachite green | 333.33 | 6.0 | NR | [113] |
| Modified breadnut peel | Malachite green | 227.0 | 3.6 | 60 | [121] |
| Walnut shell | Methylene blue | 51.55 | 6.0 | 120 | [190] |
| Tea waste | Methylene blue | 113.14 | 8.0 | NR | [191] |
| Potato peels | Crystal violet | 555 | 6.0 | 20 | [192] |
| Coffee husks | Crystal violet | NR | 3.0 | 30 | [193] |
| Banana peel | Malachite green | 243.90 | 5.0 | 720 | [194] |
| Ocotea puberula bark powder | Crystal violet | 444.34 | 9.0 | 90 | [195] |
| Fava bean peel | Methylene blue | 140.00 | 5.8 | NR | [196] |
| Terminalia arjuna sawdust | Crystal violet | 45.99 | 7.0 | 120 | [197] |
| Coconut coir dust | Methylene blue | 29.50 | 6.0 | 20 | [198] |
| Punica granatum peel | Methylene blue | NR | 7.0 | 180 | [199] |
| Waste pea shell | Malachite green | 6.20 | 7.0 | 40 | [200] |
| Wood apple shell | Malachite green | 34.56 | 7.0 | 200 | [201] |
| Pineapple peel | Methylene blue | NR | 10.0 | 30 | [202] |
| Pumice stone | Crystal violet | 6.99 | 6.5 | 120 | [203] |
| Rice straw | Crystal violet | 80.91 | 8.0 | NR | [204] |
| Copper pod flowers | Malachite green | NR | 7.0 | 100 | [205] |
| Rice husk | Malachite green | 6.5 | 7.0 | NR | [206] |
| Elaeagnus kernel powder | Malachite green | NR | 9.0 | 90 | [207] |
| Apple peels | Methylene blue | 107.52 | 6.0 | NR | [208] |
| Pineapple leaf | Crystal violet | 158.73 | 8.0 | 120 | [209] |

Elsewhere, Amode et al. [189] demonstrated the effectiveness of alkaline treatment in enhancing the adsorption capacity of sago waste powder on MB. It was observed that the maximum adsorption capacity of alkali-treated sago waste (212.8 mg g$^{-1}$) was more than double that exhibited by the untreated sago waste (83.5 mg g$^{-1}$).

### 3.2.2. Microbial biomasses

Microbial biomasses, dead or living, have been extensively employed for the elimination of cationic dyes from wastewater. Biomasses have high potential as adsorbents due to their low cost, high availability and good texture [31]. Algae, fungi and bacteria are examples of microbial biomasses utilized for dye adsorption. They can adsorb dye molecules or the enzymes secreted by them can degrade chromophores of dye molecules causing their decolorization [181]. Recently, Drumm et al. [210] utilized the wastes from the macro-fungus *Agaricus bisporus* as an eco-friendly and low cost adsorbent for the treatment of colored effluent containing CV dye. It was found that it took 120 min to attain equilibrium and it achieved a maximum adsorption capacity of 228.74 mg g$^{-1}$. Rammel et al. [123] assessed the use of *Chaetophora elegans* alga as adsorbent to remove CV from aqueous solution. It was found that the algal biomass showed quite interesting adsorption capacity (158.70 mg g$^{-1}$) under optimized operating conditions. Morales-Alvarez et al. [211] investigated the partial removal and detoxification of MG and CV from laboratory artificially contaminated water by *Pleurotus ostreatus*. They concluded that an acidic pH favors the adsorption of both dyes. Wu et al. [212] used spent substrate of *Pleurotus eryngii* to adsorb MG, safranine T and MB from aqueous solutions. Results obtained showed that more than 93% of the dyes were removed with 1 g of adsorbent, pH of 6.0 and contact time of 4 h. Yildrim and Acay [213] investigated the adsorption behavior of MB and MG onto *Pleurotus ostreatus, Armillaria tabescens and Morchella conica* mushrooms. The results showed that adsorption on all the mushrooms attained equilibrium within 120 min for both dyes.

However, some shortcomings such as low adsorption capacity and slow binding kinetics have limited the practical application of some microbial biomasses. To improve the adsorption capacity of microbial biomasses, different physical and chemical treatment methods such as surface modification and encapsulation have already been applied. The surface modification approach involved the introduction of active functional groups on the biomass surfaces using acids, bases, etc. Essekri et al. [214] reported the functionalization of brown algae (BA) by citric acid in order to enhance its adsorption capability for CV removal in aqueous solutions. The performance of the novel functionalized brown algae (BA-CA) was 273.85 mg g$^{-1}$, which was about two times higher than that of unmodified BA (146.94 mg g$^{-1}$). They concluded that the carboxyl functions of citric acid on the BA-CA surface played a vital role in CV dye removal. Shao et al. [215] investigated the adsorption of MB on the $H_2SO_4$-modified *Laminaria japonica* alga in which the maximum adsorption capacity was increased by 131.04 mg g$^{-1}$ compared to the native algal biomass. Jerold and Sivasubramanian [216] explored the removal of MG using chemically modified *Sargassum swartzii* algal specie. They reported that the maximum uptake capacity was 111.1 mg g$^{-1}$. In another study, *Cystoseira barbata* was coated with iron oxide ($Fe_3O_4$) to obtain a magnetic biomaterial and used as adsorbent for the removal of MB from aqueous solution [217]. The results showed that the maximum adsorption capacities were achieved in 300 min at pH of 2.0.

### 3.3. Nanomaterial-based Adsorbents

Nanomaterials (NMs) are defined as substances with one or more external dimension in the range of 1 to 100 nm possessing unique optical, mechanical, and magnetic properties [23,92]. The extremely large surface area-to-volume ratio, high porosity, modified surface properties, strong binding affinity, unique electron conduction properties, lighter density etc. offer NMs with excellent performances in removal of various kind of pollutants from the water environment [130,218-219]. Thus, NMs bring new opportunities for the revolution of the dye-contaminated wastewater treatment technologies. Various types of NMs such as carbon nanotubes [120], magnetic nanoparticles [220], $TiO_2$ nanoparticles [221], mesoporous silica nanoparticles [17] and $MoS_2$ nanosheets [222] have been developed for the adsorptive removal of dyes from aqueous medium. Table 6 presented the compilation of various nanomaterials used in the removal of cationic dyes together with some experimental parameters and maximum removal capacity.

Gopalakrishnan et al [223] successfully synthesized few-layered $MoS_2$ nanosheets grown into graphene dipped cellulose filter paper (M-G-CFP) by a simple hydrothermal method towards adsorption of MB dye. The hybrid M-G-CFP exhibits ultrafast removal of MB within 2 min with maximum adsorption capacity of 485.4 mg g$^{-1}$. Luo et al. [224] demonstrated the high performance of the amorphous $WO_3$ nanosheets in the removal of MB from water. Results indicate that the amount of MB adsorbed using $WO_3$ nanosheets can reach up to 600 mg g$^{-1}$. Rajabi et al. [225] successfully utilized multi-walled carbon nanotubes functionalized with the carboxylate group as an efficient adsorbent for the fast adsorption of MG from aqueous solution. The optimized pH, contact time and Langmuir adsorption capacity were found to be 10 min, 9.0,

and 11.73 mg g$^{-1}$, respectively. Ali et al. [226] used magnetic nanoparticles biosynthesized from leaf extract of Fraxinus chinesis Roxb for the removal of CV and eriochrome black T dyes from aqueous solution. The results showed that the nanoparticles exhibit >95% removal of both dyes with 10 minutes of contact time over a wide range of concentration, 10-300 mg L$^{-1}$. In another study, Jain et al. [227] experimented the removal of MG from aqueous solution by Zeolite-Iron Oxide Magnetic Nanocomposite. The nanocomposite exhibited maximum dye removal at an optimum pH of 8.0, contact time of 60 min and adsorbent dose of 5 g L$^{-1}$. The maximum Langmuir adsorption capacity was found to be 21.05 mg g$^{-1}$.

### 3.4. Polymer-based Adsorbents

Polymeric adsorbents have emerged as promising alternatives to traditional adsorbents because of their vast surface area, wide range of pore structure, high adsorption capacity, strong binding affinities, adjustable surface chemistry, remarkable flexibility, feasible regeneration under mild conditions, and good mechanical, chemical, and thermal stability [20,254]. Generally, polymer-based adsorbents can effectively adsorb many ubiquitous organic and inorganic pollutants [255]. For instance, Omer et al. [256] fabricated novel anionic adsorbent microbeads based on surface functionalization of p-benzoquinone-carboxymethyl cellulose. The fabricated adsorbent was tested for the removal of CV from aqueous solution under various experimental conditions. The results clarified that the functionalized adsorbent microbeads exhibited much higher adsorption capability (107.52 mg g$^{-1}$) compared with the native carboxymethyl cellulose microbeads. In another study, Sharma and Kumar [257] synthesized biodegradable functionalized cellulose based graft copolymers (Cell-g-HEMA and Cell-g-HEMA-co-GMA) and utilized them as sorbents for the removal of cationic dyes (MG and CV) from aqueous solutions. The percentage dye uptake are 49.42%, 67.74% for MG and CV, respectively by Cell-g-HEMA-co-GMA. Yadav et al. [258] utilized novel magnetic/activated charcoal/β-cyclodextrin/alginate polymer composite as adsorbent for the elimination of MB from aqueous solution. The obtained results indicated that the removal rate of MB was 99.53% in 90 min. Benhalima and Ferfere-Harrar [259] evaluated the use of hydrogel composite beads based on crosslinked-carboxymethyl cellulose (CMC) and dextran sulfate (DS) as adsorbent for the removal of MB from aqueous solution. The maximum adsorption capacity of CMC/DS composite beads was found to be 526 mg g$^{-1}$. Chen et al. [260] fabricated Poly(cyclotriphosphazene-co-4,4$^{'}$-sulfonyldiphenol) (PZS) nanosheets by simple precipitation polymerization method and used as an efficient adsorbent for the removal of MB from

aqueous solution. Results indicated that the neutral and basic solutions enhanced MB adsorption.

Elsewhere, poly glycidyl methacrylate/divinyl benzene was synthesized by thermal polymerization and employed as an adsorbent for the removal of MG dye from aqueous medium [261]. The maximum adsorption capacity was found to be 13.6 mg g$^{-1}$ at 310 K. Nasiri and Alizadeh [262] synthesized hydroxypropyl-β-cyclodextrin (HPβCD), HPβCD-conjugated magnetic nanoparticles (HPMN) and HPβCD-conjugated magnetic nanoparticles with polyurethane networks (HPMNPU) and used as adsorbents for the removal of CV and methyl violet dyes from aqueous solutions. The maximum adsorption capacity value of HPMNPU was approximately 1269 mg g$^{-1}$ for CV. Nakhjiri et al. [263] demonstrated the ability of Poly(AA-co-VPA) hydrogel cross-linked with N-maleyl chitosan to effectively remove CV and MB from aqueous medium. It was found that the maximum adsorption capacity of the hydrogel polymer for removal of CV and MB in 50 mg L$^{-1}$ of dye solutions was 64.56 mg g$^{-1}$ and 66.89 mg g$^{-1}$, respectively. A summary of reported works using various polymer-based adsorbents to remove cationic dyes is displayed in Table 7.

### 3.5. Emerging Adsorbents

From the recent decades, some emerging and promising adsorbents such as Schiff bases, Metal organic frameworks (MOFs), aerogels and layered double hydroxides are widely tried for cationic dyes adsorption due to their porous structure, large specific surface area and adjustable surface chemistry. In this segment, we mainly introduce briefly the recent studies on the use of the aforementioned adsorbents.

### 3.5.1. Schiff Bases

Schiff bases derived from aldehydes and primary amines have attracted attention as adsorbents for the removal of toxic dyes from the aquatic environment [286]. Recently, Amylopectin-primary amine-Schiff bases were synthesized successfully by first selective oxidation of amylopectin using sodium metaperiodate in aqueous medium followed by Schiff base formation with ethanol amine, hydrazine and semicarbazide [287]. The well characterized Schiff bases were employed as adsorbents for the removal of three cationic dyes namely MG, MB and methyl violet from their aqueous solutions. It was found that among the three Schiff bases, amylopectin dialdehyde-ethanolamine (APDA-EA) showed stronger and selective adsorption affinity towards MG achieving a maximum adsorption capacity of 89.84 mg g$^{-1}$. In another study, a novel Schiff base/SBA-15 nanoadsorbent was successfully synthesized by the reaction of salicyldehyde and (3-aminopropyl) trimethoxysilane in methanol and

Table 6. Studies on the use of nanomaterials for removal of cationic dyes from aqueous medium

| Material | Dye | $q_{max}$ (mg g$^{-1}$) | pH | Time (min) | Reference |
|---|---|---|---|---|---|
| Magnesium oxide nanoparticles | Methylene blue | 163.87 | 7.3 | 70 | [228] |
| Agar based bimetallic nanoparticles | Methylene blue | 875.00 | 8.0 | 40 | [229] |
| Carbon nanotubes | Crystal violet | 90.52 | 8.0 | 45 | [230] |
| MOS$_2$ nanosheets | Malachite green | 204.00 | NR | 300 | [231] |
| Iron nanoparticles | Methylene blue | 208.33 | 9.5 | 1 | [232] |
| Silica nanoparticles | Crystal violet | 34.50 | 12.0 | 50 | [233] |
| Mesoporous silica nanoparticles | Methylene blue | 347.20 | 10.0 | 30 | [234] |
| Mesoporous silica nanoparticles | Methylene blue | 34.23 | 7.0 | 6 | [235] |
| Silver nanoparticles | Malachite green | NR | 5.0 | 240 | [236] |
| Iron nanoparticles | Methylene blue | 230.80 | NR | 240 | [237] |
| Fe$_2$O$_3$ nanoparticles | Crystal violet | NR | 7.0 | 12h | [238] |
| Iron oxide nanoparticles | Methylene blue | 91.00 | 11.0 | 60 | [239] |
| TiO$_2$ nanoparticles | Methylene blue | 9.8 | 6.0 | NR | [240] |
| Multi-walled carbon nanotubes | Malachite green | 112.36 | 7.0 | 17 | [241] |
| Nanocomposite | Crystal violet | 500 | 12.0 | 120 | [242] |
| Carbon nanoparticles | Methylene blue | 167.00 | 12.0 | NR | [243] |
| Nanofibrous composite mat | Methylene blue | NR | 10.0 | 300 | [244] |
| Graphene nanoplatelets | Methylene blue | 225.00 | 4.0 | 45 | [245] |
| Multi-walled carbon nanotubes | Crystal violet | 57.80 | NR | 50 | [246] |
| TiO$_2$ nanoparticles | Malachite green | 6.30 | NR | 30 | [247] |
| Carbon nanotubes | Methylene blue | 132.60 | NR | 120 | [248] |
| Multi-walled carbon nanotubes | Malachite green | 142.85 | 11.0 | 80 | [249] |
| LaMnO$_3$ nanorods | Malachite green | 27.03 | NR | NR | [250] |
| Pr$_2$CuO$_4$ nanosheets | Malachite green | 3520 | NR | NR | [251] |
| Manganese oxide nanosheets | Methylene blue | 4300 | 11.0 | NR | [252] |
| Multi-walled carbon nanotubes | Crystal violet | NR | 11.0 | 150 | [253] |

Table 7. Reported studies on the removal of cationic dyes by polymer-based adsorbents.

| Adsorbent | Dye | $q_{max}$(mg g$^{-1}$) | pH | Time (min) | Reference |
|---|---|---|---|---|---|
| Cellulose nanocrystals | Methylene blue | 118.00 | 9.0 | NR | [264] |
| Chitosan composite | Crystal violet | NR | 9.0 | 40 | [265] |
| Cellulose powder | Malachite green | 2.42 | 7.2 | 30 | [266] |
| Magnetic chitosan nanocomposites | Crystal violet | 333.33 | NR | 140 | [267] |
| Alginate | Methylene blue | 572.00 | 7.0 | NR | [268] |
| Chitosan-graphene oxide | Crystal violet | 64.94 | 8.0 | 180 | [269] |
| Modified polyurethane poly(styrene-alt-maleic anhydride) | Methylene blue | 344.83 | 10.0 | NR | [270] |
| Alginate/pectin nanocomposite | Crystal violet | 619.22 | 6.1 | 240 | [271] |
| Modified chitosan composite | Malachite green | NR | 4.0 | 60 | [272] |
| Dicarboxymethyl cellulose | Methylene blue | 887.60 | 3.0 | 60 | [273] |
| EDTA-modified magnetic chitosan | Crystal violet | 227.30 | 7.0 | 60 | [274] |

| Polyaniline/TiO₂ hydrate | Methylene blue | 458.10 | NR | 120 | [275] |
|---|---|---|---|---|---|
| Poly methyl methacrylate | Methylene blue | 97.08 | 8.0 | 45 | [276] |
| Polyaniline | Methylene blue | 19.20 | NR | 90 | [277] |
| Polypyrrole | Methylene blue | 19.31 | NR | 120 | [277] |
| Hypercrosslinked polyaniline | Crystal violet | 245 | 11.0 | 60 | [278] |
| Amylopectin/poly (acrylic acid) | Malachite green | 352.11 | NR | 30 | [279] |
| Alginate-based beads | Methylene blue | 400.00 | 7.0 | NR | [280] |
| Alginate sponge | Methylene blue | 1279.00 | NR | 50 | [281] |
| Fly ash-based geopolymer | Methylene blue | 37.04 | 11.2 | 120 | [282] |
| Maleic anhydride modified cellulose | Malachite green | 370.00 | 6.0 | 30 | [283] |
| Phthalic anhydride modified cellulose | Malachite green | 111.00 | 6.0 | 60 | [283] |
| O-carboxymethyl chitosan | Crystal violet | 239.54 | 8.0 | 240 | [284] |
| Magnetic nanocomposite hydrogel | Crystal violet | 72.40 | 8.0 | 100 | [285] |

NR: Not Reported

subsequent grafting with SBA-15 [288]. The well characterized Schiff base/SBA-15 nanoadsorbent was utilized for the removal of MB. The obtained results showed that the novel adsorbent is efficient in removing MB from aqueous systems.

### 3.5. Emerging Adsorbents

From the recent decades, some emerging and promising adsorbents such as Schiff bases, Metal organic frameworks (MOFs), aerogels and layered double hydroxides are widely tried for cationic dyes adsorption due to their porous structure, large specific surface area and adjustable surface chemistry. In this segment, we mainly introduce briefly the recent studies on the use of the aforementioned adsorbents.

### 3.5.1. Schiff Bases

Schiff bases derived from aldehydes and primary amines have attracted attention as adsorbents for the removal of toxic dyes from the aquatic environment [286]. Recently, Amylopectin-primary amine-Schiff bases were synthesized successfully by first selective oxidation of amylopectin using sodium metaperiodate in aqueous medium followed by Schiff base formation with ethanol amine, hydrazine and semicarbazide [287]. The well characterized Schiff bases were employed as adsorbents for the removal of three cationic dyes namely MG, MB and methyl violet from their aqueous solutions. It was found that among the three Schiff bases, amylopectin dialdehyde-ethanolamine (APDA-EA) showed stronger and selective adsorption affinity towards MG achieving a maximum adsorption capacity of 89.84 mg g⁻¹. In another study, a novel Schiff base/SBA-15 nanoadsorbent was successfully synthesized by the reaction of salicyldehyde and (3-aminopropyl) trimethoxysilane in methanol and subsequent grafting with SBA-15 [288]. The well characterized Schiff base/SBA-15 nanoadsorbent

was utilized for the removal of MB. The obtained results showed that the novel adsorbent is efficient in removing MB from aqueous systems.

### 3.5.2. Layered Double Hydroxides

Layered double hydroxides (LDHs) are commonly known as hydrotalcite-like minerals or synthetic anionic clays. Similarly, LDHs with their average particle diameters (73–367 nm) are classified as colloidal nanoparticles [289]. LDHs compounds are considered as typically ionic layered materials because their structural characterization comprises positively charged brucite-like layers and non-framework charge compensating anions in their galleries [130]. In essence, such materials can serve as typical dual-electronic adsorbents for adsorbing cationic and anionic dyes [290]. Therefore, LDHs-based materials have been examined in recent times as potential and promising adsorbents for removing cationic dyes from the water environment. Recently, Mg/Fe and Ni/Fe LDHs with molar ratio ($M^{2+}/Fe^{3+}$) of 3 and intercalated with carbonate ions were prepared by co-precipitation method [291]. The as-prepared materials and their calcined products (CLDHs) were employed as adsorbents for the removal of MB and MG as representative of cationic dyes. The experimental results revealed the CLDHs had much higher adsorption capacities compared to LDHs. In another study, carbon-coated Zn/Al LDHs (C-Zn-Al LDH) was used in the adsorption of CV and MG dyes [292]. The adsorption capacity was maximally obtained as 129.87 and 126.58 mg g⁻¹ for CV and MG, respectively.

### 3.5.3. Covalent Organic Frameworks

Covalent organic frameworks (COFs) are a series of porous crystalline materials with powerful covalent bonds between orderly organic construction blocks, and these frames have a large surface area [293]. Due to their constructive order, high surface area, permanent porosities and high stabilities,

synthetic COFs have demonstrated excellent potential for many applications in diversified fields such as catalysis, extraction and adsorption [294]. Recently, a highly thermally stable benzimidazole based covalent organic framework (bCOF) was prepared by the reaction of perlin-tetracarboxylic anhydride, 5-aminoisophthalic acid and diaminobenzidine in polyphosphoric acid medium [295]. The synthesized porous bCOF was utilized for the removal of MB from aqueous solution, and the operating parameters such as pH of the solution, contact time and initial concentration were examined. The results demonstrated that the new bCOF exhibited maximum adsorption capacity of 63.29 mg g$^{-1}$ after only 40 min contact time at a pH of 6.0. In another study, Huo et al. [296] fabricated COF from O-linked flexible 2,4,6-tris(p-formylphenoxy)-1,3,5-triazine and utilized it as adsorbent for removing MB from water. The results demonstrate that the COF is highly stable in water and achieved a maximum of 12.31 mg g$^{-1}$ at pH of 10.0.

### 3.5.4. Aerogels

Aerogels are nanoporous materials formed by the slow replacement of liquid phase in a gel with gas through a supercritical drying or freeze-drying process [297]. Recently, aerogels have attracted empirical attention due to their exceptional properties such as highly porous three-dimensional (3D) network, large specific surface area, ultralight density, and the low mean free path of diffusion [298]. Among various aerogels, polymer aerogels, derived from biomass materials, have mostly been the focus of investigations [299]. For example, a novel three-dimensional (3D) magnetic bacterial cellulose nanofiber/graphene oxide polymer aerogel (MBCNF/GOPA) composed of bacterial cellulose nanofibers (BCNFs), Fe$_3$O$_4$ nanoparticles, graphene oxide (GO) nano-sheets, and polyvinyl alcohol (PVA) was developed by combining facile filler-loaded networks method with the vacuum freeze-drying process for the effective removal of MG from aqueous solution [300]. The results showed that the prepared MBCNF/GOPA displayed excellent MG removal efficiency (93%) with a maximum adsorption capacity of 270.27 mg g$^{-1}$. In another study inspired by the high adsorption efficiency of aerogels, a thiourea-modified nanocomposite aerogel consisting of nanofibrillated cellulose and chitosan was synthesized via facile method [301]. The novel as-synthesized aerogel was employed for the removal of CV and acid orange from aqueous solutions. The results indicated that solution pH had substantial effect on the process and maximum adsorption was achieved at pH of 6.0 for CV and 3.0 for acid orange.

### 3.5.5. Metal Organic Frameworks

Metal organic frameworks (MOFs) are a new class of crystalline organic-inorganic hybrid solids which are made by linking metal clusters or ions and organic linkers through covalent bond. Owing to their highly ordered structures, high porosity, large surface areas and modifiable surfaces, MOFs have attracted intensive attention in adsorption, catalysis, extraction, gas storage and molecular sensing [302,303]. Recently, many types of MOF-based materials have been successfully prepared and are recognized as promising adsorbents for removal of dyes from wastewater [14]. For instance, Xie et al. [304] prepared a cationic MOF (ZJU-48) from zinc, adenine and carboxyl ligands and employed it for adsorption of cationic (MB) and anionic dyes. The results show that cationic dyes are adsorbed better and the maximum loaded amount of ZJU-48 toward MB reaches 582.44 mg g$^{-1}$. Elsewhere, Shi et al. [305] synthesized a magnetic MOFs composite (Cu-MOFs/Fe$_3$O$_4$) as the adsorbent for removal of MG and lead in wastewater. It was found that the prepared Cu-MOFs/Fe$_3$O$_4$ achieved adsorption capacities of 113.67 mg g$^{-1}$ for MG, which is higher than some reported materials.

## 4. Modeling of Adsorption Process

### 4.1. Adsorption Isotherm Models

Adsorption isotherms models are important tools for the design and scale-up of an adsorption system as they describe the interaction between the dye molecules and active sites of the adsorbent surface [14]. 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. Based on the considered hypothesis concerning each model, the parameters associated with each isotherm model might offer insight regarding the adsorbent surface properties, degree of affinity of the adsorbate towards the adsorbents, distribution of exchangeable sites on the adsorbent surface and the mechanism of adsorption [9]. The removal of cationic dyes by different adsorbents can be described by various adsorption isotherm models including Freundlich, Langmuir, Temkin, Dubinin-Radushkevich (D-R) and Redlich-Peterson (R-P) models [306]. In comparison with other isotherm models, Langmuir and Freundlich models have been the most extensively explored for the simulation of cationic dyes adsorption. Readers interested in the equations together with the basic assumptions of these isotherm models can refer to a detailed review by Foo and Hameed [307].

Dandil et al. [53] applied the Langmuir and Freundlich isotherm models to study the mechanisms of adsorption of CV onto calcinated waste mussel shell. The isotherms factors were evaluated at pH 6, contact time 220 min and temperature 25°C. By way of comparing correlation

coefficients values ($R^2$) for the two isotherms, the Freundlich isotherm model showed the best fit of the isotherm data. This indicate the heterogeneous multilayer coverage of the dye on the surface of the adsorbent. These results are also similar to those reported by Sriram et al. [308]. They also applied the isotherm models to their data on MG adsorption from aqueous solution by porous silica diatomaceous earth (DE) microparticles. Their findings showed that the Freundlich model gave the best fit having an $R^2$ value of 0.972. It is interesting to note that some data will however conform to the Langmuir isotherms more than they will conform to the Freundlich, indicating adsorption on homogenous surfaces. Gurkan and Coruh [309] investigated the adsorption performance of MG by waste foundry sand using Langmuir, Freundlich and Temkin isotherms models. According to the $R^2$ value, the Langmuir model is more suitable than the other models which suggests that the adsorption is a monolayer process. Yakout et al. [310] applied Langmuir and Freundlich models to study the adsorption performance of activated charcoal to CV. It was found that the Langmuir model is more suitable than the Freundlich. The summary of modeling studies of adsorption systems that involve the aqueous-phase removal of cationic dyes by various adsorbents is diaplayed in Table 8.

*4.2. Kinetic Modeling*

Adsorption kinetics is one of the main parameters to consider while designing the adsorption system. In the past decades, many kinetic models of varied complexity have been used to determine the mechanism of adsorption process including the uptake rate, diffusion control, and mass transfer [318]. The kinetic models can generally be categorized as adsorption diffusion models and adsorption reaction models. The diffusion models are normally formulated on the basis of three consecutive steps: (i) diffusion of solute across the liquid film surrounding the adsorbent material, i.e film diffusion; (ii) diffusion of solute in the pores of the adsorbent, which is so-called intra-particle diffusion; and (iii) adsorption of adsorbate on the active site of the adsorbent. However, reaction models originating from chemical reaction kinetics are based on the whole process of adsorption without considering the mechanistic steps [319]. At present, the pseudo-first-order (PFO) and pseudo-second-order (PSO) models are the most popular empirical models employed in liquid adsorption studies. However, in most cases, PSO is superior in terms of fit to the PFO model by a least-square discrimination procedure; its qe is often much closer to the experimental value compared with that given by the PFO model. Readers interested in the derivation, features and modified version (if any) of these kinetic models can consult a comprehensive

review by Tan and Hameed [92].

Yunusa et al. [134] fitted the experimental data to pseudo-first-order and pseudo second- order models in order to investigate the mechanism of adsorption of CV and MG by using desert date seed shell as adsorbent. It was observed that the correlation coefficient of the pseudo second-order model was relatively greater than that of the pseudo-first-order model, implying that the adsorption process can be described more appropriately by the pseudo-second-order model. Based on the assumption of the pseudo-second-order model, they concluded that the adsorption of the cationic dyes onto desert date seed shell was a chemically-based sorption process. In another study, Adeyi et al. [126] applied four kinetic models i.e pseudo-first-order, pseudo-second-order, Elovich, and intraparticle diffusion (IPD) models to evaluate the adsorption mechanism of MG by thiourea-modified poly(acrylonitrile-*co*-acrylic acid) (TU-poly(AN-*co*-AA)). Results revealed that the experimental data was consistent with pseudo- second-order model. The summary of kinetic investigations of adsorption systems that involve the removal of cationic dyes from water had been displayed in Table 9. It can be observed from Table 9 that PFO-compliant kinetics is difficult to come by as the model has been shown to be valid only at the initial stage of adsorption, whereas PSO exhibits good fit over the entire time course.

*4.3. Mechanisms of Cationic Dyes Removal*

Due to the intricacy of the adsorbents utilized in cationic dyes removal, many mechanisms for their abstraction in wastewater have been proposed. Instrumental techniques such as Fourier transform infrared (FTIR) spectroscopy, scanning electron microscopy (SEM), X-ray photoelectron spectroscopy (XPS) and X-ray diffraction (XRD) have been employed by various researchers to explore the probable removal mechanism of cationic dyes by different adsorbents. In general, the adsorption of cationic dyes on diversified adsorbents is achieved by the involvement of electrostatic or ionic interactions, π-π interactions and hydrogen bonding. Besides, it has been reported that the adsorption of cationic species onto carbon surfaces is majorly due to –OH and –COO⁻ functional groups [324]. For example, recently, Yunusa et al. [134] employed FTIR spectroscopy and point of zero charge ($pH_{zpc}$) analyses to elucidate the mechanism of adsorption of CV and MG onto activated carbon (NAC) surface. They concluded that the adsorption of the dyes onto the adsorbent occurred due to strong electrostatic interaction between negatively charged groups on the carbon surface (-OH and –COO⁻) and the positively charged centers of crystal violet

Table 8. Isotherm studies of cationic dyes adsorption by various adsorbents

| Adsorbent | Dye | Best fit model | Discrimination | Reference |
|---|---|---|---|---|
| Activated carbon | Crystal violet | Langmuir | $R^2$, SSE, RMSE | [52] |
| Sawdust | Crystal violet | Langmuir | $R^2$ | [60] |
| Tea dust | Crystal violet | Langmuir | $R^2$, $\chi^2$ | [63] |
| Oat hull | Malachite green | Freundlich | $R^2$, SSE | [68] |
| Napier grass stem | Malachite green | Langmuir | $R^2$ | [70] |
| Modified clay | Crystal violet | D-R | $R^2$, $\%\Delta q_e$ | [107] |
| Teak leaf litter | Malachite green | D-R | $R^2$ | [113] |
| Activated carbon | Methylene blue | Langmuir | $R^2$ | [140] |
| Biochar | Crystal violet | R-P | $R^2$, $\chi^2$, BIC | [146] |
| Graphene oxide | Crystal violet | Freundlich | $R^2$ | [151] |
| Activated carbon | Malachite green | Langmuir | $R^2$, SSE, RMSE | [158] |
| Neem leaves | Methylene blue | Temkin | $R^2$ | [184] |
| Potato peels | Crystal violet | Langmuir | $R^2$ | [192] |
| Banana peel | Malachite green | Freundlich | $R^2$ | [194] |
| Pumice stone | Crystal violet | Langmuir | $R^2$ | [203] |
| Nanoparticles | Malachite green | Langmuir | $R^2$ | [206] |
| $MOS_2$ nanosheets | Methylene blue | Freundlich | $R^2$ | [222] |
| Alginate sponge | Methylene blue | Langmuir | $R^2$ | [261] |
| Nanocellulose | Crystal violet | Langmuir | $R^2$ | [311] |
| Biochar | Methylene blue | Langmuir | $R^2$ | [312] |
| Activated carbon | Methylene blue | Langmuir | $R^2$, RMSE | [313] |
| Pristine lignin | Malachite green | Langmuir | $R^2$ | [314] |
| *Pleurotus ostreatus* | Methylene blue | Freundlich | $R^2$ | [315] |
| *Morchella conica* | Malachite green | Freundlich | $R^2$ | [315] |
| *Morchella conica* | Methylene blue | Freundlich | $R^2$ | [315] |
| Activated carbon | Methylene blue | R-P | $R^2$, $\%\Delta q_e$ | [316] |
| Activated carbon | Crystal violet | R-P | $R^2$, $\chi^2$, SSE | [317] |

RMSE: root mean square error; SSE: sum of error squares; $\%\Delta q_e$: normalized standard deviation; $\chi^2$: chi-square test; BIC: Bayesian information criterion

Table 9. Summary of best fit kinetic models for cationic adsorption onto different adsorbents

| Adsorbent | Dye | Best fit model | Discrimination | Reference |
|---|---|---|---|---|
| Sawdust | Crystal violet | PSO | $R^2$ | [60] |
| Oat hull | Malachite green | PSO | $R^2$, SSE | [68] |
| Malt bagasse | Malachite green | PSO | $R^2$ | [69] |
| Modified clay | Crystal violet | PSO | $R^2$, $\%\Delta q_e$ | [107] |
| Teak leaf litter | Malachite green | PSO | $R^2$ | [113] |
| Biochar | Crystal violet | PSO | $R^2$ | [144] |
| Graphene oxide | Crystal violet | PSO | $R^2$ | [151] |
| Activated carbon | Crystal violet | PSO | $R^2$ | [169] |
| Neem leaves | Methylene blue | PSO | $R^2$ | [184] |
| Rice husks | Malachite green | PSO | $R^2$ | [206] |
| *Pleurotus ostreatus* | Methylene blue | PFO | $R^2$ | [213] |
| *Armillaria tabescens* | Methylene blue | PFO | $R^2$ | [213] |
| *Morchella conica* | Malachite green | PFO | $R^2$ | [213] |
| Magnetic nanoparticles | Crystal violet | PFO | $R^2$ | [226] |
| Foundry sand | Malachite green | PSO | $R^2$ | [309] |
| Activated carbon | Crystal violet | PSO | $R^2$, $\chi^2$, SSE | [317] |
| Water hyacinth | Crystal violet | PSO | $R^2$ | [320] |
| *Pleurotus eryngii* | Methylene blue | PSO | $R^2$ | [321] |
| *Pleurotus eryngii* | Malachite green | PSO | $R^2$ | [321] |
| Magnetic nanoparticles | Malachite green | PSO | $R^2$ | [322] |
| Activated carbon | Crystal violet | PSO | $R^2$, $\chi^2$, ARE(%) | [323] |

SSE: sum of error squares; $\%\Delta q_e$: normalized standard deviation; $\chi^2$: chi-square test; ARE(%): Average relative error.

($CV^+Cl^-$) and malachite green ($MG^+Cl^-$) molecules. They mechanistically postulated the process using Eqs 1 and 2:

Elsewhere, Emrooz et al. [325] employed instrumental techniques to investigate the adsorption mechanism of MB on a *Azolla*-derived micro- and mesoporous carbon surfaces. They observed shifts in characteristic peaks of some functional groups (in FTIR analysis) and crystal plains (in XRD and Raman analyses) which depicts strong electrostatic interaction or chemical bonding between some groups of adsorbed dye and some functional groups of the adsorbent. In another study, Ali et al. [322] employed FTIR, XRD and XPS analysis to investigate the probable removal mechanism of MG by phytogenic magnetic nanoparticles. They

observed that in the FTIR spectrum after MG adsorption, the stretching vibrations of –OH and –SH functional groups were altered and reduced, signifying the adsorption of cationic dye molecules with the negatively charged functionalities via electrostatic interactions. Moreover, the XPS spectra of the adsorbent after the adsorption of dye molecules indicated that the peak for –OH functional group was reduced after the interaction with dye molecules. They concluded that this illustrate the contribution of –OH functional group in the removal of cationic dye via ion-exchange or chemisorption. In another study, Kumar and Ahmad [326] reported a three-stage mechanism for the adsorption of CV by treated ginger waste as shown in Fig. 6.
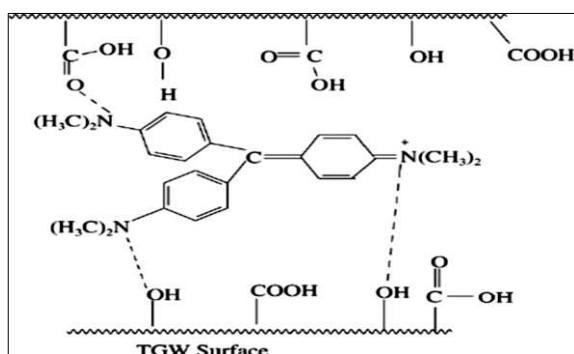
$$NAC–OH \longrightarrow NAC–O^- + CV^+Cl^- \longrightarrow NAC–O^{....}CV \tag{1}$$

$$NAC–OH \longrightarrow NAC–O^- + MG^+Cl^- \longrightarrow NAC–O^{....}MG \tag{2}$$



Fig. 6. Probable mechanism for the adsorption of CV onto treated ginger waste [326]

## 5. Concluding notes

Based on the extensive literature reviewed, the following conclusions can be drawn:

- Various adsorbents, including carbon-based materials, biomaterials, nanomaterials, polymer-based materials, layered-double hydroxides, aerogel, Schiff bases and metal-organic frameworks have been successfully applied in cationic dyes removal from aquatic environment.
- The most important parameter that influence cationic dyes adsorption is the solution pH, where a high pH value is required for maximum dye uptake.
- The Langmuir and Freundlich isotherm models are extensively used to evaluate the adsorption capacity of cationic dyes adsorbents.

- The kinetic data of adsorption of cationic dyes by different adsorbents usually follows the pseudo-second-order model.
- The major drawback of the already published adsorption studies is that their use is still at lab-scale mostly without pilot studies or commercialization. Also, the majority of the studies focused on batch mode experiments using mono-pollutant simulated solutions with only few studies using real wastewater. Furthermore, limited attempts for detailed economic analysis are available.
- Ultimately, in order cover these existing research gaps, a further work in the following direction is necessary: (i) more research on cationic dye adsorption from multi-component effluents in order to better understand the adsorption behavior of various adsorbents in complex matrices; (ii) the feasibility of recovery and recycling of the spent adsorbents should be carefully investigated; (iii) dynamic adsorption studies should be conducted for design perfection purposes and real application; (iv) study of the adsorption behavior of cationic dyes from quantum-chemical perspective for precise mechanistic insights; (v) Future researchers should investigate combined adsorbents instead of stand-alone adsorbents (v) critical economic analysis of adsorption processes in order to scale-up operation (from lab to industry).

## Conflict of Interest

The authors declare that they have no conflict of interest

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