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REVIEW ARTICLE

Utilization of tea wastes for the removal of toxic dyes from polluted water—a review

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Abstract
The presence of dyes in the aqueous system is a worldwide concern. Among the different available water treatment methods, adsorption one has attracted substantial consideration due to its unmatched advantages like selectivity, low cost, applicability verities of contaminants, high efficiency, ease and simplicity of operation, reusability of the adsorbents, etc. The utilization of these advantages depends on the appropriate choice of the adsorbent. The surplus availability and simple preparation might be the primary requirements of a promising adsorbent. In this context, tea waste materials pose themselves as potential candidates for their employment as adsorbents. In this review article, the use of unmodified raw tea waste materials as adsorbents for the remediation of water and wastewater containing dyes as pollutants has been thoroughly discussed. The review includes the characterization of tea waste-based adsorbents and their utilization for dye removal. The isotherm, kinetics, and thermodynamics accompanying the process of dye removal have also been discussed.

Keywords Tea waste · Dye pollution · Adsorbents · Wastewater treatment · Dye confiscation

1 Introduction

Water, a most valuable asset for human endurance, is confronting extraordinary worries. Water contamination has become one of the most genuine worldwide issues. The freshwater is a fundamental necessity for humans and wildlife. Nonetheless, this circumstance is still spreading; a lot of wastewater is created and discharged to the natural freshwater bodies. Among various kinds of wastewaters, dye-contaminated wastewater merits critical consideration. During the last few decades, with the consistent advancement of the printing and coloring industrialization process, many dying chemicals are discharged into the environment, particularly water systems. Synthetic organic dyes are chemicals that are widely used in different fields to color everyday usable items like textiles, leather, toys, paper, rubber, printing inks, food products, building, transport vehicles, etc. While coloring a product, a considerable volume of water is consumed, and ultimately, freshwater is converted to dye-contaminated water. For example, about 200 L of water is consumed to color 1 kg fabrics, and thereby over 1.5 million liters of water per day are spent for an average-sized mill [1]. The presence of color contaminations in the aqueous system carries an enormous hazard to humans and other living organisms. For example, the color can have an unfriendly effect on both amphibian life forms and individuals because colors can decrease sunlight transmission. Additionally, the dye-laden water also contains toxic materials. Some dye molecules are mutagenic and cancer-causing. They cause disease of the kidney, liver, conceptive framework, brain, and central nervous system [1–3]. As dyes' consumption can neither be stopped nor declined, the treatment of dye-contaminated water has become a challenging and emergent task.

The polluted wastewater containing contaminants like dyes, heavy metals, pesticides, antibiotics, etc. has been treated by employing various developed techniques. Some notable ones are advanced oxidation process [4–8], ion exchange [9–11], electrocoagulation [12–14], electrodialysis [15–17], electroflotation [18–20], ozonation [21–24], photochemical oxidation [25], coagulation/flocculation [26, 27], nanofiltration/ultrafiltration [28–33], reverse osmosis [34–37], MnO_2 oxidation [38–40], biological method (aerobic/anaerobic) [22, 41–43], chemical precipitation [44–47],

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etc. Each of these methods might be advantageous for one aspect, while it may be unfavorable on other aspects. However, most of these techniques are conventional and not universally applicable due to some limits in terms of cost, feasibility, efficiency, operational difficulty, and/or other uncontrolled factors. Among the available methods, adsorption is usually favored due to its effectiveness, simplicity, sustainability, insensitivity towards toxic materials, smooth operation, and low cost [48–50]. However, the selection of an appropriate adsorbent was always an exciting responsibility. The choice is based on several factors like availability, cost, the need for modification, mechanical stability, regenerability, non-toxicity, economic feasibility, adsorption potential, etc. In this context, it has been established that commercial adsorbents (e.g., activated carbon and silica gel) are very useful, but their high cost and regeneration problem sometimes become undesirable [51]. Thus a compromise can be often made between cost and adsorption capacity.

In this background, the utilization of domestic and agricultural wastes as adsorbents became an easy option. Many adsorbents from biomass wastes have been developed and exploited as an efficient adsorbent for the confiscation of different contaminants from water and wastewater [52, 53]. These wastes have either been exploited as such are after appropriate modification. For example, almond shell [54–56], *Azolla* [57], banana peel [58–61], biomass fly ash [62], cabbage waste [63], chitosan [64–72], *Citrus limetta* peel [2, 73, 74], *Citrus limonum* leaves [75], corn cob [76–79], *Cucumis sativus* peel [80, 81], elephant grass [82], leaves [83, 84], Bengal gram seed husk [85], *Luffa aegyptica* peel/seed [86, 87], orange peel [88–91], peanut hull [92–96], pomelo peel [97, 98], *Prunus dulcis* leaves [99, 100], *Punica granatu*m waste [101, 102], rice husk [103–107], sawdust [108–118], sugar cane bagasse [119–122], lotus seed [123], walnut shell [124, 125], etc. have been effectively used to make adsorbents for the removal of varieties of contaminants. The enormous use of these wastes as adsorbents is mainly due to their easy and cheap or priceless availability. Most of these biomasses and other materials are thrown as waste and additionally create a disposal problem. The same problem also exists for spent tea leaves or rejected tea wastes. After water, tea is the most widely consumed beverage globally, and its production and consumption are increasing continuously [126]. It is known for its good aroma, and its beverage is prepared from cured leaves of the *Camellia sinensis* plant. After making the beverage, the spent leaves become a waste like other biomasses. Due to this waste's surplus availability, spent tea leaves have attracted attention for their utilization as adsorbent. Since this waste is abundant and readily available, its transformation to an adsorbent is economically feasible, along with the additional advantage of waste handling. The scope of the present article is highlighted in the following flow chart (Fig. 1).

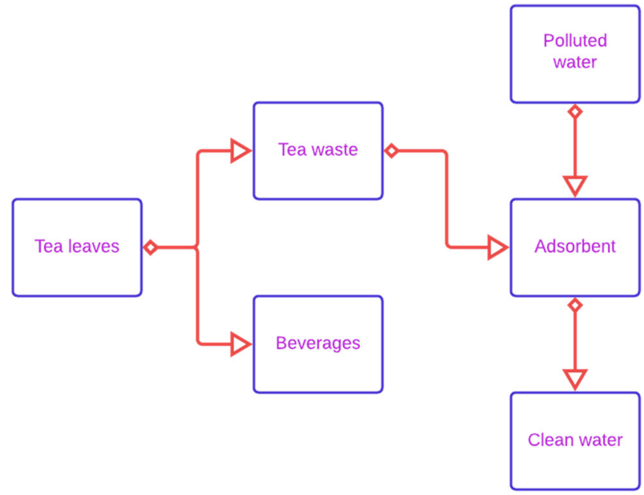


Fig. 1 Flow chart: utilization of tea waste as adsorbent for the treatment of polluted water

In addition to raw tea waste, its chemical and magnetic modification forms, along with its activated carbon, have been extensively used to treat dye polluted water. To modify, the raw tea wastes are subjected to chemical and/or thermal treatment. After the modification, the adsorbents tend to have a higher number of active binding sites, better ion exchange characteristics, and inclusion of new functional groups that favor pollutant removal. However, given the execution as an available adsorbent on a commercial basis for the treatment of polluted water, the modified forms would be undesirable because of higher cost and need for expensive apparatuses, chemicals, and skilled staff. Because of the above facts, the present article has been planned with the prime objective of reviewing the available studies on the utilization of raw tea waste as an adsorbent for the confiscation of dyes from water and wastewater. In the present article, an abbreviation, ART, will be used throughout for all those *adsorbents* that were made without any chemical or magnetic modification from *raw tea* wastes irrespective of their source. However, the source of the tea-based waste materials will also be described in the text wherever available.

2 Characterization of ART

The analytical characterizations of material play an essential role before its applications as adsorbent. The characterizations assist in relating the adsorbent properties with its claim on the adsorptive removal of different contaminants. The most common analytical methods used for the characterizations of adsorbents are point of zero charge (pHpzc); Fourier-transform infrared spectroscopy (FTIR); scanning electron microscopy (SEM); energy-dispersive X-ray (EDX); transmission electron microscopy (TEM); nuclear magnetic resonance (NMR); differential thermal analysis (DTA); thermogravimetric analysis

(TGA); differential scanning calorimetry (DSC); Brunauer, Emmett, and Teller analysis (BET); X-ray diffraction (XRD); atomic absorption spectroscopy (AAS); and spectrophotometry. Each of the above methods gives specific information, and the combination of these techniques helps to judge the suitability of materials for their potential application as adsorbent. The judicious use of a variety of a few of these techniques provides detailed surface and bulk information accompanying the materials' suitability as adsorbent. The instrumental analyses throw light on the structural, morphological, optical, and/or physicochemical features, which govern the adsorption capability and help elucidate the mechanisms involved in the process [127].

The pHpzh of an adsorbent in a solution is the pH at which the surface's net charges become zero. The adsorbent is neutral at this value, while it becomes positively charged at lower pH values and negatively charged at higher pH. Thus pHpzh plays a significant role in deciding the medium pH during the adsorption of dyes. Indeed, the role of medium pH on dye adsorption can be best judged in the light of pHpzc. For example, at lower pH, the adsorption of methylene blue by household tea waste with pHpzh of 4.3 ± 0.2 was very low due to repulsion between the cationic dye and positively charged surface as the adsorbent became positively charged through protonation of amine and carboxyl groups [128]. However, at higher pH (> pHpzc), the surface was suggested to be deprotonated. It acquired a negative charge due to the adsorption of OH^- and the carboxyl groups, and ultimately the adsorption of cationic dye was highly favored.

SEM gives information on surface morphologies, including the presence of pores, cavities, and channels. EDX coupled with SEM may be used to correlate the morphological characteristics of a material with its chemical composition and also to get a mapping of the elemental concentration profiles and perform quantitative analysis [127]. Many researchers have reported the use of SEM for surface characterizations of ART. It is a useful technique in judging the change in the surface morphology of ART after the adsorption. For instance, SEM images of ART, before and after the adsorption of acid blue 25 (an anionic dye), illustrated in Fig. 2, indicated that the waste tea residue was porous and irregular while it had comparatively fewer cavities after adsorption, which suggested the successful penetration of the dye into the pores [129]. The researchers also performed the EDX analyses on unadsorbed and adsorbed tea residue and reported the increase in weight % of carbon (from 65.93 to 69.11%) and inclusion of N (3.36%) and S (0.62%) after adsorption, which indicated the effective adsorption of dye.

FTIR is the most commonly used technique in getting information on the active sites and the presence of functional groups. It gives important information on the presence of functional groups that can be utilized for the abstraction of toxic dyes. The FTIR of tea waste shown in Fig. 3 and the

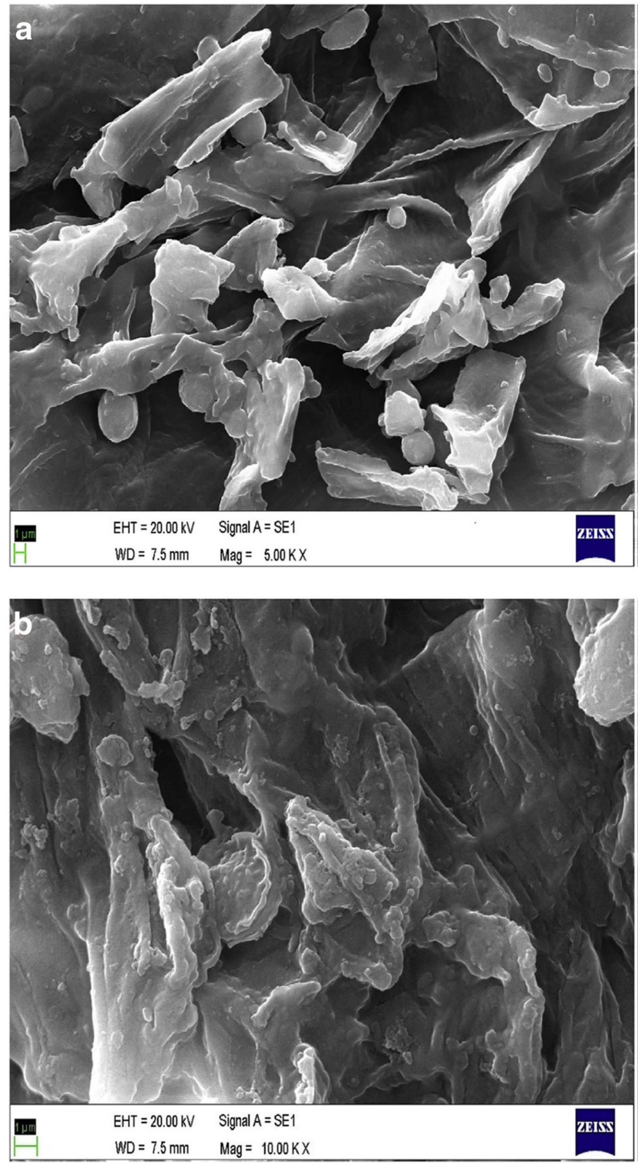


Fig. 2 SEM images of (a) unadsorbed ART and (b) acid blue 25 adsorbed ART (figure is taken from [129] with permission from the publisher

presence of –OH, –CH, C=C, and C=O, secondary amine, N–H bending, $–CH_3$ bending, C–O stretching, $–SO_3$ stretching, and P=O, C–O, C=O groups were suggested [130]. Based on shifting of peaks (3416 to 3406 cm^{-1}, 1651 to 1644 cm^{-1}, 1530 to 1537 cm^{-1}, 1371 to 1385 cm^{-1}, 1320 to 1331 cm^{-1}, 1237 to 1244 cm^{-1}) after adsorption, the authors suggested the involvement of –OH, –C=C, or C=O, amine groups, and –CH towards the interactions with MB.

The XRD studies give information on the crystalline and amorphous nature of the adsorbents. The XRD examinations of black tea samples were carried out, and the pattern showed a broad peak at 20.2° (2θ), and the absence of any sharp peak indicated the presence of significant species in the form of amorphous texture [131]. Being involved in surface phenomena, knowledge of surface properties of adsorbent is essential.

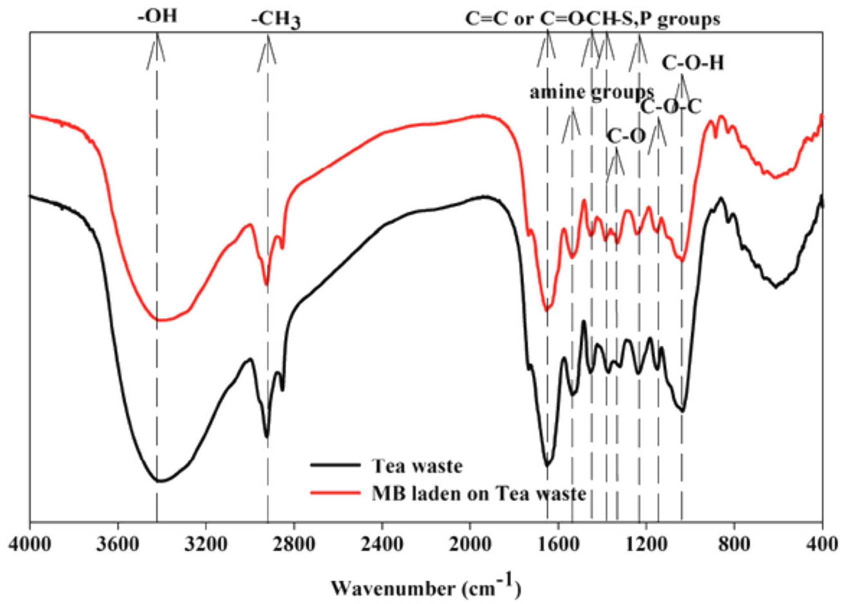


Fig. 3 FTIR spectra of ART before and after adsorption methylene blue dye—figure is reproduced from [130] as distributed by Creative Commons Attribution License

To investigate the phases, the XRD studies on tea wastes and its modified forms have been carried out by many researchers [132–136]. The BET method has been commonly used to examine the surface area. Literature indicated that a number of investigators had determined the surface area tea waste–based adsorbent. The BET surface area of biomass waste is strongly dependent on its source, method of adsorbent preparation, and other laboratory and environmental conditions. The BET surface area of tea waste materials was reported to be ranging from 0.222 to 68.82 m^2/g [129–131, 137–141]. As expected, like other biomasses, the surface area of tea wastes is very low, particularly when compared with other commercial adsorbents like activated carbon. However, the low surface area does not necessarily imply poor adsorption potential of any adsorbent [142]. The thermoanalytical techniques like DTA/TGA/DTG/DSC are used to examine the thermal stability of the material. AAS and spectrophotometry are used to determine the concentration of metals and dyes, respectively. The utilization of these and other techniques for the characterization of tea waste–based adsorbents is included in Table 1.

3 Adsorption, isotherm, and kinetic studies

In this section, the utilization of ART for the removal of different dyes will be discussed. Since, to develop a suitable adsorbate-adsorbent system, accurate knowledge of adsorption equilibrium and kinetics is essential, the literature on isotherm and kinetics is also included in this section.

ART was generally prepared by washing the tea waste many times with tap, deionized, or distilled water to eliminate dust, dirt, and other visible or invisible impurities. These samples were dried, ground to a powdered form, and then sieved to get particles of desirable size. The dry mass of the tea leaves carries hydroxyl, carboxylate, phenolic, and oxyl groups [146]. The presence of carboxylic protons, hydroxyl protons, and lactone protons in ART is accountable for its ion-exchange nature, which provides its scope to be used as an adsorbent [147]. The spent tea leaves without any chemical or physical treatment were utilized for the effective sequestration of the cationic methylene blue [126]. The authors reported a high monolayer adsorption capacity of 300.052 mg/g at 303 K. The author stated that adsorption capacity was increased with an increase in initial dye concentration, and equilibrium adsorption capacity (q_e) rose from 8.0299 to 102.1376 mg/g with the increase in initial dye concentration from 30 to 390 mg/L (Fig. 4). The initial dye concentration was suggested to provide the driving force to overcome all mass transfer resistances of the dye between the aqueous and solid phases, which resulted in the enhancement in adsorption at a higher initial dye concentration. Based on thorough analysis, the researcher concluded that the data fitted the Langmuir model better than Freundlich and Temkin isotherms and adsorption of methylene blue on ART occurred as a homogeneous monolayer surface coverage. Further, the kinetic results were best obeyed by the pseudo-second-order (PSO) model.

In another work, the removal of this cationic dye by household tea waste was also studied by carrying the batch experiments in changing laboratory parameters like initial dye concentration, contact time, adsorbent dose, and medium pH [128]. The adsorption by ART was reported to be dependent on initial dye concentrations, contact time, medium pH, and adsorbent dose. Further, the isotherm results followed the Langmuir model, confirming the monolayer adsorption with the capacity of 85.16 mg/g, and the PSO model best represented the kinetics of the process. Employing the intensive

Table 1 Characterization of ART used for removal of dyes

Dye	Original source of tea waste	Characterization techniques used	Results	Reference
Methylene blue	Spent tea bags from collected from tea stall	SEM	Adsorbent had a rough surface morphology having pores of different sizes. The dye-loaded surface was covered with dye molecules	[126]
Methylene blue	Household tea waste	pHpzc, FTIR	pHpzc = 4.3 ± 0.2 FTIR: Involvement of bonded –OH, aliphatic C–H group, C=O stretching mode conjugated with the NH_2, C–O stretching of ether group, $–SO_3$ stretching	[128]
Methylene blue	Rejected tea obtained from tea plantation	FTIR, SEM, BET	FTIR: Involvement of bonded –OH, C=O stretching, aromatic nitro compound, and –C–C– group Adsorbent had a rough surface morphology with some pores. The dye-loaded surface was covered with dye molecules BET: Surface area = 4.2 m^2/g, total pore volume = 0.0045 cm^3/g, average pore diameter = 4.3 nm	[137]
Methylene blue	Tea waste obtained from a tea factory	FTIR, SEM, XPS, solid state ^{13}C-NMR, BET	FTIR: Involvement of bonded –OH, –C=C or –C=O, –CH, amine groups in interaction with MB SEM: Adsorbent had a heterogeneous porous surface BET: Surface area = 0.913 m^2/g, pore volume = 0.007 cm^3/g, average pore diameter = 2.611 nm ^{13}C-NMR: Carbon and oxygen functional groups might be played key role during the interaction with dye	[130]
Methylene blue	Black tea	pHpzc, FTIR, XRD, BET	pHpzc = 5.55 ± 0.21 FTIR: Details are not available XRD: Major species present in adsorbent were of amorphous texture BET: Surface area = 0.738 m^2/g	[131]
Reactive green 19, reactive violet 5	Black tea from commercial tea bags	FTIR, TG/DTA	FTIR: Involvement of bonded –OH, aliphatic C–H, C=O or C=C, and C–O stretching of ether groups TG/DTA: Loss of hygroscopic water (65 °C), degradation of cellulose (300 °C) and lignin (450 °C) occurred	[143]
Congo red	Tea waste from tea plant	FTIR, SEM, BET	FTIR: Involvement of bonded –OH, aliphatic C–H, –C=N or –C=O stretching, –C–C–, amine groups SEM: Adsorbent surface had highly heterogeneous pores BET: Surface area = 4.6 m^2/g, pore volume = 0.0052 cm^3/g, average pore diameter = 4.7 nm	[138]
Acid blue 25, acid green 25	Waste tea residue of tea shops	FTIR, zeta potential, SEM/EDX, BET, DLS	FTIR: Based on shifting of the peak from 3311.78 to 3296.35 cm^{-1} (acid blue 25)/3290.56 cm^{-1} (acid green 25) belonging to the strong stretching band of the O–H or N–H group, authors suggested the involvement of hydrogen bonding in adsorption Zeta potential: Isoelectric point at pH = 5.64 SEM: The image before adsorption has the porous and irregular structure of ART, whereas comparatively fewer cavities were observed in image after adsorption, which revealed penetration of the dye into the	[129, 144]

Table 1 (continued)

Dye	Original source of tea waste	Characterization techniques used	Results	Reference
			pores and hence changed in morphology after adsorption BET: Decrease in surface area from 68.82 m^2/g (before adsorption) to 23.47 m^2/g (after adsorption) affirmed the penetration of dye leading to blockage of pores	
Acid green 25	Waste tea residue of tea shops			
Astrazon blue FGRL (AB)	Households discharged tea dust	FTIR, SEM, BET	FTIR: Involvement of bonded O–H and aliphatic C-H groups, symmetric bending of CH_3, secondary amine group, C–O stretching of ether group, and –CN– stretching SEM: The adsorbent consisted of fibers with open stomata and no considerable difference was observed in the surface morphology after the adsorption of dye BET: Surface area = 0.871 m^2/g	[139]
Crystal violet	Tea dust from tea factory	pHpzc, FTIR, SEM, BET	pHpzc = 4.2 FTIR: Involvement of stretching frequency of bonded O–H group and C–H bond of methyl and methylene groups, carbonyl group, secondary amine group, C–O and $–SO_3$ stretching SEM: The adsorbent contained fibers with pores and rough surfaces BET: Surface area = 59.9 m^2/g, pore volume = 0.031 cm^3/g, average pore diameter = 5.33 nm	[140]
Acid orange 7, basic yellow2	Tea waste from local Shop	pHpzc, FTIR, SEM	pHpzc = 5.15 FTIR: Involvement of O–H group, > C=O, secondary amino groups, C–O, C–H stretchings SEM: Rough and porous surface texture	[145]
Astrazon red 6B	Spent tea leaves and tea bags	FTIR, SEM, BET,	FTIR: Involvement of bonded O–H, aliphatic C–H, carboxyl C=O, amide, $-CH_3$, C–O ether, $–SO_3$, –CN groups, etc. SEM: TL had rough surface morphology with pores of different sizes offering suitability for dye adsorption. TB fibers were swollen after the dye adsorption BET: Surface area = 0.222 m^2/g (TL)	[141]

analysis of the adsorption/desorption experiments, the authors suggested that the adsorption was reversible and obeyed an ion-exchange mechanism. The unmodified form of the spent tea leaves (black tea) obtained from commercial tea bags was used for the adsorption study of azo dyes and observed to have very poor removal efficiency (< 10 %) [143]. However, on thermal activation of the adsorbent, considerable enhancement in the removal efficiency was observed, and it reaches nearly 100%. Since thermal treatment was also associated with significant weight loss, the authors suggested compromised heating of the adsorbent at 300 °C for 1 h when 98.8% and 72.8% of removal efficiencies were, respectively, achieved for reactive green 19 and reactive violet 5. In a similar work, the spent tea leaves of commercial tea bags were also used to remove other azo dyes, viz., reactive blue 19, reactive red 120, reactive violet 5, and reactive green 19 from wastewater [148]. The waste material obtained from the tea plantation, i.e., rejected tea, was also used to prepare adsorbent and tested to remove methylene blue [137]. The effect of initial dye concentration, contact time, temperature, adsorbent dose, and solution pH on the adsorption methylene blue by this adsorbent was thoroughly studied and reported that the adsorption capacity increases with the increase of temperature. Further, the equilibrium adsorption rises from 18.6 to 134 mg/g, with an increase in the initial dye concentration from 50 to 500 mg/L, which was suggested to be an enhancement in mass

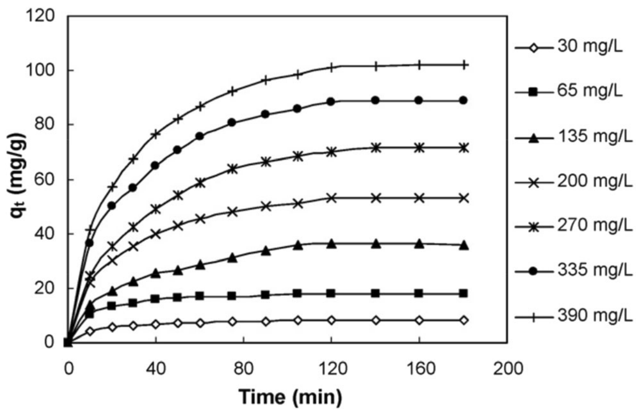


Fig. 4 Effect of contact time and initial dye concentration on the adsorption capacity ART for the confiscation of methylene blue—figure is taken from [126] with permission from the publisher

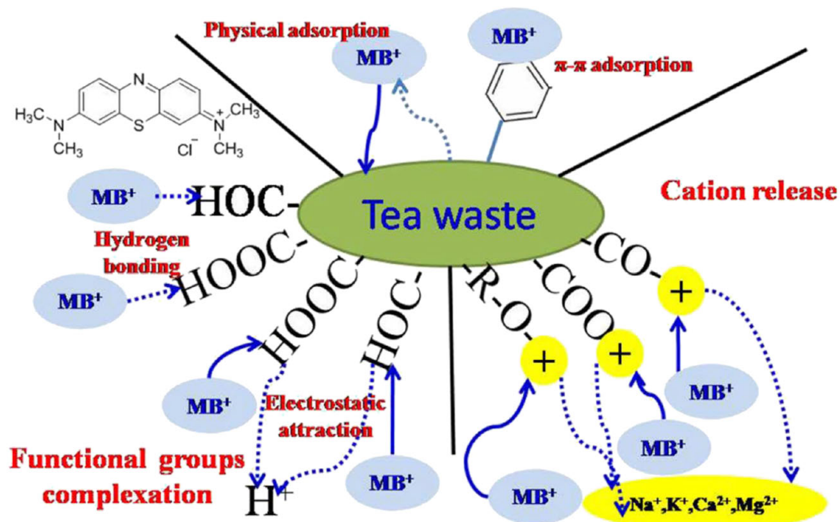
transfer driving force at higher initial dye concentration resulting from the higher dye adsorption.

In a significant work, the adsorption ability of ART was examined by using an anionic dye (Congo red) by conducting the batch experiments under the changing laboratory conditions like initial dye concentration, adsorbent dose, contact time, solution pH, and temperature and the data were analyzed in the light of different isotherm (Langmuir, Freundlich, and Temkin and kinetic (PFO, PSO, IPD) models [138]. Based on the observation of a slight decrease of adsorption at lower pH, the authors suggested the adsorption mechanism to occur by the factors involving ion exchange, hydrogen bonding, π–π stacking interaction, or chelation. In another notable work [130], the color removal of methylene blue dye by tea waste was suggested to be accomplished by different mechanisms, such as hydrogen bonds, electrostatic interaction, functional group complexation, and π–π binding and ion exchange as shown in Fig. 5.

In a very recent work, adsorption of acid blue 25 (anionic dye) onto ART was investigated by performing both batch and continuous experiments, and the experimental results were stated to be best obeyed by Redlich-Peterson (R-P) isotherm and PFO kinetic models [129]. The removal of dye was suggested to be ensured by physisorption and had good reusability of ART up to three repetitive cycles. Based on the continuous studies conducted in a packed bed, the authors also suggested the commercial applicability of this waste-based adsorbent. The removal of methylene blue has also been reported by tea wastes, and the results were followed by second-order kinetics and Langmuir isotherm with maximum adsorption capacity (q_m) of 104.9, 173.4, and 210.8 mg/g at 15, 25, and 40 °C, respectively [149].

Some researchers reported that the adsorption of dyes by ART also obey Freundlich isotherm. For example, the adsorption of Astrazon blue FGRL dye by an adsorbent made from households' discharged tea dust was well signified by the Freundlich equation indicating the adsorption-complexation reactions that occurred during the adsorption [139]. The applicability of the used black tea for the treatment of water containing Rhodamine-B was examined by carrying out the experimental studies in the batch method, and the data were interpreted in light of simple first and second order along with PSO kinetic equations [150]. This adsorption was well obeyed by PSO kinetics and Langmuir model with the q_m of 53.2 mg/g at the acidic pH of 2. The spent black tea collected from the cafeteria was employed to remove Congo red from water and influences of different laboratory parameters such as time, temperature, adsorbent dose, pH, and dye concentration on adsorption were studied [151]. A maximum dye removal of > 80% was reported to be obtained within 5 min at adsorbate concentration of 5 mg/L, adsorbent dose of 0.1 g, and medium pH of 6 at room temperature. The investigational data were suggested to be conformed to Langmuir and Freundlich isotherms.

Fig. 5 Mechanism for removing methylene blue dye by tea waste—figure is reproduced from [130] as distributed by Creative Commons Attribution License



In another significant work, the adsorbent made from tea waste collected from tea factory was analyzed by FTIR, SEM, BET, XPS, and solid-state ^{13}C-NMR and exploited for the confiscation methylene blue from water [130]. Based on the thorough analyses of the experimental data in the light of different isotherm (Langmuir, Freundlich, Temkin, Dubinin-Radushkevich) and kinetic (PFO, PSO, Elovich, two-compartment) models, the authors suggested that the adsorption might be consisted of two stages: a fast adsorption stage and a slow adsorption stage. The former stage was completed within 5 min with the removal rate of 90%. The adsorption was well followed by PSO kinetics and Langmuir isotherm with a q_m of 113.1461 mg/g. The interaction mechanism between adsorbent and adsorbate was suggested to be associated with the electrostatic attraction, ion exchange, hydrogen bond, π–π binding. Thus the presence of the organic groups in tea waste played a significant role in the adsorption of dye. In another work, black tea waste powder was utilized as an effective adsorbent for methylene blue and different experimental factors like contact time, initial dye concentration, solution pH, temperature, and adsorbent dosage thoroughly studied, and the data were examined in light of different kinetics (PFO, PSO, intraparticle diffusion, Boyd, and Elovich) and isotherm (Langmuir, Freundlich, Dubinin-Radushkevich, and Temkin) models [131]. Based on the multiple regeneration/adsorption studies, the authors reported that adsorbent remained efficiently more than 75% after five cycles using NaOH as a regenerative reagent and thus be used for many times.

The application of spent green tea leaves collected from tea processing company was demonstrated for the decolorization of raw textile wastewater samples with a true color of 868 ADMI (American Dye Manufacturers' Institute values) which demonstrated and reported that the true color removal efficiency of the raw textile wastewater was high in acidic solution and at high temperature, suggesting the endothermicity of the process [152]. A low value of 13.9 kJ/mol of activation energy specified that the process was controlled by diffusion with the physisorption mechanism. It was also reported that green tea waste adsorbent showed outstanding performance in color removal with the q_m of 775 ADMI/g at 26 °C, which was higher than that of the commercial powder-activated carbon (526 ADMI/g).

In a relatively recent work, the adsorption of crystal violet using tea dust was studied thoroughly under the different laboratory experimental conditions in batch mode [140]. The adsorption of the dye was reported to be fast during the initial 20 min, and then, it continued slowly up to 100 min and lastly attained saturation. The adsorption capacity was observed to be increased with initial dye concentrations (50 to 200 mg/L) at any time. This was suggested to be due to the predominance of mass transfer driving force at higher initial dye concentration. The adsorption data had a better correlation with PSO than PFO, while the unified approach modeled the isotherm and kinetics successfully well and suggested to be beneficial for the adsorption of crystal violet onto ART. The data were well fitted with both Freundlich and Langmuir model equations with the q_m value of 175.4 mg/g.

In a notable work, the adsorption of both acidic (acid orange 7) and basic (basic yellow 2) dyes was studied by both batch and column modes [145]. The batch studies indicated that the removal of both dyes was reliant on pH, initial dye concentration, contact time, adsorbent dose, and temperature, and the equilibrium was reached in 4 and 3 h for acid orange 7 and basic yellow 2, respectively. The authors reported that lower pH (with an optimum value of 2) was more favorable for acid orange 7 because of the protonation of phenolic and carboxylic groups. However, maximum removal of basic yellow 2 was observed at pH 6, which was suggested to be the fact that at this pH, the amino, carboxylic, and phenolic groups are not ionized and therefore the adsorption of cationic basic yellow 2 was executed by the van der Waals interactions with the electron-rich adsorbent.

Astrazon red 6B, a basic dye, was removed in batch experiments using two different adsorbents made from spent tea leaves and tea bags and proposed to be competent and inexpensive adsorbents [141]. The adsorption behavior of both adsorbents was analyzed and reported that the Langmuir model best followed the adsorption equilibrium of Astrazon red 6B dye onto the latter adsorbent. In contrast, that with former was followed well by the Freundlich isotherm. Further, the kinetics accompanying both the adsorbents were the same and well represented by PSO.

Spent tea leaves were used to examine their effectiveness in decontamination of wastewater containing crystal violet by studying the effect of different laboratory variables like adsorbent dose, solution pH, and temperature, and agitation time has been investigated [153]. The dye uptake was reported to be increased with temperature and medium pH. The PSO best represented the kinetics of adsorption, and rate constants were 8.5×10^{-3}, 22.2×10^{-3}, and 42.0×10^{-3} g/mg min for the initial adsorbate concentrations of 10, 20, and 30 mg/L, respectively. In another work, waste green tea biomass was used to study the adsorption of malachite green [154]. In this work, the experimental data were thoroughly analyzed in light of different isotherms (Langmuir, Freundlich, Dubinin-Radushkevich, and Temkin) and kinetic (PFO, PSO, intraparticle film diffusion, Elovich) models and best fittings of data were stated to be obeyed by Dubinin-Radushkevich isotherm and PSO kinetic models. The removal was suggested to be chemisorption with the Langmuir maximum monolayer coverage of 14.08 mg/g. The experimental conditions and isotherm and kinetic models accompanying the adsorption of different dyes by ART have been summarized in Table 2.

The confiscation of methyl orange from contaminated water by green tea waste of tea plant was studied by conducting batch experiments under the changing conditions of initial dye

Table 2 Experimental conditions, isotherm and kinetic models accompanying the adsorption of different dyes by ART

Dye	Experimental conditions	Optimum conditions	Isotherm models used	q_m (mg/g)	Kinetic models used	Best isotherm/kinetic models	Reference
Methylene blue	Idc: 30 to 390 mg/L AdD = 0.2 to 4.8 g/L, Time: up to 180 min pH = 2 to 11	AdD = 3.5 g/L	Lang, Frd, Tem	300.052	PFO, PSO, IPD	Lang, PSO	[126]
Methylene blue	Idc: 10 to 50 mg/L AdD = 1.0 to 3.0 g/L Time: up to 360 min pH: 2 to 10	Temperature: 27 °C AdD: 1.5 g/L pH: 8	Lang, Frd	85.16	PFO, PSO,	Lang, PSO	[128]
Reactive green 19, reactive violet 5	Idc: 50 to100 mg/L Thermal activation: 200 to 400 °C for up to 2 h	Thermal activation: 300 °C for 1 h	-	-	-	-	[143]
Methylene blue	Idc: 50 to 500 mg/L Temperature: 30 to 50 °C AdD: 0.25 to 2.5 g/L pH: 2 to 8	AdD = 2.5 g/L	Lang, Frd	147 (30 °C) 154 (40 °C) 156 (50 °C)	PFO, PSO, IPD	Lang, PSO	[137]
Congo red	Idc: 50 Tto 250 mg/L Temperature: °C AdD: 2.5 to 15 g/L pH: 3 to 13	-	Lang, Frd, Tem		PFO, PSO, IPD	Lang, PSO	[138]
Acid blue 25	Idc: 100 to 300 mg/L Temp.: 15 to 45 °C AdD: 0.5 to 5.0 g/L Stirring time: 0 to 360 min pH: 1 to 10	pH: 1 AdD: 3.5 g/L Temp: 3 °C	Lang, Tem, RP	127.14	PFO, PSO	RP, PFO	[129]
Congo red	Idc: 5 to 500 mg/L Temperature: 35 °C pH: 1 to 13	-	Lang, Frd, Tem	-		Lang, Frd	[151]
Astrazon blue FGRL (AB)	Idc: 25 to 200 mg/L Temperature: °C AdD: 0.5 to 5.0 g/L Time: up to 75 min. pH: 2 to 10	Idc: 50 mg/L Temperature: °C AdD: 5.0 g/L Time: 30 min. pH: 5	Lang, Frd	263.16	-	Frd	[139]
Methylene blue	Idc: 100 to 500 mg/L AdD: 1 to 10 g/L pH: 3 to 11	Idc: mg/L Temperature: 35 °C AdD: 4 g/L pH:	Lang, Frd, Tem, DR	113.1461	PFO, PSO, EL, TC	Lang, PSO	[130]
Methylene blue	Idc: 100 to 2200 mg/L Temperature: 15 to 50 °C AdD: 1 to 10 g/L Time: up to 120 min pH: 2 to 11	-	Lang, Frd, Tem, DR	302.63	PFO, PSO, EL, Boyd, IPD	Lang, PSO	[131]
Crystal violet	IDc: 50 to 200 mg/L Temperature: °C		Lang, Frd	175.4	PFO, PSO, unified approach model		[140]

Table 2 (continued)

Dye	Experimental conditions	Optimum conditions	Isotherm models used	q_m (mg/g)	Kinetic models used	Best isotherm/kinetic models	Reference
	AdD: 1 to 10 g/L pH:					Lang, Frd PSO, unified approach model	
Acid orange 7 (AO-7), basic yellow 2 (BY-2)	IDc: 40 to 100 mg/L (AO-7), 100 to 250 (BY-2) Temperature: 30 to 50 °C AdD: 2 to 20 g/L pH: 2 to 9	IDc: mg/L Temperature: °C AdD: 4.0 g/L (AO-7), 2.0 g/L (BY-2) pH: 2 (AO-7), 6 (BY-2) Equilibrium time: 240 min (AO-7), 180 min (BY-2)	Lang, Frd	5.73 (AO-7) 43.88 (BY-2	PFO, IPD	Lang, Frd, PFO	[145]
Astrazon red 6B	IDc: 25 to 200 mg/L AdD: 0.25 to 2 g/L pH: 2 to 10 Time: up to 240 min	IDc: 25 to 200 mg/L AdD: 0.5 g/L pH: 10 (TL), 9 (TB) Time: 90 min	Lang, Frd, Tem	4.5×10^{-3} (TL), 0.011 (TB)	PFO, PSO	Frd (TL), Lang (TB), PSO	[141]
Malachite green	IDc: 75 TO 445 mg/L Time: up to 240 min	-	Lang, Frd, Tem, DR	14.08	PFO, PSO, EL, IPD	DR, PFO	[153]

Abbrevations: *IDc* initial dye concentration, *AdD* adsorbent dose, *pHpzc* point of zero charge, *Lang* Langmuir, *Frd* Freundlich, *Tem* Temkin, *RP* Redlich-Peterson, *DR* Dubinin-Radushkevich, *PFO* pseudo-first-order, *PSO* pseudo-second-order, *IPD* intraparticle diffusion, *AFO* Avrami fractional order, *PF* power fraction, *EL* Elovich, *DLS* dynamic light scattering, *TC* two-compartment model, q_m maximum adsorption capacity

Table 3 Thermodynamic parameters accompanying the adsorption of different dyes by ART

Dye	Thermodynamic parameters			Temperature range (K)	Reference
	ΔG° (Sign)	ΔH° (kJ/mol)	ΔS° (J/K mol)		
Congo red	Negative	19.9619	80.1	303–323	[138]
Acid blue 25	Negative	17.59	230	288–318	[129]
Acid green 25	Negative	9.43	153	288–318	[144]
Astrazon blue FGRL (AB)	Positive	19.61	− 82.48	293–323	[139]
Methylene blue	Negative	− 18.50	− 18.90	288–323	[131]
Methylene blue	Negative	11.356	20.563	288–318	[149]
Acid orange 7	Negative	78.2312	336.1	293–333	[145]
Basic yellow 2	Negative	7.332	91.9	293–333	

concentration, adsorption time, temperature, and ART dose [155]. The experimental parameters were optimized by the response surface methodology based on Box-Behnken design. The optimal conditions corresponding to the maximal removal of MO (58.2%) were reported to be 9.75 mg/L (initial dye concentration), 63.8 min (contact time), and 3.90 g/L (adsorbent dose). The three-dimensional response surface plots reported the effects of parameter interaction for dye removal by the spent tea adsorbent. The experimental data modeling was also performed in the continuous fixed-bed adsorption of Congo red by tea waste [156]. The investigators studied the influence of different factors like the adsorbent mass, dye concentration, flow rate, and initial pH. They used three models, namely, Adams-Bohart, Thomas, and Yoon-Nelson, to predict the adsorption breakthrough curves of dye onto tea waste adsorbent. They observed that the Adams-Bohart model reasonably predicted the early parts of the adsorption with the correlation coefficient of about 0.9 while the Yoon-Nelson Thomas models better projected the breakthrough curves at all the investigational factors. They also applied the bed depth service time model at various bed depths and suggested the fitness of this model for the best explanation of the column data.

4 Thermodynamic studies

Since, to develop a suitable adsorbate-adsorbent system, accurate knowledge of thermodynamic is essential. The adsorption process should be thermodynamically spontaneous either in terms of exothermicity or entropy enhancement or by both. Moreover, the absorption should also be kinetically supported even in a very low contact time of adsorbent with an adsorbate. On the scale of thermodynamics, all the adsorbent processes are characterized by three parameters, namely, changes in Gibbs free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°). These parameters play a significant role in understanding the nature of adsorption as they give useful information on the feasibility and exothermicity or endothermicity of the process. Additionally, ΔS° highlights the nature of randomness at the solid-liquid interface. These three parameters are interrelated as:

$$\Delta G^{o} = \Delta H^{o} - T\Delta S^{o}$$

The parameter ΔG° is related to equilibrium constant (K_c) and given as:

$$\Delta G^{o} = -RT\ln K_c$$

where R (8.314 J/K mol) and T represent the universal gas constant and thermodynamic temperature in Kelvin, respectively. From the above equations, the following linear equations can be generated to calculate the values of ΔS° and ΔH° very easily from the intercept and slope of ln K_c versus 1/T plot.

$$\ln K_c = -\frac{\Delta H^{\circ}}{RT} + \frac{\Delta S^{\circ}}{R}$$

The thermodynamic behavior of adsorption of dyes by ART is summarized in Table 3. The thermodynamic parameters accompanying adsorption, in most cases, suggested the spontaneous and endothermic nature of the process with an increase in randomness at the solid-liquid interface. This table further indicates that the magnitude and sign of thermodynamic constants are reliant on the nature of dyes and ART. For example, the removal of methylene blue was reported to be exothermic with a decrease in randomness at the solid-liquid interface by a research group [131] while an opposite behavior, i.e., endothermicity along with enhancement in randomness (disorder) at the solid-liquid interface, was reported by another research group [149]. However, in both the above cases, the process was thermodynamically feasible.

5 Conclusions and scope for future work

Water contamination by toxic dyes has become a severe issue due to tremendous hazardous and dangerous impacts on humans and other living organisms. This review article focuses on the progress related to the confiscation of dyes from water and wastewater employing the adsorbents made from spent or rejected tea available as waste in huge amount worldwide. Such adsorbents offer ample opportunities for researchers to utilize them to treat toxic and polluted water. The effect of various experimental factors like the concentration of dye solution, adsorbent dose, adsorbate-adsorbent contact time, medium pH, and the temperature on the removal of dyes by ART as reported by different research groups has been thoroughly surveyed and discussed. In most of the published studies, excellent or satisfactory adsorption was obtained by using tea-based adsorbents. However, the extent of adsorption is strongly dependent on the nature of dyes, source of tea waste, and laboratory conditions. The reported studies were also commonly focused on the characterization of adsorbents and analyses of adsorption data in the light of equilibrium, kinetics, and thermodynamics accompanying the adsorption. The PSO kinetic and Langmuir models fitted dye removal data nicely in the majority of reported work.

Based on the in-depth analyses of literature on the topic, it has been proposed that there is a need for more research on the treatment of dye-contaminated wastewater. The suggestions for future work can be recommended as below:

1) Since spent or rejected tea leaves are widely available as waste in huge quantities, it is advisable to properly utilize them in making new biodegradable, inexpensive, efficient adsorbents on a large scale.
2) The coverage of the study must be widened, i.e., more and more dyes should be included in the study.
3) Since the maximum adsorption capacity was reported to be from very low (0.0045 mg/g) to a moderately high (300.052 mg/g), the different batch parameters should be properly optimized for the further improvement of adsorption capacity.
4) To the best knowledge of the author, all the studies have been carried out on synthetic dye solutions. Therefore there is an urgent need to focus the work on real effluent samples of different industries.
5) Since the combination of pollutants is found in industrial effluents, there is a need for research focusing on the competitive adsorption of dyes from mixtures.
6) With few exceptions, studies are generally carried out in batch mode, which does not have scope for industrial applications. Thus additional investigation on the adsorptive use of ART should be planned in continuous systems.

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