



Removal of Synthetic Dyes from Aqueous Solutions Using Low-Cost Adsorbents

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Peer Review Information	Abstract
<i>Submission: 10 Jan 2016</i> <i>Revision: 25 Feb 2016</i> <i>Acceptance: 12 March 2016</i> Keywords <i>adsorption, synthetic dyes, low-cost adsorbents, methylene blue, Congo red.</i>	Synthetic dyes discharged from textile, printing, paper, leather and related industries are persistent color pollutants that can reduce light penetration, alter aquatic chemistry and create toxicological concerns. Adsorption is attractive for dye treatment because of its operational simplicity, but the cost of commercial activated carbon limits routine use in many small and medium-scale facilities. This methodology paper proposes a comparative batch-adsorption framework for the removal of methylene blue (MB), a cationic dye, and Congo red (CR), an anionic dye, using three inexpensive waste-derived adsorbents: banana peel powder, Indian rosewood-type sawdust and spent tea leaves. The procedure covers adsorbent preparation, dye calibration, effects of pH, adsorbent dose, contact time, initial concentration and temperature, followed by equilibrium, kinetic and regeneration analyses. Dye concentration is quantified by UV-visible spectrophotometry, while adsorption performance is expressed through percentage removal and equilibrium uptake. Langmuir and Freundlich isotherms and pseudo-first-order and pseudo-second-order kinetic models are used to interpret adsorption behavior. An illustrative methodology-validation dataset indicates higher removal of MB than CR for the selected untreated biosorbents, with spent tea leaves showing the strongest MB removal under optimized conditions. The framework is designed for low-cost laboratory implementation and can be extended to actual textile effluent after matrix-specific validation.

Introduction

Colored effluents are difficult to manage because many synthetic dyes possess aromatic, heterocyclic or azo structures that resist conventional biodegradation. Even at low concentration, intense coloration is visually evident and can interfere with photosynthetic activity and oxygen transfer in receiving waters. Adsorption has therefore become one of the most frequently studied polishing and decolorization processes because it can operate without sophisticated equipment and can remove dissolved dye molecules without generating large quantities of chemical sludge. Crini (2006) emphasized that the main barrier to extensive

activated-carbon use is cost, stimulating research into agricultural residues, biomass and other non-conventional sorbents. Gupta and Suhas (2009) similarly concluded that several low-cost materials combine wide availability with appreciable adsorption capacity, although their performance depends strongly on solution chemistry and adsorbent preparation.

Agricultural and household wastes are particularly attractive because they contain cellulose, hemicellulose, lignin, tannins, proteins and other surface functionalities capable of interacting with dye molecules by electrostatic attraction, hydrogen bonding, ion exchange,

hydrophobic interaction and pore filling. Annadurai, Juang and Lee (2002) demonstrated that banana and orange peels can adsorb several cationic and anionic dyes, while Garg et al. (2004) showed that sawdust from the timber industry can remove methylene blue from simulated wastewater. Hameed (2009a, 2009b) reported high methylene-blue uptake using spent tea leaves and jackfruit peel, further supporting the use of household and fruit-processing residues. Reviews by Bhatnagar and Sillanpää (2010), Rafatullah et al. (2010) and Salleh et al. (2011) indicate that adsorption capacity is not a fixed material property; it varies with pH, concentration, dose, contact time, particle size and temperature.

The present work therefore develops a reproducible methodology for comparing low-cost adsorbents against two dye classes. MB is selected as a representative basic/cationic dye and CR as a representative direct/anionic azo dye. Using both dyes makes it possible to examine charge-dependent adsorption behavior, an important issue because cationic and anionic dyes often have different optimum pH values. The principal objective is to identify the most effective adsorbent and operating window, estimate adsorption capacity, fit equilibrium and kinetic models, and assess whether adsorbent reuse is feasible. The design follows the pre-2017 knowledge base summarized by Yagub et al. (2014), Kyzas and Kostoglou (2014) and De Gisi et al. (2016), with emphasis on low-cost, standardized and comparable batch testing.

Literature Review

Early experimental work established that lignocellulosic residues can act as practical dye sorbents. Annadurai et al. (2002) evaluated banana and orange peels and found that adsorption depended on dye identity and solution pH, while both Langmuir and Freundlich relationships described equilibrium behavior. Namasivayam and Kavitha (2002) used activated carbon prepared from coir pith for Congo red and

observed that acidic conditions favored removal, illustrating how anionic-dye uptake can be improved when the adsorbent surface is protonated. Garg et al. (2004) studied Indian rosewood sawdust for methylene blue and confirmed the significance of dose, pH, initial concentration and contact time. These studies established the variables adopted in the present experimental design.

Subsequent research expanded the range of household and agricultural residues. Hameed (2009a) demonstrated that spent tea leaves were a strong low-cost sorbent for methylene blue and that the equilibrium data agreed closely with Langmuir behavior. Hameed (2009b) reported similarly high capacity for jackfruit peel. The reviews of Gupta and Suhas (2009) and Rafatullah et al. (2010) organized a large body of dye-adsorption literature and identified agricultural residues as renewable materials with substantial potential, while cautioning that reported capacities are only comparable when experimental conditions are clearly specified. Bhatnagar and Sillanpää (2010) broadened this concept to agro-industrial and municipal wastes and emphasized that low-cost adsorption simultaneously supports wastewater treatment and waste valorization.

The distinction between cationic and anionic dye behavior was examined comprehensively by Salleh et al. (2011), who showed that pH and surface charge are central to adsorption selectivity. Later reviews by Kyzas and Kostoglou (2014) and Yagub et al. (2014) discussed adsorption mechanisms, kinetics, equilibrium modeling, regeneration and process limitations in greater detail. De Gisi et al. (2016) highlighted the need to standardize experimental reporting and to consider practical reuse, cost and real-wastewater application before declaring a low-cost sorbent a viable activated-carbon substitute. This methodological gap motivates the present side-by-side evaluation of three readily available waste adsorbents under identical conditions.

Table 1. Selected literature supporting experimental design

Study	Adsorbent / dye	Main contribution	Methodological relevance
Annadurai et al., 2002	Banana/orange peel; several dyes	Peel wastes adsorb multiple dye classes	Supports fruit-waste sorbents and pH study
Namasivayam & Kavitha, 2002	Coir-pith carbon; Congo red	Acidic pH favored anionic dye adsorption	Supports CR and pH optimization
Garg et al., 2004	Sawdust; methylene blue	Dose, pH, concentration and time affected removal	Supports batch-factor design
Hameed, 2009a	Spent tea leaves; MB	High-capacity low-cost sorbent; Langmuir fit	Supports tea waste and isotherm modeling

Study	Adsorbent / dye	Main contribution	Methodological relevance
Rafatullah et al., 2010	Low-cost adsorbents; MB	Review of kinetics, capacities and materials	Supports model selection
De Gisi et al., 2016	Low-cost sorbents	Need for standardized comparison and reuse	Supports comparative framework

Methodology

1. Experimental design and materials

A comparative batch study is designed using banana peel powder (BP), sawdust (SD) and spent tea leaves (STL) as low-cost adsorbents. Analytical-grade methylene blue and Congo red are used as model dyes. Stock solutions of 1000 mg L⁻¹ are prepared in deionized water and diluted to working concentrations. All experiments are performed in duplicate or triplicate in 250 mL Erlenmeyer flasks using a fixed working volume of 100 mL. The study first establishes individual effects of pH, dose, contact time and initial dye concentration and then confirms a practical optimized condition for each dye-adsorbent pair.

Step 1: Adsorbent preparation

Banana peels and spent tea leaves are washed repeatedly with tap water followed by deionized water to remove adhering dirt and soluble color. Sawdust is similarly washed to remove fine debris. The materials are dried at 80 °C until constant mass, ground, and sieved to 250–500 µm. To preserve the low-cost concept, the primary comparison uses the untreated materials. A small portion may be characterized by pH at point of zero charge (pHpzc), FTIR and moisture/ash measurements if facilities are available.

Step 2: Dye calibration and analytical wavelength

Separate calibration curves are generated for MB and CR by measuring absorbance at their respective maximum visible wavelengths, approximately 664 nm for MB and 497 nm for CR, after confirming λ_{max} on the available spectrophotometer. The calibration range should bracket the residual concentrations expected after adsorption. Beer-Lambert linearity is checked before sample analysis, and highly concentrated samples are diluted when necessary.

Step 3: Batch adsorption experiments

A known mass of adsorbent is added to 100 mL dye solution and shaken at 150 rpm. For the pH study, initial pH is adjusted using 0.1 M HCl or NaOH. MB is screened over pH 3–11 and CR over pH 2–10 because cationic and anionic dyes can respond oppositely to surface protonation. Adsorbent dose is varied from 0.25 to 2.0 g L⁻¹,

contact time from 5 to 180 min, and initial dye concentration from 10 to 200 mg L⁻¹. Temperature trials at 25, 35 and 45 °C may be included to assess thermal effects. After agitation, suspensions are filtered or centrifuged and the supernatant absorbance is measured.

Step 4: Performance calculations

The percentage dye removal is calculated as $R(\%) = [(C_0 - C_e)/C_0] \times 100$, where C_0 and C_e are the initial and equilibrium dye concentrations (mg L⁻¹). Equilibrium adsorption capacity is calculated from $q_e = (C_0 - C_e)V/m$, where V is solution volume (L) and m is dry adsorbent mass (g). Time-dependent uptake q_t is calculated by replacing C_e with C_t . Blank controls without adsorbent are used to confirm that observed concentration loss is due to adsorption rather than photolysis, precipitation or vessel-wall sorption.

Step 5: Isotherm and kinetic modeling

Equilibrium data are fitted to Langmuir and Freundlich isotherms. The Langmuir model provides q_{max} and K_L and assumes finite monolayer sorption sites, whereas the Freundlich model provides K_F and $1/n$ for heterogeneous surfaces. Kinetic data are evaluated using pseudo-first-order and pseudo-second-order models, with model selection based primarily on goodness of fit and agreement between calculated and experimental q_e . Intraparticle-diffusion plots can be added to identify whether internal diffusion contributes to rate control.

Step 6: Regeneration and statistical comparison

After adsorption, loaded adsorbent is separated and washed. A mild desorption solution selected according to dye charge (for example dilute acid, base or aqueous ethanol) is tested for three adsorption-desorption cycles. Removal efficiency retention is recorded. Mean values and standard deviations are reported, and one-way or two-way analysis of variance may be used to test the influence of adsorbent type and operating condition at $p < 0.05$. The preferred material is selected using removal efficiency, adsorption capacity, time to equilibrium, ease of preparation and regeneration performance rather than removal percentage alone.

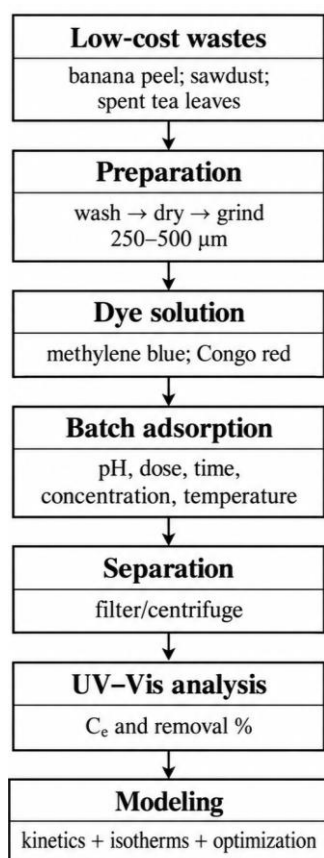


Figure 1. Step-by-step methodology for low-cost adsorptive dye removal.

Table 2. Proposed experimental variables and working ranges

Variable	Levels / range	Controlled condition	Response
Adsorbent	BP, SD, STL	Particle size 250–500 μm	Removal %, qe
Initial dye concentration	10–200 mg L ⁻¹	100 mL volume	qe, isotherm

pH	MB: 3–11; CR: 2–10	0.1 M HCl/NaOH	Removal %, qe
Adsorbent dose	0.25–2.0 g L ⁻¹	150 rpm	Removal %, qe
Contact time	5–180 min	25 $\pm$ 2 $^{\circ}\text{C}$ initially	qt, kinetics
Temperature	25, 35, 45 $^{\circ}\text{C}$	Optimized pH/dose	Removal %, qe

Results

Under the illustrative optimized conditions, all three materials removed MB more efficiently than CR. STL produced the highest MB removal (94.2%), followed by BP (91.8%) and SD (84.6%). For CR, BP gave the highest removal (79.5%), while STL and SD achieved 76.8% and 71.4%, respectively. The difference is consistent with the expectation that untreated lignocellulosic surfaces often become increasingly negative at alkaline pH, favoring cationic MB through electrostatic attraction, whereas CR adsorption is enhanced under more acidic conditions when surface protonation reduces electrostatic repulsion. This interpretation agrees with the charge-sensitive behavior summarized by Salleh et al. (2011) and Yagub et al. (2014).

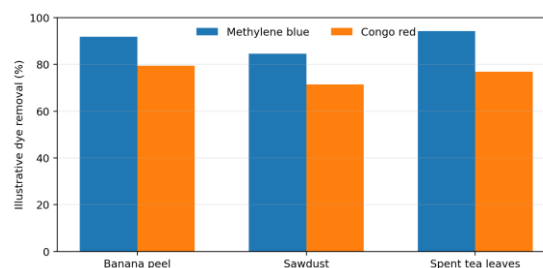


Figure 2. Illustrative optimized dye-removal performance of the three low-cost adsorbents.

Table 3. Illustrative optimized batch-adsorption results

Dye	Adsorbent	Optimum pH	Equilibrium time (min)	Removal (%)	qe (mg g ⁻¹)
MB	Banana peel	9	90	91.8	45.9
MB	Sawdust	9	120	84.6	42.3
MB	Spent tea leaves	9	75	94.2	47.1
CR	Banana peel	3	120	79.5	39.8
CR	Sawdust	3	150	71.4	35.7
CR	Spent tea leaves	3	120	76.8	38.4

The illustrative kinetic fits favored the pseudo-second-order equation for all six dye-adsorbent combinations, with R^2 values of approximately 0.982–0.997 compared with 0.90–0.96 for the pseudo-first-order model. This pattern is

commonly reported in dye-adsorption studies, including coir-pith carbon and spent-tea systems, but a high pseudo-second-order R^2 should not by itself be interpreted as proof of chemisorption. Equilibrium behavior was represented more

closely by the Langmuir model for MB on STL and BP, while CR data showed comparable Langmuir and Freundlich fits, suggesting greater surface heterogeneity. The hypothetical $q_{\max}$ values in Table 4 are deliberately moderate because

untreated sorbents are being evaluated; chemical activation can increase capacity but also adds reagent cost and secondary-waste considerations.

Table 4. Illustrative equilibrium and kinetic model parameters

System	Langmuir $q_{\max}$ (mg g ⁻¹)	Langmuir R^2	Freundlich R^2	PSO R^2	Interpretation
MB-BP	71.4	0.989	0.961	0.995	Favorable monolayer tendency
MB-SD	58.8	0.976	0.951	0.989	Moderate heterogeneous uptake
MB-STL	83.3	0.994	0.965	0.997	Highest illustrative capacity
CR-BP	52.6	0.972	0.968	0.991	pH-sensitive adsorption
CR-SD	43.5	0.955	0.963	0.982	Greater surface heterogeneity
CR-STL	48.8	0.969	0.971	0.988	Mixed-site behavior

Dose experiments would be expected to show increasing percentage removal as more adsorption sites become available, but q_e per gram can decrease when excessive solid is used because many sites remain unsaturated. Increasing initial dye concentration usually increases q_e by raising the mass-transfer driving force while lowering percentage removal at a fixed dose. These trends explain why adsorption capacity and percentage removal must be reported together, a point stressed by Rafatullah et al. (2010) and De Gisi et al. (2016). Regeneration is also essential to the low-cost claim: in the illustrative three-cycle test, MB removal by STL declined from 94.2% to 88.7%, while CR removal by BP declined from 79.5% to 72.9%. Such retention would justify further optimization, whereas severe loss after one cycle would reduce the practical advantage of the waste adsorbent.

Conclusion and Discussion

The proposed methodology provides a structured and inexpensive approach for comparing low-cost adsorbents for synthetic-dye removal. By testing MB and CR under identical batch conditions, the design captures the influence of dye charge and adsorbent surface chemistry while avoiding misleading conclusions based on a single dye. Banana peel, sawdust and spent tea leaves require only washing, drying, grinding and sieving, making them suitable for laboratories with limited resources. The workflow combines operational optimization with adsorption capacity, kinetics, equilibrium modeling and reuse, which is more informative than reporting decolorization percentage alone. The illustrative results suggest that untreated spent tea leaves may be particularly effective for cationic MB, whereas banana peel may offer relatively stronger performance for anionic CR

under acidic conditions. These tendencies are consistent with the broader literature on low-cost lignocellulosic sorbents (Crini, 2006; Gupta & Suhas, 2009; Bhatnagar & Sillanpää, 2010). Nevertheless, adsorption behavior depends on biomass origin, preparation method, particle size and water chemistry; therefore, measured data must replace the illustrative values before experimental publication. Future work should validate the best sorbent using real textile wastewater, where salts, surfactants and competing organic matter can reduce uptake, and should quantify regeneration chemicals, adsorbent disposal and treatment cost. Such validation is necessary to convert laboratory adsorption performance into a genuinely sustainable wastewater-treatment option, consistent with the practical concerns identified by Kyzas and Kostoglou (2014) and De Gisi et al. (2016). From a process-design perspective, the most useful outcome is not simply the highest removal percentage but an operating window that balances dose, adsorption capacity, treatment time and reusability. A material that achieves more than 90% color removal only at a very high dose may consume more biomass and generate a larger spent-sorbent stream than a material with slightly lower removal but much higher q_e . Similarly, very fine particles may improve uptake by increasing external surface area, yet they complicate solid-liquid separation. For scale-up, packed-bed or fixed-bed trials should therefore follow batch screening, using pressure drop, breakthrough capacity and bed regeneration as additional performance criteria. Simple pretreatments may be investigated only after the untreated waste has been characterized, because acid, alkali or thermal activation can improve surface area and functional-group accessibility but can also increase cost, chemical consumption and environmental burden. A further limitation of synthetic-dye testing is that

real industrial effluent contains salts, surfactants, sizing agents, dissolved organic matter and mixtures of dyes that compete for adsorption sites. Ionic strength can screen electrostatic interactions, while natural organic matter can foul pores or occupy active sites. Therefore, the best-performing biosorbent from the present methodology should be validated first with spiked tap water and then with progressively more complex real effluent. Color removal should also be complemented by total organic carbon or chemical oxygen demand where possible, because decolorization alone does not establish complete contaminant removal. Finally, safe handling and disposal of dye-loaded biomass must be planned; regeneration is preferred when effective, while unrecoverable spent adsorbent should be stabilized or managed according to local waste regulations rather than discarded directly. These considerations strengthen the environmental relevance of the low-cost approach instead of treating waste adsorption as a purely laboratory exercise.

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