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RESEARCH ARTICLE

EFFECT OF INITIAL ADSORBATE CONCENTRATION ON THE ADSORPTION EFFICIENCY OF METHYLENE BLUE USING LOW-COST NATURAL ADSORBENTS

A Muspira [1] , Andrews. N[2], R Lavanya[3] , Senthilkumar G P[4]* and Pradeepa A K[5]
[1]PERI Institute of Technology, Chennai - 48
[2]PERI College of Arts and Science, Chennai -48
[3]PERI College of Physiotherapy, Chennai -48
[4]PERI College of Pharmacy, Chennai -48
[5] PERI College of Nursing, Chennai -48

*Corresponding Author
Senthilkumar G P4

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Abstract: The discharge of untreated dye effluents into water bodies remains a major environmental concern due to their toxicity, persistence, and resistance to biodegradation. Adsorption using low-cost natural materials offers an effective and economical approach for dye removal. In this study, groundnut shell powder and spent tea waste were evaluated as biosorbents for the adsorption of methylene blue from aqueous solutions. Batch adsorption experiments were conducted by varying the initial dye concentration from 100 to 500 mg/L while maintaining constant adsorbent dosage (0.5 g) and contact time (100 minutes). The results showed a strong dependence of adsorption efficiency on dye concentration. Spent tea waste exhibited the highest removal efficiency of 70% at 100 mg/L, followed by a gradual decrease at higher concentrations, reaching 65% at 500 mg/L. Groundnut shell showed a fluctuating trend, with dye removal ranging from 30% at 100 mg/L, decreasing to 15% at 200 mg/L, and increasing again to 54% at 500 mg/L. The findings confirm that both natural adsorbents are suitable for methylene blue removal, with tea waste demonstrating superior performance. Optimization of parameters such as contact time, pH, and temperature could further enhance adsorption efficiency for large-scale wastewater treatment applications.

Keywords: Methylene blue; Adsorption; Initial dye concentration; Groundnut shell; Tea waste; Natural biosorbent; Wastewater treatment; Low-cost adsorbents.

INTRODUCTION

Synthetic dyes are among the most prominent pollutants found in industrial wastewater, particularly from textile, dyeing, leather, and paper manufacturing sectors. Due to their complex aromatic structures, dyes such as methylene blue are highly stable, resistant to degradation, and capable of imparting intense coloration even at low concentrations. Their presence in water bodies adversely affects aquatic ecosystems, inhibits photosynthesis, and poses risks to human health. Therefore, the removal of dyes from wastewater has become a critical environmental priority.

Among various treatment methods, adsorption is widely recognized as one of the most efficient and versatile techniques for dye removal due to its simplicity, cost-effectiveness, and high removal capacity. In recent years, the use of low-cost, plant-based biosorbents has received increasing attention as sustainable alternatives to activated carbon. Agricultural residues such as orange peels, neem leaves, coconut shells, rice husk, sesame hull, tea waste, and groundnut shells have been successfully utilized for the adsorption of methylene blue and other dyes, as reported by several researchers (Rahman et al., 2011; Santhi & Manonmani, 2012; Kyzas et al., 2013).

Groundnut shell and spent tea waste, being abundant and biodegradable, possess lignocellulosic components such as cellulose, hemicellulose, and lignin that contain functional groups capable of binding dye molecules. Their availability and minimal processing requirements make them promising candidates for wastewater treatment applications. Previous studies have demonstrated that adsorption efficiency is strongly influenced by parameters such as initial dye concentration, adsorbent dose, contact time, pH, and temperature. However, understanding the effect of initial adsorbate concentration is crucial, as it determines the driving force for mass transfer and directly impacts the adsorption capacity of the biosorbent.

The present study investigates the influence of initial methylene blue concentration on its adsorption onto two low-cost natural adsorbents—groundnut shell powder and spent tea waste. The findings aim to contribute to the development of economical and eco-friendly dye removal techniques suitable for industrial wastewater treatment.

LITERATURE REVIEW

Reviews and foundational overviews. Several comprehensive reviews synthesize the state of dye adsorption on low-cost materials and highlight the primary variables affecting performance. Rafatullah et al. (2010) provided an early, widely cited review on methylene blue adsorption using inexpensive biosorbents, summarizing typical capacities, common

experimental ranges, and the role of operating parameters. Kyzas et al. (2013) reviewed trends in adsorbent materials for dyeing wastewaters and emphasized the growing shift toward biomass-derived adsorbents. More recent broad reviews further underscore the importance of cost, sustainability, and parameter optimization in selecting adsorbents (Aragaw et al., 2021; Ho et al., 2022; Adeoye et al., 2025). These works collectively establish the theoretical and practical context for studying how initial adsorbate concentration influences adsorption performance (Rafatullah et al., 2010; Kyzas et al., 2013; Aragaw et al., 2021; Ho et al., 2022; Adeoye et al., 2025).

Effect of initial adsorbate concentration. The initial dye concentration is a critical driver of adsorption capacity and removal efficiency because it sets the concentration gradient (driving force) for mass transfer. Several experimental studies explicitly examined this effect across different low-cost adsorbents. Silva et al. (2019) and Choi et al. (2019) reported that increasing initial methylene blue concentration typically increases the amount adsorbed per unit mass (q_e) but may reduce percent removal due to site saturation at higher concentrations. Dbik et al. (2021), Jawad et al. (2020), and Garba and Waken (2020) used factorial or response-surface approaches to show clear nonlinear relationships between initial concentration, adsorbent dose, and contact time. Radoor et al. (2022) also demonstrated that initial concentration strongly modifies observed kinetics and isotherm fits in studies of eco-friendly biosorbents (Silva et al., 2019; Choi et al., 2019; Dbik et al., 2021; Jawad et al., 2020; Garba & Waken, 2020; Radoor et al., 2022). Vickneswari M et al (2025), Revathi K et al (2025), Revathi K et al (2025), Vickneswari M et al (2025), Vickneswari M et al (2025), P Priyadharshini et al (2025) and P Priyadharshini et al (2025)

Plant-based and leaf/tea/groundnut adsorbents. Numerous studies validate the use of leaf biomass, spent tea, and other agricultural residues for methylene blue removal. Silva et al. (2019) and Amar et al. (2021) reported usable uptake capacities for untreated and acid-treated seed/leaf powders, with performance sensitive to initial concentration and dose. Yilmaz et al. (2021) and Zulkurnain (2022) specifically explored tea-waste derived carbons and demonstrated improved capacity after alkali activation, while Ibrahim et al. (2014) reviewed capacities across many low-cost adsorbents and emphasized the recurring influence of initial concentration on reported capacities. Studies on agricultural wastes such as sesame cake, banana leaf, and papaya/fruit peels corroborate that raw leaf biomasses often perform well at lower concentrations but require activation/modification for high-concentration treatment (Yilmaz et al., 2021; Zulkurnain, 2022; Ibrahim et al., 2014; Rangabhashiyam et al., 2018; Mane & Bhusari, 2012).

Casuarina and related plant wastes. Casuarina biomass has been investigated as an adsorbent precursor and shows promise in methylene blue or other dye removal. Nakbanpote et al. (2017) and Zuki et al. (2017) reported that microwave or thermal treatment of *Casuarina equisetifolia* seed/biowaste improves porosity and adsorption performance; they also documented the dependence of uptake on initial dye concentration. Puspitasari et al. (2023) extended Casuarina needle studies to kinetic and thermodynamic modeling for reactive dyes, confirming the typical concentration dependence seen with other biomasses (Nakbanpote et al., 2017; Zuki et al., 2017; Puspitasari et al., 2023).

Activation, chemical modification and characterization. Chemical and thermal activation consistently improve adsorption capacity at higher initial concentrations by increasing surface area and functional site availability. Kulkarni et al. (2020) reviewed how activation (KOH, H_3PO_4, microwave) enhances pore structure; Rahmani et al. (2024) showed that an acidic modification of palm leaf waste significantly improved MB uptake across a range of initial concentrations. Zulkurnain (2022) and Garba and Waken (2020) provided experimental evidence that NaOH or acid activation of tea wastes produces activated carbons that maintain high percent removal even as initial concentration increases. These studies highlight that activation reduces the drop in percent removal typically observed at high initial adsorbate loads (Kulkarni et al., 2020; Rahmani et al., 2024; Zulkurnain, 2022; Garba & Waken, 2020).

Isotherms, kinetics and modelling approaches. Careful modeling is essential to interpret concentration effects. The pseudo-second-order kinetic model and Langmuir/Freundlich isotherms remain the most widely used frameworks to describe dye adsorption (Ho & McKay, 1999; Ibrahim et al., 2014). Box–Behnken and RSM methods have been applied to quantify the interaction between initial concentration and other parameters (Dbik et al., 2021; Yilmaz et al., 2021). Handayani et al. (2024) and Yetgin et al. (2024, 2025) provided more recent modelling studies showing that linear vs. nonlinear isotherm fitting and the chosen regression method can materially affect derived parameters when initial concentration ranges expand (Dbik et al., 2021; Yilmaz et al., 2021; Handayani et al., 2024; Yetgin et al., 2024/2025).

pH and concentration interplay, and multi-parameter studies. Since pH alters dye speciation and adsorbent surface charge, many studies treat initial concentration and pH jointly. Jawad et al. (2020) and Rangabhashiyam et al. (2018) observed that at higher initial concentrations the mitigating or amplifying effect of pH on percent removal becomes more pronounced. Rebecca studies that combine concentration, pH and dose typically find that optimal operating windows at bench scale must be carefully chosen to avoid

misleading extrapolation to industrial effluents (Jawad et al., 2020; Rangabhashiyam et al., 2018).

Recent large-scale and comparative works. Radoor et al. (2022) and Sah et al. (2022) compared numerous low-cost adsorbents under standardized concentration ranges and concluded that while many biomasses work well at low to moderate MB concentrations, only activated or composite materials sustain high percent removal at industrially relevant concentrations. Bakhtaoui et al. (2025) and Mihret et al. (2025) provide newer data on scale-up and activated carbons from agricultural residues, reinforcing that performance at high initial concentrations is achievable but generally requires activation or hybrid material design (Radoor et al., 2022; Sah et al., 2022; Bakhtaoui et al., 2025; Mihret et al., 2025).

Gaps and recommended directions. Across the literature, common gaps include limited testing with real industrial effluent (most work uses synthetic MB solutions), inconsistent reporting of concentration ranges, and under-explored regeneration/desorption performance at high initial loads. Several authors urge standardized testing ranges and inclusion of life-cycle or techno-economic assessments when reporting capacities (Rafatullah et al., 2010; Adeoye et al., 2025; Ibrahim et al., 2014). Recent modeling and design studies (Handayani et al., 2024; Yetgin et al., 2024/2025) point toward the need for predictive tools that incorporate initial concentration effects alongside pH, dose, and contact time to design robust, scalable adsorption systems.

MATERIAL AND METHODS

Materials and preparation of adsorbents. Groundnut shells and spent tea waste were collected locally. Both materials were washed several times with distilled water to remove dust and soluble impurities, boiled at 100 °C for 30 min, oven-dried at 80 °C for 48 h, crushed, and sieved through a 25-mesh sieve to obtain a uniform particle size. Prepared adsorbents were stored in airtight bottles until use.

Adsorbate preparation. A stock solution of methylene blue (MB) was prepared by dissolving 0.5 g of MB in 1 L of distilled water and stirring at 400 rpm and 80 °C for 1 h to ensure complete dissolution. Working solutions of 100, 200, 300, 400 and 500 mg·L^{-1} were prepared by appropriate dilution of the stock.

Batch adsorption experiments. Batch studies were performed in 250-mL conical flasks containing 100 mL of MB solution. For the initial concentration study, aliquots of MB solution at 100–500 mg·L^{-1} were taken and a constant adsorbent mass of 0.5 g (either spent tea powder or groundnut shell powder) was added to each flask. Flasks were agitated on a rotary shaker for 100 minutes at ambient temperature. After agitation, suspensions were centrifuged at 7000 rpm for 15 min and the supernatant was analyzed.

Analytical method and calculations. Final dye concentration was determined using a Systronics Photoelectric Colorimeter (wavelengths 315 nm and 515 nm, as appropriate for MB calibration). Percentage removal and adsorbed amount (q, mg·g^{-1}) were calculated as:

$$\% \text{ Removal} = \frac{C_0 - C_f}{C_0} \times 100$$

$$q = \frac{(C_0 - C_f)\,V}{W}$$

where C0 and Cf are initial and final concentrations (mg·L^{-1}), V is solution volume (L), and W is adsorbent mass (g). All experiments were performed in duplicate and average values reported.

RESULTS AND OBSERVATIONS:

Summary of experimental results. Table A (below) presents the observed percent MB removal at different initial concentrations for the two adsorbents using a fixed adsorbent dose of 0.5 g and a 100-minute contact time.

Table A — Percent MB removal vs. initial concentration (adsorbent dose 0.5 g, t = 100 min)

Initial [MB] (mg·L^{-1})	Spent tea powder — % removal	Groundnut shell — % removal
100	70%	30%
200	65%	15%
300	33%	33%
400	50%	43%
500	66%	54%

Observed trends and interpretation.

1. Spent tea powder performance. Spent tea powder shows high removal efficiency at the lowest initial concentration tested (70% at 100 mg•L^{-1}). Efficiency declines at mid concentration (33% at 300 mg•L^{-1}) but increases again at the highest concentration (66% at 500 mg•L^{-1}). This non-monotonic behavior suggests a combination of factors: at low concentration there is a large ratio of available adsorption sites to dye molecules (high % removal). At intermediate concentrations, partial saturation of high-affinity sites reduces percent removal. The rebound at higher concentration may indicate multilayer adsorption, changes in dye aggregation state, or experimental variability (e.g., mixing efficiency, measurement sensitivity). Additional characterization (BET surface

area, FTIR) and kinetic/isotherm fitting would clarify whether Langmuir (monolayer) or Freundlich (multilayer/heterogeneous) models are more appropriat

2. Groundnut shell performance. Groundnut shell shows lower removal at low-to-moderate concentrations (30% at 100 mg•L^{-1}, 15% at 200 mg•L^{-1}), but removal improves as concentration increases (54% at 500 mg•L^{-1}). The initial low efficiency indicates fewer readily available high-affinity sites compared with tea waste; the later improvement suggests that groundnut shell either exposes additional adsorption sites at higher dye load (perhaps via dye–dye interactions promoting deposition) or that measurement variability influenced mid-range values. Overall, groundnut shell demonstrates promise at higher concentrations but underperforms spent tea at low concentrations.
3. Comparison and practical implications. Under the chosen experimental conditions (0.5 g adsorbent, 100 min), spent tea waste is the better adsorbent at low concentrations, while groundnut shell becomes more competitive at higher concentrations. For practical wastewater treatment, percent removal alone is not sufficient: adsorption capacity (q, mg•g^{-1}) and treatment goals must be considered. If the target is high percent color removal at low influent concentrations, spent tea is preferable; for treating very concentrated dye effluents, groundnut shell (possibly after activation) may be viable..
4. Possible mechanisms. Both adsorbents are lignocellulosic and contain functional groups (–OH, –COOH, phenolic groups) that can bind MB via electrostatic attraction, hydrogen bonding and π–π interactions. Differences in porosity, surface area, and surface chemistry between tea waste and groundnut shell likely account for the different concentration-dependent behavior. Spent tea often contains polyphenols and high surface heterogeneity which provide many high-affinity sites; groundnut shell tends to be more carbonaceous and may require activation to reveal micropores.
8.
9. Limitations and sources of variability. Nonlinear trends may arise from experimental variability (mixing, measurement at two wavelengths, centrifugation efficiency), dye aggregation at higher concentrations, or insufficient equilibration time for some concentration ranges. Also, using a single adsorbent dose and contact time restricts interpretation of intrinsic capacity; fuller parametric studies (varying dose and time) and isotherm/kinetic modelling are recommended.

CONCLUSION

The study demonstrates that initial adsorbate concentration strongly influences the observed percent removal of methylene blue for both spent tea powder and groundnut shell when adsorbent mass and contact time are held constant. Under the tested conditions (0.5 g adsorbent, 100 min), spent tea powder achieved high percent removal at low MB concentration (70% at 100 mg•L^{-1}) and generally outperformed groundnut shell at lower loads. Groundnut shell showed increased removal at higher concentrations (up to 54% at 500 mg•L^{-1}), indicating different adsorption dynamics between the materials. Raw, unmodified biomasses show moderate efficacy; chemical or thermal activation and optimization of operational parameters are likely necessary to achieve high and consistent removal across a broad concentration range.

FUTURE SCOPE

1. Isotherm and kinetic modelling. Fit Langmuir, Freundlich, Temkin isotherms and pseudo-first/second order kinetics to quantitatively describe adsorption behavior and extract capacities (q_m) and rate constants.
2. Adsorbent characterization. Perform BET, SEM, FTIR and pH_pzc (point of zero charge) analyses to identify surface area, morphology and active functional groups governing adsorption.
3. Optimisation studies. Study effects of adsorbent dose, contact time, temperature and pH in a factorial or RSM design to identify optimal operating windows.
4. Activation/modification. Prepare activated carbons (chemical activation with KOH/H_3PO_4 or thermal/microwave activation) from groundnut shell and tea waste to compare raw vs. activated performance at high dye concentrations.
5. Regeneration and reuse. Investigate desorption protocols and cyclical reuse to evaluate economic feasibility and lifetime of biosorbents.
6. Real effluent testing & scale-up. Validate performance with actual textile effluents that include competing ions and other organics; carry out pilot-scale column studies.
7. Techno-economic & LCA assessment. Analyze cost, energy, and environmental impact of using raw or activated agro-wastes for dye removal at scale.

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