

# Surface chemistry and adsorption behavior of methylene blue on functionalized carbon materials: A comprehensive study

Maria Ulfa<sup>a,\*</sup>, Rizki Fauzia Hanifa<sup>a</sup>, Novia Amalia Sholeha<sup>b</sup>

<sup>a</sup>Chemistry Education Study Program, Sebelas Maret University, Surakarta 57126, Indonesia

<sup>b</sup>College of Vocational Studies, Bogor Agricultural University (IPB University), Bogor 16151, Indonesia

## Article history:

Received: 19 April 2025 / Received in revised form: 8 November 2025 / Accepted: 22 November 2025

## Abstract

The increasing release of synthetic dyes, particularly methylene blue (MB), from textile effluents has become a major environmental and health concern, highlighting the urgent need for efficient remediation strategies. Adsorption remains one of the most effective techniques for dye removal due to its simplicity, low cost, and high efficiency. This review discusses the surface chemistry and adsorption behavior of MB on functionalized carbon-based materials, emphasizing how physicochemical characteristics, surface modifications, and functional groups influence adsorption capacity and selectivity. Recent progress in developing engineered carbonaceous adsorbents—such as activated carbon (AC), graphene derivatives, carbon nanotubes, and hybrid carbon composites—has significantly improved the removal performance of MB through enhanced structural and chemical interactions. The ACHC-KOM-1 carbon composite, for instance, exhibits remarkable photocatalytic-assisted adsorption, achieving complete MB degradation under optimized conditions. Surface functional groups and pore architecture are decisive factors governing adsorption efficiency. Oxygen-containing moieties, including carboxyl (–COOH) and hydroxyl (–OH), create active sites that facilitate electrostatic attraction and hydrogen bonding with cationic MB molecules. Nitrogen functionalities (–N), introduced via heteroatom doping, enhance electron-donating properties and  $\pi$ – $\pi$  interactions between MB aromatic rings and the conjugated carbon framework, thereby strengthening molecular affinity. Pore dimensions further regulate accessibility and diffusion, with micropores (<2 nm) providing strong confinement and high adsorption energy, while mesopores (2–50 nm) promote rapid diffusion and prevent pore blockage. The synergistic combination of abundant surface functionalities and hierarchical porosity governs the overall adsorption capacity, kinetics, stability, and regeneration potential of carbon-based adsorbents for dye removal. The mechanistic framework presented here distinguishes biomass-derived and non-biomass carbon adsorbents, enabling rational design of high-performance materials. These findings offer practical optimization guidelines for industrial-scale methylene blue removal while supporting sustainable, circular-economy-aligned water purification technologies.

**Keywords:** Adsorption; carbon; methylene blue; parameters; regeneration

## 1. Introduction

Industrial operations represent major sources of water pollution, contributing up to 80% of the contaminants found in lakes, rivers, and oceans [1–3]. The global population increase from 5.3 billion to 7.8 billion over the past three decades has intensified pressure on freshwater resources, challenging the capacity to maintain clean water supplies. Among the most persistent pollutants are synthetic dyes such as methylene blue (MB), methyl orange (MO), and rhodamine red (RR), which not only discolor aquatic environments but also threaten human health and ecological balance [4–6]. The

growing demand for sustainable water purification methods has stimulated extensive research focused on developing innovative solutions to mitigate methylene blue contamination. Conventional treatment techniques—coagulation, ion exchange, adsorption, chemical precipitation, electrolysis, reverse osmosis, and electrodialysis—have been widely applied to control water pollution [7–10]. However, advances in materials science have highlighted adsorption based on carbon-derived materials as a particularly promising strategy for efficient dye removal. The present review examines recent progress in optimizing methylene blue adsorption on carbon-based adsorbents, emphasizing emerging opportunities for restoring and safeguarding global freshwater systems.

Conventional methods are insufficient for effectively

\* Corresponding author.

Email: [ulfa.maria2015@gmail.com](mailto:ulfa.maria2015@gmail.com)

<https://doi.org/10.21924/cst.10.2.2025.1711>



tackling the increasing issue of methylene blue (MB) contamination. The increasing demand for efficient water purification methods has focused considerable attention on carbon-based materials, noted for their high surface area, versatile functionality, and strong chemical reactivity [1]. Current investigations address the limitations of traditional carbon by focusing on material modification to improve physicochemical properties and overall performance. Modified carbon exhibits unique structural and functional properties that facilitate enhanced efficacy in environmental remediation [11, 12]. Composite carbon materials combine the benefits of carbon with supplementary reinforcing agents, addressing various needs in scientific and industrial contexts. Multiple studies have validated the advantages of modified carbon materials, such as increased corrosion resistance, elevated specific stiffness, enhanced thermal insulation, decreased density, and superior mechanical strength. The integration of functional materials into carbon matrices has produced advanced adsorbents tailored for complex pollutants like methylene blue, laying the groundwork for notable advancements in water treatment [13–15].

Carbon derived from the pyrolysis of biomass or non-biomass in oxygenated and inert environments, specifically within a nitrogen gas stream, is a crucial element in effective water purification systems. Techniques including hydrothermal carbonization, slow pyrolysis, and rapid pyrolysis are utilized to transform plant residues and animal waste into sustainable adsorbent sources [7]. Non-biomass carbon, produced via industrial processes and synthetic precursors, offers an alternative pathway for the development of engineered adsorbent materials. Biomass-derived carbon composites are gaining attention due to their abundant availability, cost-effectiveness, and highly porous surface morphology, making them a superior alternative to conventional activated carbon (AC) for the removal of various pollutants. Target contaminants encompass polycyclic aromatic hydrocarbons, pharmaceuticals, nutrients, pesticides, trace metals, and dyes, with methylene blue identified as one of the most persistent substances [8-13]. The significant porosity of biomass carbon composites improves adsorption efficiency, providing an effective and sustainable method for water decontamination [16-22]. The ongoing progress in carbon material research establishes these composites as both economically feasible and environmentally sustainable alternatives for enhancing global water quality.

While recent comprehensive insights, such as those presented by Bello et al. [16], have comprehensively outlined the synthesis techniques, structural categorization, and various applications of carbon-based nanomaterials for environmental remediation, this collection adopts a very different approach by concentrating explicitly on the mechanistic interplay between surface chemistry and methylene blue adsorption behavior in functionalized carbon materials. In contrast to previous studies that largely highlight materials synthesis and broad adsorbent classification, this study provides a complete analysis of how specific surface functional groups (such as carboxyl, hydroxyl, and nitrogen-containing groups) and pore characteristics (including micropores, mesopore distribution, and BET surface area) collaboratively modulate the adsorption capacity, selectivity, and kinetics of methylene

blue removal. By synthesizing contemporary experimental data (2020–2025) on molecular-level interactions—including electrostatic forces,  $\pi$ - $\pi$  folding, hydrogen bonding, and van der Waals interactions—the research elucidates fundamental principles supporting the design of high-performance adsorbents. Furthermore, it places significant emphasis on the comparative evaluation of biomass-derived versus non-biomass-derived carbon adsorbents, highlighting how precursor selection, pyrolysis parameters, and post-synthesis changes result in varying adsorption mechanisms and regeneration potentials. This mechanistic emphasis not only enhances our understanding of adsorbent-adsorbate chemistry but also provides practical insights for the rational design of next-generation carbon materials specifically targeted for sustainable wastewater treatment applications.

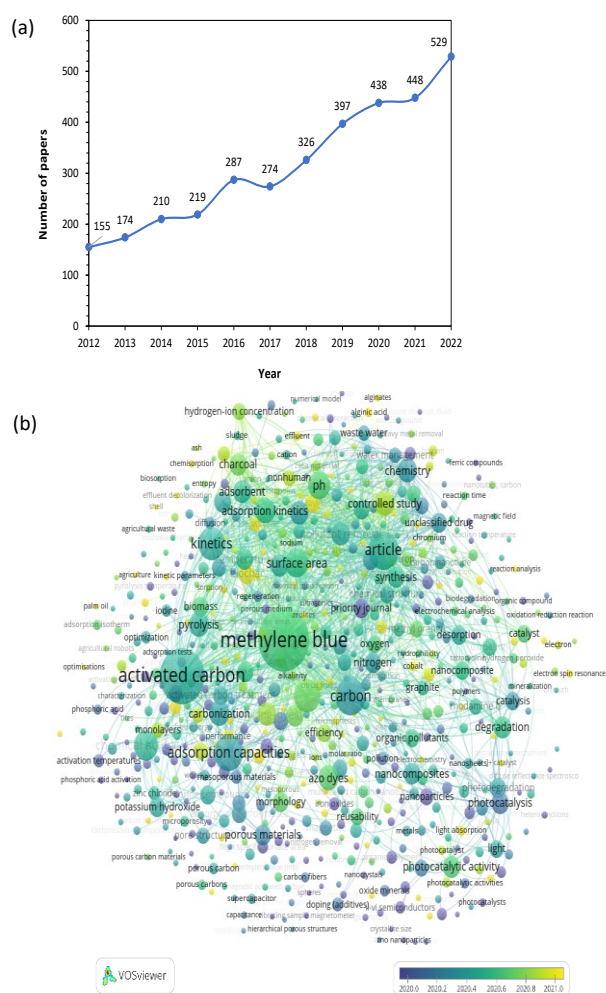


Fig. 1. Number of carbon publications for methylene blue since 2012-2022 from scopus.com and VOS viewer related carbon publication

Fig. 1 presents a visual analysis comprising two primary elements: a line graph illustrating annual publication growth and a network visualization of research keywords generated by VOS viewer. The line graph illustrates a consistent increase in publication frequency from 2012 to 2022, with a significant rise observed in the latter years. This pattern indicates an increase in academic involvement and technological advancement in the field, especially regarding activated carbon and adsorption techniques. The significant

increase in research productivity highlights a unified scientific endeavour to tackle current environmental issues using advanced purification technologies. The VOS viewer network analysis illustrates the interconnections among key research topics, including “methylene blue,” “activated carbon,” “adsorption capacities,” and “photocatalysis.” The significance of these keywords indicates a focused investigation into adsorption mechanisms and the efficiency of materials in contaminant removal. Terms like “surface area,” “kinetics,” and “carbonization” highlight the continuous enhancement of physicochemical properties, while “photocatalytic activity” and “nanocomposites” indicate a focus on the incorporation of nanotechnology and sustainable chemical design. This bibliometric assessment illustrates the intersection of chemistry, environmental science, and materials engineering in tackling global sustainability issues.

Carbon materials not derived from biomass exhibit enhanced structural and textural properties, rendering them efficient adsorbents for the removal of dyes. Examples include single-walled carbon nanotubes (SWCNTs), multi-walled carbon nanotubes (MWCNTs), ordered mesoporous carbon (OMC), and graphene. The materials demonstrate significant mechanical strength, extensive surface area, tuneable pore size, effective water transport, and superior chemical stability, which are critical for methylene blue adsorption. Graphene is a two-dimensional (2D) arrangement of  $sp^2$ -hybridized carbon atoms that exhibits exceptional chemical, mechanical, and electrical properties. Graphene possesses a specific surface area of approximately  $2620 \text{ m}^2 \text{ g}^{-1}$  and can be functionalized with oxygen-containing groups to produce graphene oxide (GO) or reduced graphene oxide (rGO), which are effective nano-adsorbents for large-scale water purification applications [23].

This review discusses recent developments in the synthesis and modification of metal oxide–carbon composites, specifically  $\text{TiO}_2$ ,  $\text{ZnO}$ , and  $\text{Fe}_3\text{O}_4$ , which demonstrate superior photocatalytic activity, chemical stability, and cost efficiency relative to alternative semiconductors. Although metal oxides face inherent challenges related to electron–hole recombination, their combination with graphene significantly alleviates this issue due to graphene's exceptional electron mobility and ability to facilitate charge separation [24,25]. Recent developments in nanotechnology and materials chemistry have facilitated the controlled, bottom-up synthesis and functionalization of carbon-based materials, enabling precise adjustments of surface properties and improved photocatalytic performance.

## 2. Carbon as An Adsorbent for Methylene Blue

Carbon-based adsorbents have attracted considerable interest in analytical chemistry because of their beneficial properties, such as high thermal stability, excellent extraction efficiency, substantial adsorption capacity, and strong selectivity. Examples comprise graphene, carbon nanotubes, and biopolymers including chitosan and biochar. Numerous wastewater treatment methods, including chemical precipitation, ion exchange, solvent extraction, nanofiltration, adsorption, and reverse osmosis, have been extensively investigated. Adsorption is notable for its simplicity, cost-effectiveness, and high removal efficiency. The efficacy of

methylene blue (MB) removal is significantly influenced by the type and source of the carbon employed. Carbon materials are classified into two categories: biomass-based and non-biomass-based, based on their precursor sources.

### 2.1. Biomass based carbon

Carbon obtained from biomass pyrolysis is acknowledged as a renewable resource, exhibiting varied physicochemical properties and extensive applications in energy production, agriculture, climate change mitigation, and environmental management [57]. Biochar derived from biomass improves soil fertility, increases crop productivity, enhances nutrient content, boosts water retention, and mitigates greenhouse gas emissions. Biomass-derived carbon demonstrates potential applications in catalysis, supercapacitor production, gas adsorption, and pollutant remediation. The utilization of organic waste as a precursor enhances sustainable waste management, rendering biomass-based carbon a viable, cost-effective, and efficient alternative for methylene blue adsorption.

The yield of carbon and its properties are affected by various factors, such as the type of biomass, the reaction medium, and the processing conditions. Pyrolysis, encompassing both slow and fast processes, serves as the principal technique for biomass conversion, with the heating rate, temperature, and residence time influencing the resultant carbon characteristics [59, 60]. Multiple raw materials, including plant residues, municipal solid waste (MSW), wood biomass, and animal manure, have been investigated as potential feedstocks. Recent studies (2018–2022) indicate positive results utilizing precursors such as Areca catechu nut powder, polyurethane elastomer–sucrose, tannins, spent coffee grounds, soybean dregs, eggshells, peanut shells, and corncobs.

Table 1 provides a comparative analysis of adsorption studies utilizing various biomass-derived and polymer-based precursors, highlighting the effects of pyrolysis temperature, residence time, and precursor type on the performance of adsorbents. Activated nanocarbons (ANCs) derived from Areca catechu nut powder exhibited a surface area of  $2132 \text{ m}^2/\text{g}$ , a pore volume of  $3.4 \text{ cc/g}$ , and a maximum adsorption capacity ( $Q_{\text{max}}$ ) of  $333.3 \text{ mg/g}$ . In a similar manner, soybean dreg-based SDB-6-K demonstrated a remarkable surface area of  $2611 \text{ m}^2/\text{g}$  and a  $Q_{\text{max}}$  of  $2636 \text{ mg/g}$ , indicating enhanced efficiency. The EFAC1 series exhibited a distinct trend of enhanced surface area and adsorption performance correlating with extended pyrolysis duration at  $1000^\circ\text{C}$ ; specifically, EFAC1 (1.6 h) achieved a surface area of  $2172 \text{ m}^2/\text{g}$ , a pore volume of  $1.08 \text{ cc/g}$ , and a  $Q_{\text{max}}$  of  $592 \text{ mg/g}$ . Adsorbents derived from spent coffee grounds (e.g., N-CGNM4) and peanut shells (PSAC) exhibited moderate capacities, indicating the influence of precursor variability. The diverse array of raw materials, including agricultural residues and polymer composites, demonstrates current initiatives aimed at creating sustainable adsorbents for environmental remediation. Results highlight the significance of green chemistry and materials engineering in developing scalable, environmentally sustainable solutions that align with circular economy and sustainability goals.

Table 1. Synthesis method of carbon and its physicochemical properties

Sample	Pyrolysis condition		Precursor	Characteristic			Ref
	T (°C)	t (h)		Surface area (m <sup>2</sup> /g)	Pore volume (cc/g)	Q <sub>max</sub> (mg/g)	
ANCs	400	3	<i>Areca catechu Nut Powder</i>	2132	3.4	333.3	[17]
EF	900	1	<i>Polyurethane elastomer -sucrose</i>	370	0.21	-	[6]
EFAC2	900	1	<i>Polyurethane elastomer -sucrose</i>	328	0.22	-	[6]
EFAC2	1000	1	<i>Polyurethane elastomer -sucrose</i>	633	0.41	-	[6]
EFAC1	1000	0.5	<i>Polyurethane elastomer -sucrose</i>	334	0.16	-	[6]
EFAC1	1000	0.8	<i>Polyurethane elastomer -sucrose</i>	896	0.43	-	[6]
EFAC1	1000	1.2	<i>Polyurethane elastomer -sucrose</i>	1058	0.50	-	[6]
EFAC1	1000	1.6	<i>Polyurethane elastomer -sucrose</i>	2172	1.08	592	[6]
EFAC1	1000	2.1	<i>Polyurethane elastomer -sucrose</i>	1421	0.71	-	[6]
TC-05	800	2	Tanin	380	0.23	5.78	[13]
CGM <sup>1</sup>	1000	1	Spent coffee grain	69	0.03	216	[18]
NCGM <sup>2</sup>	1000	1	Spent coffee grain	317	-	-	[18]
CGNM <sup>3</sup>	1000	1	Spent coffee grain	369	-	-	[18]
N-CGNM <sup>4</sup>	1000	1	Spent coffee grain	544	2.05	623	[18]
SDB-6-K	800	1	<i>dried soybean dreg powder</i>	2611	2.21	2636	[19]
CQD- CNT	850	1	<i>eggshells</i>	324	-	299.4	[20]
PSAC	800	2	<i>Peanut Shell (Dried peanut shell)</i>	868	0.459	555.6	[21]
YMA-8	900	2	<i>Corncob</i>	1004	0.899	232.2	[3]
	1000	1	<i>Orange peel</i>				[22]
	800	1	<i>Pamelo Peel</i>				[23]
	850	1	<i>Shaddoc peel</i>				[24]
	800	2	<i>Banana Peel</i>				[25]
	900	2	<i>Waste orange</i>				[26]
	800	1	<i>Cornstalk</i>				[27]
	850	1	<i>bagasse</i>				[28]

Pyrolysis, carbonization in inert gas, activation with CO<sub>2</sub>, dry carbonization, and biomass gasification are commonly utilized methods for producing carbon from natural sources. Additionally, CaCl<sub>2</sub> is incorporated as an ammonia adsorbent, and MgO serves as a hard template (Fig. 2). The standard pyrolysis process occurs at temperatures ranging from 800 to 1000 °C for a duration of 0.5 to 2 hours. While certain studies have focused on optimizing a single variable to maximize porosity, the prevailing conditions continue to favor low temperature and brief duration. Pyrolysis or carbonization at 1000 °C under an inert gas stream for a brief duration is typically preferred for time efficiency compared to lower temperatures maintained for extended periods. Excessive micropore formation can hinder adsorbate accessibility, leading to a reduced adsorption capacity when compared to carbon pyrolyzed at 1000 °C for an extended period, which facilitates complete precursor conversion and improved porosity.

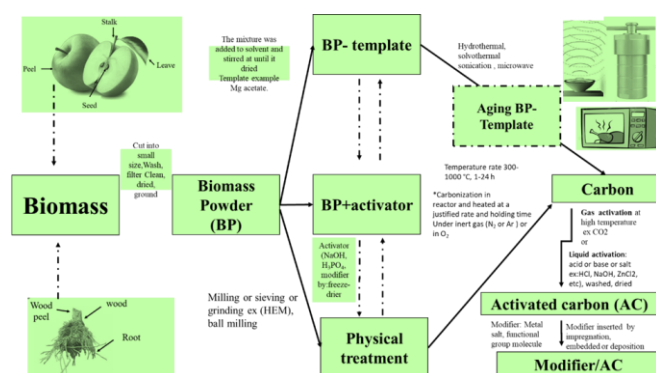


Fig. 2. Scheme of carbon production by biomass

The integration of carbon surface components or functional groups derived from natural materials has resulted in the creation of diverse composites and structural modifications. Carbon and carbon-based composites typically display porous structures that improve methylene blue adsorption capacity. XRD analysis of carbon derived from natural sources reveals peaks at 24–29° (002) and 43.5° (100), suggesting a graphite-like structure, while the broad spectra indicate the presence of an amorphous phase. The presence of modifiers significantly affects adsorption capacity. Ti-modified activated carbon (TiOAC29) demonstrated a decrease in pore size from 7 nm to 6 nm because of pore blockage, with capacities recorded at 37.63 mg/g and 479.18 mg/g, respectively. Certain samples, including ANCs with a pore size of 0.355 nm and a capacity of 333.3 mg/g, demonstrate significant adsorption despite their small pore dimensions, whereas CGM exhibited a reduced capacity of 216.33 mg/g. The primary factors distinguishing ANC (2320 m<sup>2</sup>/g and 3.4 cc/g) from CGM (69 m<sup>2</sup>/g and 0.03 cc/g) are its high surface area and pore volume. The narrow pore structure of ANC enhances chemical adsorption by facilitating stable bonding with methylene blue, thereby reducing desorption. Methylene blue molecules confined within pores exhibit strong interactions that inhibit their release, resulting in high adsorption efficiency. Conversely, adsorbents characterized by large pores and restricted surface areas exhibit rapid yet diminished physical adsorption, resulting in desorption prior to reaching equilibrium. High surface area and pore volume are the primary factors influencing methylene blue adsorption,

with pore size serving as a secondary consideration; pore dimensions near the dye's molecular size (0.9–1.2 nm) facilitate enhanced adsorption capacity.

## 2.2. Non-biomass carbon

Non-biomass carbon adsorbents are synthesized mainly from petroleum-based polymers, industrial residues, and synthetic organic compounds via controlled carbonization and activation processes. The materials demonstrate significant structural uniformity, adjustable porosity, and improved mechanical and thermal stability, rendering them appropriate for advanced environmental and industrial applications. In contrast to biomass-derived carbon, non-biomass carbon allows for precise control over physicochemical properties, including pore size distribution, surface functionality, and graphitic order, which facilitates reproducible performance in adsorption and catalysis. Production of synthetic carbon typically entails elevated energy consumption and dependence on non-renewable feedstocks, leading to increased environmental impact. Non-biomass carbons exhibit significant stability during repeated adsorption-desorption cycles and under extreme operational conditions; however, their relatively low surface reactivity and limited presence of oxygen-containing functional groups can diminish their affinity for polar contaminants like methylene blue. Current research increasingly emphasizes the hybridization of non-biomass carbons with metal oxides or heteroatom dopants to improve surface activity, thereby addressing the performance gap with more sustainable biomass-based alternatives.

Table 2. Comparison of Characteristics of Biomass and Non-Biomass Based Carbon Adsorbents for Methylene Blue Adsorption

Adsorbent Type	Precursor	Surface Area (m <sup>2</sup> /g)	Pore Volume (cc/g)	Qmax MB (mg/g)	Ref
Biochar biomassa	Areca catechu nut	2132	3.4	333.3	[17]
Biochar biomassa	Soybean dreg	2611	2.21	2636	[19]
Biochar biomassa	Spent coffee grain	544	2.05	231	[18]
Biochar biomassa	Corn cob	1004	0.89	232	[29]
Biochar biomassa	Peanut shell	868	0.45	555	[21]
Karbon non-biomassa	Graphene oxide	2620	~2.0	~350	[30]
Karbon non-biomassa	TiO <sub>2</sub> -modified AC	479	0.37	37.6-479	[31]
Karbon non-biomassa	CNTs	500–1200	1.5–3.0	100–400	[30]

Biomass-based carbon adsorbents are generally derived from agricultural waste such as peanut shells, soybeans, coffee beans, and corn cobs, which have been processed through pyrolysis and chemical/physical activation processes to produce biochar with very large surface areas and pore volumes. Biomass adsorbents offer the main advantages of abundant availability, low production costs, and contribution to circular economy efforts through the utilization of organic waste. The methylene blue adsorption capacity (Qmax) of



biomass such as soybeans even reaches >2000 mg/g, with surface and pore trends that allow for optimal absorption.

On the other hand, non-biomass carbon adsorbents, such as graphene oxide, carbon nanotubes (CNTs), and metal-modified carbon (TiO<sub>2</sub> or ZnO), have advantages in terms of structural regularity, precisely tunable pore size, and multifunctional potential (electrocatalysis, water processing, energy materials). Although their Q<sub>max</sub> is generally not as high as certain biomass, non-biomass carbon offers chemical stability, surface properties that can be further modified, and broad applications in industries that require specific and durable adsorption. Some recent research results even show that the combination of biomass and non-biomass carbon (hybrid-composite) can improve regeneration efficiency and performance in repeated applications.

In general, biomass carbon is more competitive for large-scale applications requiring cost-efficiency and sustainability, while non-biomass carbon is superior for precision engineering and multifunctional applications. Selecting the most appropriate adsorbent depends largely on the characteristics of the waste to be treated, efficiency targets, and environmental and operational cost considerations.

### 3. Carbon Structural Parameters and Synthesis Approach

Carbon demonstrates significant structural tunability and compositional adaptability, facilitating the creation of various composites with improved adsorption properties. The optimization of adsorption parameters is essential for efficient and cost-effective pollutant removal, extending beyond material design. Recent studies (2020–2022) have highlighted the advancement of adsorbents that function efficiently with low dosages, high pollutant concentrations, and brief contact times, all while achieving substantial adsorption capacities.

The data in Table 2 clearly demonstrates the correlation between structural parameters and adsorption efficiency. The specific surface area (SBET) and pore diameter (D) significantly affect adsorption performance, as indicated by removal efficiency and adsorption capacity (q). For instance, ACHC-KOH (1:1) and ACHC-KOH (1M), exhibiting SBET values of 743.8 and 703.9 m<sup>2</sup>/g, show adsorption capacities of 314.05 and 357.38 mg/g, respectively. This suggests that activation conditions and KOH concentration substantially alter pore architecture. Materials like ANC (2132.1 m<sup>2</sup>/g) and IUAC (1486.3 m<sup>2</sup>/g) demonstrate nearly complete methylene blue removal (~99%) and significant adsorption capacities (369.3 and 344.83 mg/g), indicating that well-developed pore networks and extensive surface areas significantly improve molecular diffusion and the availability of active sites.

In contrast, OLPAC has a smaller surface area (168 m<sup>2</sup>/g) and a larger pore diameter (5.18 nm), resulting in a lower adsorption capacity (38 mg/g). This underscores the necessity for a balance among surface area, pore size, and surface chemistry. Adsorbents like TWAC (878.63 m<sup>2</sup>/g) and PC5A8 (206 m<sup>2</sup>/g) demonstrate significant removal efficiencies (88.65% and 87%), indicating that adsorption behaviour is influenced by operational parameters such as contact time (t) and initial pollutant concentration. The findings highlight the

relationship between textural properties and adsorption dynamics. A strategic alignment of surface functionality, pore hierarchy, and process conditions is crucial for improving adsorption efficiency, thereby validating the potential of engineered carbon materials as sustainable and high-performance adsorbents for water purification.

Table 3. Correlation between surface area and pore diameter of carbon to adsorption

Samples	S <sub>BET</sub> (m <sup>2</sup> /g)	D (nm)	t (min)	C <sub>0</sub> (mg/L)	W (g)	V (mL)	Removal	q (mg/g)	Ref
AC-CSL	193	0.93	90	400	8	1000	97%	94.9	[32]
AC-CTW	469	0.9	90	400	0.16	25	99%	182.48	[33]
PC5A8	206	3.79	120	30	0.03	50	87%	139.72	[34]
ACHC-KOH 1:1	744	1.22	960	500	1	10	63%	314.05	[35]
ACHC-KOH 1M	704	1.5	960	500	1	10	71%	357.38	[35]
TWAC	879	2.85	360-420	300	0.02	200	89%	265.95	[36]
ANC	2132	3.29	180	300	280	1000	~99%	369.3	[17]
IUAC	1486	1.88	180	200	1.5	ND	99%	344.83	[37]
N-CM	22	24.8	4320	50	0.5	50	89%	118.98	[38]
OLPAC	168	5.18	20	100	0.02	50	96%	38	[26]

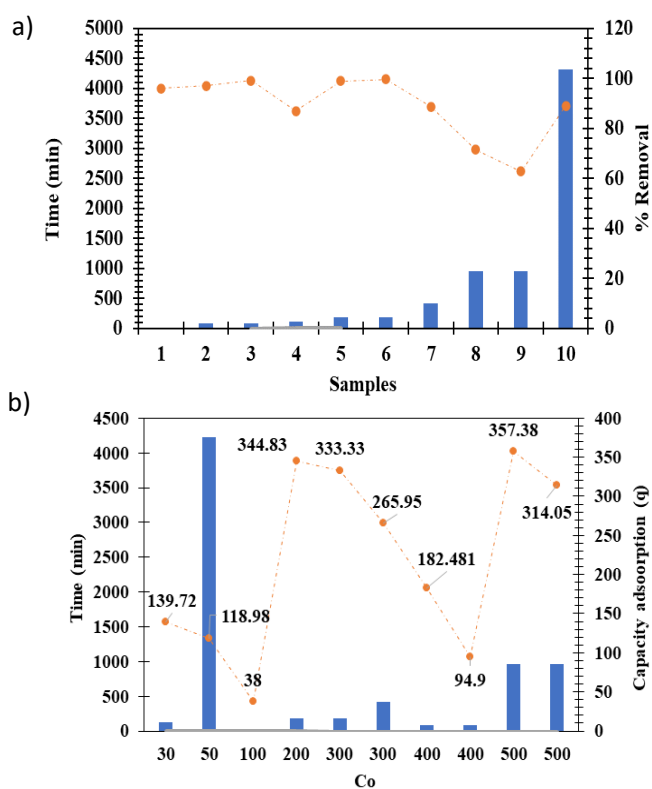


Fig. 3. (a) Relationship between contact duration and percent removal and (b) Relationship between initial concentration vs contact time and adsorption capacity

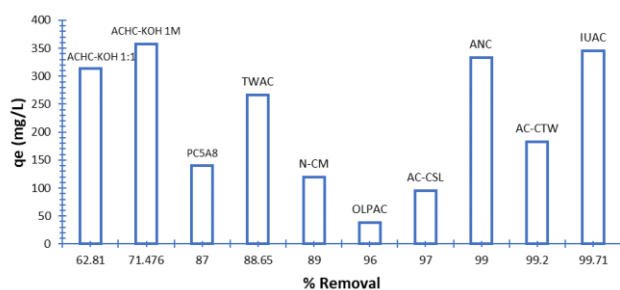


Fig. 4. Correlation of percent removal with carbon-based adsorption capacity

Table 3 and Fig. 3–4 present a summary of the relationship between condition parameters and methylene blue (MB) elimination based on studies conducted from 2020 to 2022. Samples exhibiting the highest surface area and pore volume (2131 m<sup>2</sup>/g and 3.29 cm<sup>3</sup>/g) achieved complete removal of MB, indicating enhanced adsorption performance. The ANC sample achieved maximum adsorption capacity at an adsorbent concentration of 2800 mg/mL after 180 minutes. In comparison, the N-CM sample exhibiting the lowest surface area (21.7 m<sup>2</sup>/g) and larger pore size attained an 89% removal efficiency, corresponding to 30% of the ANC capacity, while necessitating merely one-sixth of the adsorbent. The enhanced performance was attributed to nitrogen modification, which improved the electrostatic attraction between methylene blue and nitrogen–oxygen functional groups. Although N-CM exhibits high affinity, it necessitates a contact time of 72 hours, resulting in increased energy consumption. Fast-adsorbing materials like PC5A8 demonstrated superior efficiency, achieving 87% removal of MB within 120 minutes using only 25 mg of adsorbent, and exhibiting an adsorption capacity exceeding 180 mg/g. This positions PC5A8 as a more viable option for large-scale MB removal, characterized by reduced operational costs and energy requirements. Adsorption times in various studies ranged from 0.5 to 66 hours, with adsorption capacities between 20 and 350 mg/g. Most experiments utilized contact times of less than 16 hours, typically ranging from 1 to 2 hours, to avoid surface saturation and the desorption of MB molecules. Figure 5 indicates that initial concentration values ranging from 20 to 500 mg/L significantly affected adsorption capacity. Concentrations around 50 mg/L resulted in significant adsorption with moderate contact times, while 500 mg/L indicated saturation conditions for assessing maximum performance. The elevated initial concentration improved the diffusion gradient and adsorption rate until the active sites were fully occupied, while low porosity restricted adsorption efficiency.

The synthesis routes of carbon-based adsorbents typically preserve analogous structural frameworks; however, they significantly affect adsorption performance via variations in activation methods, dopant incorporation, and textural optimization. Controlled synthesis effectively minimizes contact time and adsorbent dosage while preserving high removal efficiency, thereby facilitating the advancement of cost-effective and scalable materials for the elimination of methylene blue.

### 3.1. Carbon synthesis method

Table 3 presents a summary of synthesis strategies for the

removal of methylene blue (MB), highlighting the methodological diversity in the production of effective carbon-based adsorbents. Methods including hydrothermal treatment, sol–gel processing, thermal polymerization, and chemical activation are designed to enhance adsorption capacity (Q<sub>max</sub>) and removal efficiency. The 66HP6Cs adsorbent, derived from coal tar pitch via a melamine sponge template, demonstrated a Q<sub>max</sub> of 631.0 mg/g and a 99.8% removal rate of MB. In contrast, BNC-1000, synthesized through salt templating utilizing ionic liquids and ZnCl<sub>2</sub>/NaCl, achieved a capacity of 500 mg/g with an efficiency exceeding 99%. Template-assisted synthesis effectively enhances pore structure and improves adsorption behavior by promoting uniform porosity and surface accessibility.

Advanced templating and activation methods enhance structural characteristics and adsorption efficiency. The HG@MS adsorbent, produced through a hydrothermal method using polystyrene microspheres, attained a capacity of 595.2 mg/g and an 86% removal rate of MB. Materials like TPNi, templated with Pluronic® F-127, and LSPC, which incorporates Fe<sub>2</sub>O<sub>3</sub> as a hard template, demonstrate the efficacy of hierarchical pore formation in pollutant uptake. Biomass-derived carbons, such as corn cob-based C-HPCs and lignin-based LSPC, represent sustainable synthesis methods that leverage renewable feedstocks, achieving high removal efficiency and advantageous textural properties.

Chemical modification and activation processes are crucial for enhancing adsorption performance. RF/TiO<sub>2</sub>, synthesized through sol–gel methods, exhibited a Q<sub>max</sub> of 212.56 mg/g, indicating the efficacy of hybrid metal–carbon frameworks for selective adsorption. EFAC1, synthesized from sucrose via CO<sub>2</sub>-assisted thermal polymerization, achieved a yield of 592 mg/g, thereby validating the efficacy of thermal activation in creating a high surface area and active sites. The synthesis route, precursor selection, and template design collectively determine pore distribution, surface functionality, and structural stability, forming the basis for the development of next-generation adsorbents aimed at efficient methylene blue removal and enhanced water purification.

Methods are essential to produce carbon materials that possess structural and textural properties conducive to effective methylene blue removal. The template method, utilizing either soft or hard templates, is among the most effective techniques for synthesizing highly porous and ordered carbon. Commonly employed techniques following self-assembly with soft templates, such as P123, F123, CTAB, or CTAC, include evaporation-induced self-assembly (EISA), sol–gel processes, and hydrothermal methods. The carbonization of organic precursors within rigid molds like MCM-48 and SBA-15 facilitates the production of ordered mesoporous carbon. The procedure generally consists of an aging phase of 1–72 hours, succeeded by high-temperature treatment at 900–1000 °C in an inert gas atmosphere, and template elimination utilizing NaOH. Subsequent research has shown the synthesis of ordered mesoporous carbons (OMCs) featuring uniform pore diameters through the carbonization of sucrose or furfuryl alcohol within the silica channels of SBA-15 or MCM-48. The adjustable mesoporous structure of SBA-15 enables the synthesis of nanoporous carbon with pore sizes that can be varied between 3.0 and 5.2 nm, resulting in materials characterized by significant adsorption capacity.

Carbon sources exhibit significant diversity, ranging from natural biomass to composite materials, with the majority derived from renewable resources. The production of carbon adsorbents typically requires calcination at high temperatures to eliminate impurities and enhance porosity. Techniques for template preparation, including sol–gel synthesis and salt-template pyrolysis, have exhibited significant diversification. The incorporation of sulfuric acid in carbonization enhances the process, resulting in improved yield and structural integrity. The IUAC adsorbent, synthesized through these techniques, attained a capacity of 344.83 mg/L for 200 mg/L methylene blue with just 1.5 g of material, indicating remarkable efficiency.

Multiple synthesis routes have been investigated for carbon production, including system templating (using salt or hard templates), thermal processes (such as solvothermal, hydrothermal, or CO<sub>2</sub>-activated carbonization), and homogenization methods like wet spinning. The IUAC sample demonstrates a surface area of 1,486.3 m<sup>2</sup>/g and a pore size of 1.88 nm, suggesting predominant microporosity and chemical adsorption characteristics. The ACHC:KOH 1M + AC-CTW composite exhibits greater methylene blue uptake compared to ACHC:KOH 1M alone, indicating that synergistic surface modification improves adsorption kinetics.

Table 4. Synthesis method for methylene blue removal

Sample	Method	Precursor	Aging Time (Min)	Carbonization Temperature °C	Template	Q <sub>max</sub> (mg/g)	% Removal	Ref
TPNi	EISA	Chestnut wood tannis	120 min	900	Pluronic® F-127		<b>84%</b>	[16]
RF/TiO <sub>2</sub>	Sol-gel method	TiO <sub>2</sub>	4320 min (72 h)	ND	RF gels	212.56	<b>75%</b>	[2]
HG@MS	Hydrothermal	GO frm hummers method graphene oxide	12 h	140	Microsphere polystyrene	595.2	<b>86%</b>	[39]
ACFs (EFAC1)	Thermal polymerization; direct in CO <sub>2</sub>	Sucrose ordinary table sugar	100 min	1000	Polyurethane elastomer	592	-	[40]
LSPC	Hard template	Lignin from EHR of corn stalk	2 h dab 12 h	400 dan 150	Fe <sub>2</sub> O <sub>3</sub>	423.73	<b>92.3 %</b>	[27]
66HP6Cs	carbonization and chemical activation	CTP (Coal tar Pich), Nitrogen	2 h	700	Melamine sponge	631.0	<b>99.8%</b>	[41]
C-HPCs (YMA-4)	carbonization and subsequent CO <sub>2</sub> activation process	Corn cob	2 jam, activation CO <sub>2</sub> 1 jam	900	Mg(C <sub>2</sub> H <sub>3</sub> O <sub>2</sub> ) <sub>2</sub>	230	-	[3]
CDs/MIL-88B (Fe)/Bi <sub>2</sub> S <sub>3</sub>	multistep method (Solvothermal method, thermal, hydrothermal,	Glucose	2 h	400 C	MOF (MIL-88B (Fe))		60 %	[42]
CTC <sub>4-900</sub>	programmed temperature method	Coal tar	60	900	calcium oxide	1001	-	[43]
CMC/GAs	wet spinning-	Graphene Oxide	ND	ND	CaCl <sub>2</sub>	222.72		[44]
BNC-1000	salt-templating	[EMIM][BIm <sub>4</sub> ]	2 h	1000	Ionic Liquid ZnCl + NaCl	500	<b>&gt;99%</b>	[45]



### 3.2. Parameter adsorption and its kinetic study

Table 4 presents a summary of the kinetic modeling related to the adsorption of methylene blue on carbon-based adsorbents, with a focus on the parameters of the pseudo-second-order model (PSO). Many materials demonstrate high correlation coefficients ( $R^2 \geq 0.999$ ), indicating strong alignment with the PSO model and suggesting that chemisorption is the primary mechanism involved. N,A-PCSS-1 and SDB-K-3 exhibit remarkable adsorption capacities (Equilibrium Adsorption Capacity ( $\text{mg g}^{-1}$ ) ( $q_e$ ) = 434.8 and 1155.7  $\text{mg/g}$ ), highlighting the influence of advanced carbon engineering and surface functionalization on adsorption efficacy.

The kinetics of adsorption are significantly influenced by the structural and chemical properties of the adsorbent. PET NF-MWCNTs exhibit rapid adsorption characterized by a high-rate constant ( $k = 0.2038$ ) and moderate capacity ( $q_e = 7.43 \text{ mg/g}$ ), making them suitable for efficient pollutant removal. Reduced graphene oxide (rGO) demonstrates a balanced performance, featuring a moderate rate constant ( $k = 1.4 \times 10^{-3}$ ) alongside a high capacity ( $q_e = 111.1 \text{ mg/g}$ ), highlighting the role of functionalized graphene surfaces in enhancing adsorption. Elevating initial concentrations ( $C_0$ ) in materials like AZOP-550 enhances capacity ( $q_e$  up to 222.22  $\text{mg/g}$ ), thereby confirming scalability under increased pollutant loads. SDB-K-3 exhibits a stable PSO correlation ( $R^2 \approx 1.000$ ) across different  $C_0$  values, whereas GALaO873 attains a  $q_e$  of 137.9  $\text{mg/g}$  with extended contact times, indicating its suitability for continuous or long-term treatment applications.

Kinetic analysis indicates that the PSO model provides a more accurate representation of methylene blue adsorption compared to the pseudo-first order (PFO) model, as evidenced by the linear relationship between time and adsorption capacity. The PFO model posits that surface diffusion is the rate-limiting step, while PSO kinetics suggest that chemisorption occurs via valence forces and electron transfer between the adsorbent and adsorbate. The influence of time, adsorbent mass, and initial concentration on model linearity is minimal when structural and textural advantages are present. Higher initial concentrations enhance both the rate and capacity of adsorption, suggesting more significant driving forces for dye uptake. For AZOP-550, increasing  $C_0$  from 50 to 100  $\text{mg/L}$  results in an approximate 90% enhancement of the reaction rate. Carbon-based adsorbents typically achieve equilibrium within 90 minutes, as demonstrated by HCWS3, PET NF-MWCNTs, and rGO. Extended contact durations enhance capacity, even under comparable initial conditions.

Several kinetic models were employed to evaluate the kinetic data of MB removal from carbon-based materials. The MB removal process is elucidated through pseudo-first order and second-order models, which predict the chemical kinetics of the interaction between methylene blue as an adsorbate and carbon-based materials as an adsorbent. The pseudo second order kinetic model presented in Table 4 provides a more accurate representation of the adsorption data compared to the pseudo first order kinetic model, owing to its superior linearity in the correlation between time and adsorption capacity.

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}(t) \quad (9)$$

The following pertains to the pseudo first-order concept, characterized by a rate-limiting adsorption of a target adsorbate molecule governed by heterogeneous diffusion control.

The pseudo-first-order model suggests that the adsorption of methylene blue is governed by a rate-limiting surface reaction. The robust driving force from MB molecules, along with a substantial specific surface area and elevated initial concentration, improves the overall adsorption rate. Kinetic data demonstrate that elevated initial concentrations decrease adsorption rates while concurrently enhancing overall adsorption capacity, illustrating the interplay between diffusion limitations and surface saturation effects. The pseudo-second order (PSO) plots in Table 5 illustrate distinct kinetic variations. In AZOP-550, increasing the initial concentration from 50 to 100  $\text{mg/L}$  resulted in approximately a 90% increase in reaction rate, thereby confirming a concentration-dependent enhancement in adsorption performance. An increase in methylene blue concentration enhances the rate of equilibrium and improves adsorption capacity, illustrating the effectiveness of well-structured carbon materials. A comparative analysis indicates that most adsorbents, including HCWS3, PET NF-MWCNTs, and rGO, necessitate a minimum of 90 minutes to achieve equilibrium, even when employing identical initial concentrations and comparable mass ratios. Prolonged contact duration improves the diffusion and adhesion of MB molecules to accessible active sites, highlighting the role of adsorbent structure and surface chemistry in adsorption kinetics.

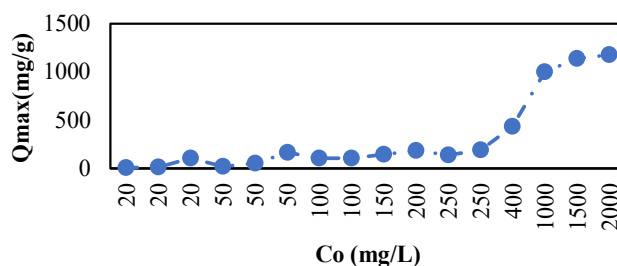


Fig. 5. The trend of adsorption capacity

### 3.3. The effect of functional groups and pore characteristics on the adsorption mechanism of methylene blue

The adsorption mechanism of methylene blue on carbon materials is synergistically influenced by the presence of surface functional groups ( $-\text{COOH}$ ,  $-\text{OH}$ ,  $-\text{N}$ ) and the pore characteristics of the adsorbent used (Fig. 6). The combination of these two aspects determines the adsorption capacity, selectivity, and process rate in real wastewater treatment applications.

At the molecular level, functional groups distributed on the carbon surface play a crucial role as active sites for interaction with methylene blue, a cationic dye often used as a model pollutant. The carboxyl group ( $-\text{COOH}$ ) is a key component that provides a negative charge on the adsorbent surface, particularly at pH values above the zero charge point ( $\text{pH}_{\text{pzc}}$ ), where it undergoes deprotonation. The negative charge on the

carboxyl group effectively attracts positively charged methylene blue molecules through electrostatic interactions, resulting in a significant increase in adsorption capacity. This mechanism is even more optimal in alkaline environments, as the number of negatively charged sites increases and competition with hydrogen ions decreases. Studies have also

shown that activation of the carbon surface with phosphoric acid ( $\text{H}_3\text{PO}_4$ ) or alkali treatment such as NaOH can increase the concentration of acidic groups on the surface, strengthening the electrostatic attraction and expanding the appropriate pore distribution [53,54].

Table 5. Kinetic Model PSO of Methylene blue adsorption on carbon

Samples	k	qe (mg/g)	qe <sub>exp</sub>	R2	Co (mg/L)	W (mg)	V (mL)	t (min)	Ref
PET NF-MWCNTs	0.2038	7.427	7.0972	0.999	20	8	30	120	[1]
rGO	$1.4 \times 10^{-3}$	111.1	104.3	0.999	20	15	100	90	[46]
	$1.33 \times 10^{-2}$	49.26	48.89	1.000	50	500	500	120	[47]
	$0.13 \times 10^{-2}$	101.01	100.08	1.000	100	500	500	120	[47]
AZOP-550	$0.03 \times 10^{-2}$	142.86	141.52	0.999	150	500	500	120	[47]
	$0.01 \times 10^{-2}$	185.19	180.59	0.999	200	500	500	120	[47]
	$0.01 \times 10^{-2}$	222.22	190.19	0.999	250	500	500	120	[47]
SPAC-8	0.200	100.0	100.2	0.999	100	10	10	ND	[24]
N,A-PCSS-1	0.00067	434.8	433.01	0.999	400	15	50	180	[34]
	0.00356	1000.0	996.37	1.000	1000	25	25	120	[48]
SDB-K-3	0.00221	1129.1	1136.83	0.9999	1500	25	25	120	[48]
	0.00086	1155.7	1174.7	0.9998	2000	25	25	120	[48]
TC	$1.11 \times 10^{-3}$	155.0	159.0	0.961	50	5 g	50	120	[49]
GALaO873	$3.88 \times 10^{-4}$	137.9	135.2	0.999	250	1 g	25	600	[50]
HCWS3	$3.78 \times 10^{-4}$	2.25	2.26	0.99	20	0.1 g	50	15	[51]
GFSA	0.0037	20.044	19.1803	0.992	50	3	50	270-360	[52]

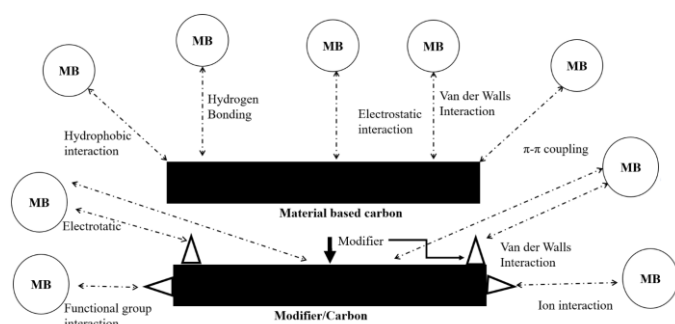


Fig. 6. The mechanism of the reaction between methylene blue and carbon adsorbents

In addition, hydroxyl ( $-\text{OH}$ ) groups, both aliphatic and phenolic, play an important role through the mechanism of hydrogen bond formation. Aliphatic hydroxyls tend to act as hydrogen bond donors, while phenolic groups act more as acceptors or contribute to electrostatic interactions. In modified lignin or carbon systems, the contribution of  $-\text{OH}$  groups to the adsorption capacity can reach more than half of the total amount of methylene blue adsorbed, where modifications to increase the amount of  $-\text{OH}$  can double or even triple the adsorption efficiency. Spectroscopic data, such as FTIR, confirm a shift in the vibrational band in the hydroxyl region after interaction with methylene blue—this is evidence of the formation of new hydrogen bonds between the adsorbent and adsorbate [55,56].

Nitrogen ( $-\text{N}$ ) groups, which can be incorporated into carbon materials through doping or chemical modification, also play a significant role. One of the main effects of the presence of nitrogen atoms in the carbon structure is an increase in the  $\pi$ -electron density and the formation of additional active adsorption sites. Nitrogen doping causes electron transfer from the carbon atom to the nitrogen atom due to the difference in electronegativity, thereby

strengthening ionic attraction or creating sites for hydrogen bonding and  $\pi$ - $\pi$  interactions. Nitrogen in the pyridinic, pyrrolic, or graphitic forms acts as an electron donor and increases the surface affinity for positively charged or aromatic molecules, including methylene blue [57,58].

The occurrence of  $\pi$ - $\pi$  stacking interactions between the aromatic structure of carbon and the aromatic ring of methylene blue also contributes to the adsorption process. This interaction occurs when the benzene ring of carbon and MB overlaps, increasing the bond through electron delocalization and shortening the intermolecular distance, thereby accelerating the adsorption kinetics and maintaining the stability of the adsorbent-adsorbate complex, especially in carbon materials with good gravity [59].

Furthermore, van der Waals forces and cation exchange contribute to the diversity of adsorption mechanisms. Van der Waals forces facilitate the initial attachment process, while cation exchange allows the migration of MB ions to the surface via ionizable hydroxyl groups. The overall mechanism is a synergistic combination of hydrogen bonding, electrostatic interactions, pore contacts, and  $\pi$ - $\pi$  stacking.

In terms of pore characteristics, the distribution and size of the adsorbent pores determine the accessibility and efficiency of the mechanisms described above. Mesopores (2–50 nm), especially those in the range of 3–3.5 nm, are ideal for methylene blue adsorption because they are comparable to the dimensions of MB molecules (approximately 0.9–1.2 nm). The mesoporous structure allows MB to easily penetrate the adsorbent and interact with the active functional groups. Carbon materials dominated by mesoporous materials have an adsorption capacity of up to 2555 mg/g, much more effective than materials that rely solely on micropores or macropores. Conversely, pore sizes that are too small (micropores  $< 2$  nm) can hinder diffusion and decrease the adsorption rate, although they can sometimes increase the strength of chemical

bonds at highly specific active sites. If the pore size is too large (macropores >50 nm), the physical interactions become weak and allow the dye to return to the solution, decreasing the adsorption efficiency [60,61].

The optimal pore distribution—a hierarchical structure with a combination of micropores, mesopores, and macropores—functions like a transport network: Macropores act as diffusion channels, mesopores facilitate access of MB molecules to the micropores, and micropores serve as high-energy adsorption sites. This combination not only provides high adsorption capacity but also accelerates the kinetics and effectiveness of material use in wastewater treatment processes.

Surface area and total pore volume remain the main parameters determining the maximum adsorption capacity of carbon materials. There is a positive correlation between the specific surface area (BET surface area) and the MB adsorption capacity, but a truly optimal pore distribution plays a more important role in the actual efficiency of the application. The ideal design of an adsorbent should lead to the dominance of mesopores with a diameter of ~3–4 nm, a large pore volume (>1 cc/g), a surface area >1000 m<sup>2</sup>/g, and a surface rich in –COOH, –OH, and –N groups [62].

#### 4. Regeneration

Table 6 provides data on the regeneration efficiency of carbon-based materials utilized for methylene blue adsorption, emphasizing aspects of reusability and sustainability. The results correspond with the study's emphasis on enhancing adsorption efficiency. Regeneration is a critical factor in assessing the practicality and economic viability of adsorbents, illustrating the impact of different methods—such as solvent batch processes, photocatalysis, and thermal regeneration—on adsorption efficiency over multiple cycles. Fe<sub>3</sub>O<sub>4</sub>@C/GO nanocomposites and PPC demonstrate high retention of removal efficiency, exceeding 95% after five cycles of repeated use. The findings validate the capability of advanced carbon composites to attain elevated adsorption performance while ensuring sustained reusability over time.

The regeneration methods outlined in Table 6 offer valuable insights into the design of carbon-based adsorbents tailored to environmental and operational contexts. Photocatalytic regeneration, as demonstrated by CuOSC-3, provides a sustainable, solvent-free method driven by sunlight, maintaining removal efficiencies greater than 93% after five cycles. Regeneration utilizing solvent-based methods with ethanol or acid solutions provides scalable and efficient alternatives. Declining efficiency in specific materials, exemplified by F-C-G beads exhibiting 61% retention by the fifth cycle, underscores the necessity for improved material stability and degradation resistance during regeneration.

Future research may combine nanotechnology with green chemistry to enhance the reusability of carbon-based adsorbents. Hybrid systems that integrate photocatalytic or magnetic components could improve regeneration efficiency. Enhancing regeneration processes to reduce energy and solvent usage will aid in sustainable material design. Table 6

illustrates the feasibility of cyclic regeneration and highlights the need for ongoing advancements in carbon material engineering to achieve effective and sustainable pollutant removal.

Batch solvent regeneration is the predominant technique employed for the reuse of methylene blue adsorbents. Common solvents comprise water, ethanol, methanol, glacial acetic acid, hydrochloric acid (HCl), and sodium hydroxide (NaOH), with organic solvents like ethanol favored for their advantageous polarity and adsorption compatibility. The immersion-based batch method facilitates extended interaction between the adsorbent and solvent, thereby improving adsorbate desorption. The PPC and Fe<sub>3</sub>O<sub>4</sub>@C/GO materials, regenerated with ethanol at a 2:1 adsorbent-to-solvent ratio, demonstrated enhanced performance across five cycles, sustaining an average removal efficiency of 95.5% with a mere 3.4% reduction per cycle. The polar groups of ethanol enhance interactions with the nitrogen moiety of methylene blue, thereby facilitating desorption more efficiently than post-adsorption carbon.

Thermal regeneration, while straightforward, necessitates significant energy input and frequently causes structural damage to the carbon framework, resulting in diminished performance in later cycles. In contrast, solvent-based regeneration maintains microstructural integrity by disrupting the physical interactions between methylene blue and carbon, avoiding structural collapse. The duration of regeneration does not inherently relate to enhanced performance; materials regenerated for one hour frequently demonstrate longer lifespans and improved adsorption recovery compared to those subjected to longer treatment durations.

The structural and textural properties of carbon materials significantly affect stability during regeneration. Table 6 presents evidence of robust regeneration performance after five cycles, especially for Fe<sub>3</sub>O<sub>4</sub>@C/GO, PPC, DFM-10, and OAC-9, which exhibit ordered frameworks and elevated surface areas. Modifiers like Fe<sub>3</sub>O<sub>4</sub> anchored on GO improve the long-term stability and retention of methylene blue removal capacity. The choice of solvent that aligns with the physicochemical properties of the adsorbent is essential for maintaining regeneration efficiency and avoiding degradation of the carbon structure over multiple cycles.

The reaction mechanism between methylene blue and carbon adsorbents encompasses various interactions, such as hydrogen bonding, pore adsorption, electrostatic attraction between oxygen atoms in carboxylate groups,  $\pi$ – $\pi$  interactions on aromatic rings, and cation exchange through hydroxyl functionalities. Van der Waals forces play a significant role in the overall adsorption process. The rising body of research on methylene blue adsorption via carbon-based materials highlights the increasing scientific and environmental significance of these systems. Recent advancements have resulted in the creation of efficient, sustainable, and cost-effective adsorbents. This progress demonstrates significant efforts to enhance adsorption mechanisms and material properties, specifically surface area, pore structure, and regeneration capacity, which are critical for effective water purification applications.

Table 6. Regeneration carbon after methylene blue adsorption

Samples	Regeneration method	Solvent	Regeneration condition	% Removal in cycles					Ref.
				1 <sup>st</sup>	2 <sup>nd</sup>	3 <sup>rd</sup>	4 <sup>th</sup>	5 <sup>th</sup>	
F-C-G beads	Solvent Batch	HCl 0.1 M	8 h	91	83	71	66	61	[63]
DFM-10	Solvent Batch	HCl/ethanol	3 h, 25C	90.05	87	85	84	83.15	[64]
CuOSC-3	Photocatalytic cycles	Water	12:00 p.m. and 2:00 p.m. for 5 sunny days	98	97.88	96	95	93	[65]
TPCNPs	Solvent Batch, sonification and using magnet	10 mL of 1 M NaOH, 10 mL ethanol, distilled water	110C, 1h	98.93	97.29	96.32	91.24	88	[66]
PPPC	Solvent Batch	ethanol	Solvent: adsorbent 2:1 Weight-to-Volume Ratio (w/v), 2h	100	97.08	96	94.60	92.3	[23]
OAC-9	Solvent Batch	Methanol and glacial acid (9:1)	Immersing MB into methanol and glacial acid)	99.17	89.24	83.2	82.41	82.34	[22]
mPVA/AC CG	Solvent Batch	ethanol	Immersing MB into ethanol, sonification and using magnet)	80	79	76	74	72.1	[67]
CaAlg/f-CNT-CD-MFA composite	Solvent Batch	Water, ethanol, 0.1 mol/L HCl	40 water + 25 HCl : 20 mg, 3h – 60 °C, Immersing MB into ethanol and HCl)	82	96	99			[68]
Fe <sub>3</sub> O <sub>4</sub> @C/GO nanocomposites	Solvent Batch	ethanol	120 min Immersing MB into ethanol)	100	100	98.85	97	95.5	[69]
BR30%-25M-10 Activated char briquettes	Thermal regeneration	ND	Drying at 50 °C for 4 h, Heated up a CO <sub>2</sub> /N <sub>2</sub> atmosphere at 800 °C for 0.5 h	93.78	92.55	89.16			[70]

The ongoing increase in research output highlights the adaptability of carbon-based materials in tackling wider pollution control issues, as shown in Fig. 7 and Fig. 8. Methylene blue functions as a model contaminant, offering a useful framework for the design of adsorbents aimed at the removal of various pollutants from aqueous environments. Recent advancements in the synthesis, modification, and functionalization of carbon materials have enhanced their potential for scalable and sustainable wastewater treatment. The ongoing trends of global industrialization and population growth highlight the necessity for materials that integrate high efficiency with environmental compatibility, facilitating widespread application in purification systems.

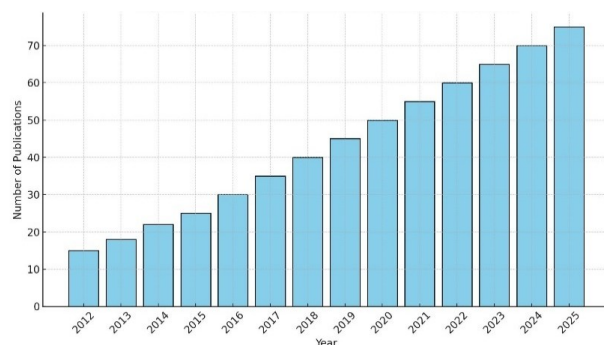


Fig. 7. Publication of methylene blue

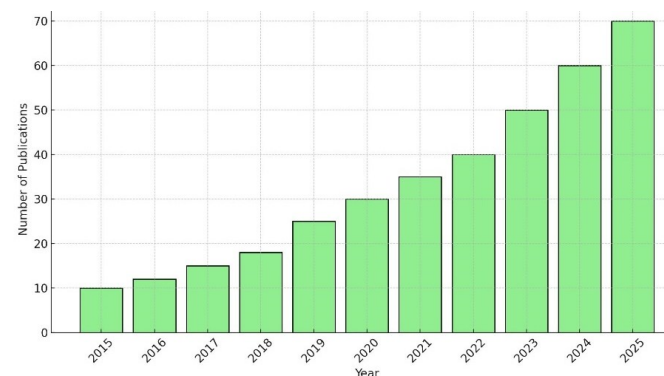


Fig. 8. publication of synthesis material for methylene blue removal

Recent innovations broaden the use of carbon-based adsorbents beyond the realm of wastewater treatment. Future research may examine functions in air purification, energy storage, and catalytic processes, thereby enhancing multifunctional environmental technologies. Enhancing comprehension of adsorption mechanisms and structure–property relationships will improve material design and performance. The increasing academic focus underscores the critical importance of carbon-based materials in advancing sustainable environmental management and enhancing ecological and human well-being worldwide.

Recent years have shown a notable increase in research activity concerning the synthesis of materials for the removal of methylene blue. The increasing number of publications in recent years indicates that the scientific community is acknowledging the essential role of advanced materials in addressing environmental challenges. The upward trend underscores the urgency of developing innovative solutions for the removal of pollutants, such as methylene blue, from wastewater, reflecting increasing concerns regarding environmental contamination and the necessity for effective remediation strategies.

Recent advancements in adsorption science have yielded carbon-based materials exhibiting increased adsorption capacity, improved structural stability, and accelerated regeneration cycles. The enhancements increase the efficiency of methylene blue removal and promote long-term operational sustainability. The integration of diverse synthesis methods, including hydrothermal, sol-gel, and hard-template techniques, illustrates adaptability to different environmental conditions, thereby rendering the materials suitable for industrial-scale water treatment. Continuous advancements in adsorbent synthesis offer considerable promise for environmental remediation in response to the escalating global water contamination resulting from industrialization and urbanization. Integrating material science with environmental engineering in interdisciplinary research can enhance the development of sustainable, high-performance technologies for large-scale pollutant removal, thereby reinforcing future strategies for water purification and ecosystem protection.

## 5. Future Aspects of Carbon Materials

The persistent issue of methylene blue contamination in aquatic environments is being addressed through innovative approaches, notably the application of carbon-based materials. Materials derived from biomass have emerged as promising candidates for efficient and cost-effective water purification. The surface area and porosity of biomass-derived carbons, including those from nut powder and soybean dregs, enhance their adsorption capacity, rendering them effective in the removal of methylene blue. The conditions of pyrolysis, such as temperature and duration, are essential for optimizing properties, facilitating the creation of adsorbents with specific characteristics aimed at enhancing pollutant removal efficiency. The adaptability of carbon-based materials is augmented through modifications that integrate advanced functional groups and composites, thereby enhancing their adsorption efficiency. The integration of metal oxides such as  $\text{TiO}_2$  and  $\text{ZnO}$  with carbon materials has enhanced photocatalytic activity and mitigated electron recombination issues, establishing new benchmarks for the efficiency of methylene blue degradation. Graphene, characterized by its high surface area, is gaining attention in the field for its remarkable adsorption properties, facilitating potential large-scale applications in water treatment. The advancement of the field reveals the interaction among green chemistry principles, nanotechnology, and material science, which consistently unveils new possibilities. Research on carbon-based adsorbents provides sustainable solutions to global water pollution and demonstrates significant advancements toward

cleaner ecosystems for future generations.

Future research must extend beyond laboratory-scale evaluations to rigorously assess the scalability, operational stability, and environmental safety of biomass-derived carbon adsorbents in realistic wastewater conditions. Industrial effluents frequently comprise intricate combinations of dyes, heavy metals, and organic pollutants that can compete for active sites, thereby diminishing adsorption efficiency—an issue that has not been adequately explored. Systematic investigations into regeneration performance, adsorption-desorption cycles, and the structural integrity of adsorbents following repeated use are essential for evaluating long-term feasibility. Furthermore, integrating carbon-based materials with advanced treatment methods like photocatalysis, electrochemical oxidation, or membrane filtration may produce synergistic effects for concurrent adsorption and degradation. Prioritizing comprehensive techno-economic and life-cycle assessments is essential to ensure that emerging materials comply with the principles of green chemistry and the circular economy. Integrating multi-functionalized carbon composites into scalable treatment systems is essential for transforming promising laboratory results into robust, sustainable technologies for industrial-scale water purification.

### 5.1. Future perspectives

The development of carbon adsorbents for water and air pollution remediation continues to undergo rapid evolution, with various promising research directions opening up significant opportunities for improving efficiency, sustainability, and future industrial applicability [71]. One key trend gaining increasing attention is the application of artificial intelligence (AI) and machine learning in the design and optimization of carbon adsorbent materials [72]. This data-driven approach enables rapid prediction of  $\text{CO}_2$  and methylene blue adsorption capacity based on parameters such as surface area, micropore volume, temperature, and pressure, with high accuracy ( $R^2 > 0.94$ ) without the need for expensive and time-consuming repeated experiments. Hybrid models such as ANN-XGBoost have even demonstrated predictive capabilities with  $R^2$  reaching 0.97, enabling more efficient material screening and design of next-generation adsorbents [73]. The integration of genetic algorithms with machine learning also paves the way for systematic optimization of activated carbon synthesis parameters, reducing experimental effort while maximizing adsorption performance in carbon capture, water purification, and energy storage applications.

On the materials side, the development of multifunctional hybrid carbon composites is a very promising research focus to expand the application of adsorbents beyond traditional structural functions. Hybrid composites that combine carbon fibers with carbon nanotubes (CNTs), graphene, or metal-modified carbon particles (such as  $\text{TiO}_2$ ,  $\text{Fe}_3\text{O}_4$ ) are able to integrate mechanical properties, energy storage capabilities, as well as sensor and electromagnetic interference (EMI) shielding functions in a single material. Recent research has shown that carbon fiber composites reinforced with CNTs or biomass-modified carbon (e.g., from coconut shells) not only improve thermal stability and resistance to water absorption,



but also offer effective EMI shielding suitable for industrial applications. This approach is in line with the trend towards smart material systems that can adapt to environmental conditions and provide multifunctional responses.

However, the main challenges that must be overcome to realize large-scale industrial applications are the economic aspects, scalability, and long-term stability of carbon adsorbents. Although activated carbon from biomass offers lower production costs and high sustainability, large-scale production processes still face challenges related to heat and mass transfer, pressure drop, mechanical integrity, and product quality consistency. Furthermore, regeneration and recycling of saturated adsorbents are crucial issues for maintaining the economic and environmental sustainability of adsorption technology. The development of low-energy regenerating adsorbents with good hydrothermal and oxidative stability is a priority for future research. Alternative synthesis strategies that avoid hazardous organic solvents and utilize abundant metals and low-cost organic units also need to be explored to accelerate the commercialization of carbon adsorbents in post-combustion carbon capture and wastewater treatment [74–75].

Moving forward, the development of porous carbon adsorbents from biomass for CO<sub>2</sub> capture and organic pollutant remediation will increasingly rely on machine learning-based rational design approaches, with a focus on pore structure optimization, surface functionalization through heteroatom doping (nitrogen, sulfur, oxygen), and the formulation of particle, monolith, or fiber shapes suitable for industrial implementation. Research will also continue to explore AI-designed generative carbon materials with CO<sub>2</sub> capacities exceeding 96% of the existing hypothetical material database, accelerating the identification of high-performance adsorbent candidates. Furthermore, the integration of carbon adsorbents with other carbon capture technologies, such as membranes and pressure/temperature adsorption-desorption (PSA/TSA)-based systems, will be a critical area for improving overall system efficiency and lowering operational costs. Thus, the combination of materials innovation, intelligent computing approaches, and a well-thought-out scale-up strategy will be key to successfully realizing carbon adsorbents as practical and sustainable solutions to future global environmental challenges.

## 6. Conclusion

This study confirms that the use of biomass and non-biomass-based carbon materials as adsorbents provides an effective solution to reduce methylene blue pollution in waters. Carbon from biomass pyrolysis such as soybeans, peanut shells, and coffee grounds shows a very high adsorption capacity, for example, dried soybean dreg powder can reach a Q<sub>max</sub> of up to 2636 mg/g, while non-biomass carbon such as TiO<sub>2</sub>-modified activated carbon offers stability and efficiency in repeated applications with a capacity range of 37–479 mg/g, depending on the engineering and modifications used. This quantitative comparison shows that optimal surface area and pore volume, especially in biomass carbon, significantly increase the amount of methylene blue that can be adsorbed by more than tenfold compared to

conventional commercial carbon, although characteristics such as chemical resistance and multifunctional potential are more superior for non-biomass carbon. Kinetics and isotherm parameter studies revealed that the adsorption mechanism is predominantly influenced by a combination of electrostatic interactions, hydrogen bonding,  $\pi$ -stacking, and the role of surface functional groups, with mesopore pore distribution (3–4 nm) and BET area above 1000 m<sup>2</sup>/g as key factors for effectiveness. The selection, design, and modification of carbon adsorbents based on application needs—both efficiency, regeneration, and sustainability aspects—are key strategies to address the challenges of current and future industrial dye wastewater treatment

## Acknowledgements

The authors would like to express their sincere gratitude to Sebelas Maret University for the financial support provided through the Penelitian Unggulan Terapan A scheme under contract number 369/UN27.22/PT.01.03/2025. This support has been instrumental in facilitating the research activities and development presented in this study. The authors extend their heartfelt appreciation to the university for the resources, guidance, and opportunities that have significantly contributed to the successful completion of this work.

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