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Recent Advances in Methylene blue removal using plant-derived adsorbents

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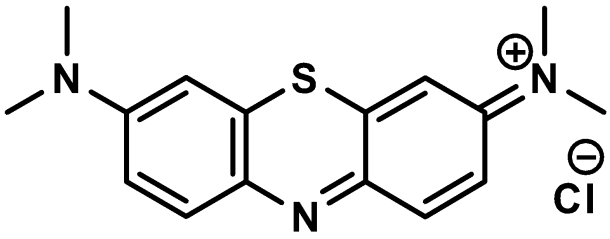
ABSTRACT

Methylene blue (MB) is a widely used synthetic dye that can cause environmental concerns when released into wastewater. Among various treatment methods, adsorption has received considerable attention for its simplicity and effective dye removal. This review examines plant-derived materials and agricultural wastes as adsorbents for MB removal, including rice residues, pine-derived materials, duckweed, watermelon waste, spent coffee grounds, fruit peels, water hyacinth, and peanut-derived materials. The reviewed studies show that both minimally processed biomass and modified materials can effectively adsorb MB. Various synthetic methods for adsorbent materials include pyrolysis, chemical activation, magnetic modification, and the formation of composites or beads. Depending on the preparation methods, adsorption performance can vary, and the adsorbents can also be easily recovered and reused. However, differences in experimental conditions make direct comparisons among studies difficult, and many studies are limited to batch experiments on MB removal. Overall, plant-derived adsorbents show promising potential for the removal of MB, but further studies regarding standardized conditions, real wastewater, continuous-flow systems, and longer-term regeneration studies are needed.

1. INTRODUCTION

Methylene blue (MB), a synthetic cationic dye, has strong coloration and chemical stability, which makes it widely used in industrial, medical, and scientific fields [1–10]. Environmental concerns can be raised when MB-containing wastewater is released into aquatic systems. Because MB can affect water quality and aquatic environments, effective methods for its removal from contaminated water are needed.

Figure 1. The structure of Methylene Blue



Various physical, chemical, and biological methods have been investigated for MB removal [11]. Because it requires relatively simple operation and shows high potential for MB removal, adsorption has received considerable attention [12–32]. Although activated carbon is widely used as an adsorbent, its production and regeneration can increase treatment costs. This leads to the development of alternative adsorbents produced from more renewable and inexpensive materials.

Agricultural wastes and other plant-derived materials are promising sources of adsorbents for MB [12–32]. This is because they are widely available and may contain functional groups that interact with MB. These materials can be used directly or modified via treatments such as pyrolysis, chemical activation, magnetic modification, or incorporation into composite materials. This review discusses plant-derived adsorbents for MB removal, focusing on how they are prepared, how well they adsorb MB, how they can be modified, and whether they can be recovered and reused for wastewater treatment.

2. METHODS

Google Scholar was used to collect 111 initial articles published between 2010 and 2026. Keywords such as "Methylene blue; removal; adsorption"; "Methylene blue; removal; bioadsorption"; "Methylene blue; removal; biosorbents"; "Methylene blue; removal; adsorbents" were used. Articles published before 2010 were removed due to focusing on more recent studies. After the screening, 88 articles remained for further evaluation. Articles are included only if the articles were peer-reviewed and investigates plant-derived or agricultural-waste-derived absorbents for methylene blue removal and reported absorption performance. Articles that did not discuss plant-derived absorbents or methylene blue absorption were discarded. A total of 19 papers were selected for inclusion in this review. The selected literature (i.e., references #14 to #32) consists of peer-reviewed articles.

3. APPLICATIONS AND ENVIRONMENTAL IMPACT OF METHYLENE BLUE

MB has been widely used in industrial and scientific fields due to strong color, chemical stability, redox properties, and ability to interact with biological tissues [1,2,8–10]. MB also has several medical and diagnostic applications [3–7]. Acquired methemoglobinemia, a condition in which hemoglobin is oxidized and becomes less able to carry oxygen effectively, has been treated with MB [3]. It has also been used as a surgical and diagnostic dye to improve the visualization of anatomical structures and has been

investigated for fluorescence-guided imaging [3]. In addition, MB can act as a photosensitizer in antimicrobial photodynamic therapy, in which light activation generates reactive oxygen species that damage microorganisms [4]. When MB is released into wastewater, it can negatively affect aquatic environments [1]. Light penetration in water could be diminished due to its strong coloration, and its chemical stability can make removal of MB from wastewater challenging. Exposure to high concentrations may also harm aquatic organisms and human health. These concerns highlight the need to remove MB effectively from contaminated water.

4. GENERAL STRATEGIES FOR METHYLENE BLUE REMOVAL

Various treatment technologies have been investigated for the removal of MB from contaminated water [11–32]. Various factors, such as dye concentration, wastewater composition, removal efficiency, cost, and environmental impact, need to be considered to select appropriate treatment. Because no single method is suitable for all wastewater conditions, physical, chemical, and biological approaches have been developed for MB removal.

Table 1. Comparison of common methods for methylene blue removal

Method	Principle	Advantages	Limitations
Coagulation-flocculation	Aggregation of contaminants into removable flocs	Relatively simple operation	Chemical use and sludge generation
Membrane filtration	Physical separation through membranes	High removal efficiency	Membrane fouling and maintenance costs
Biological treatment	Microbial degradation of contaminants	Mild operating conditions	Longer treatment time; limited degradation of stable dyes
Advanced oxidation processes	Oxidation by highly reactive species	Effective dye degradation	May require chemicals, a catalyst, or energy
Photocatalytic degradation	Light-assisted catalytic degradation	Can degrade dye molecules	Requires a suitable catalyst and light source

Adsorption	Transfer of MB from water to an adsorbent	Simple operation; relatively low energy requirement	Potential adsorbent recovery and regeneration

Common methods to remove MB from wastewater include coagulation-flocculation, membrane filtration, biological treatment, advanced oxidation processes, photocatalytic degradation, and adsorption [11]. Each method has its own mechanism and limitations. Coagulation-flocculation and membrane filtration physically remove MB from wastewater. However, sludge can be formed or regular membrane maintenance is required. Advanced oxidation processes and photocatalytic methods can eventually degrade MB into substances which can be easily removed, but they require additional chemicals, catalysts, or energy. Biological treatment can work under relatively mild conditions, but the chemical stability of MB may result in slower or incomplete degradation.

Among the MB removal methods, adsorption has received considerable attention due to its relatively simple operation, effective dye removal, and low energy requirements [12–32]. During adsorption, MB molecules are transferred from the aqueous solution to the surface or internal structure of an adsorbent. Activated carbon has been commonly used owing to its high surface area and adsorption performance. However, the production and regeneration costs have prompted efforts to develop alternative adsorbent materials.

Plant-derived materials and agricultural by-products have therefore been increasingly investigated as renewable alternatives for MB adsorption. These materials can contain functional groups that interact with MB, while their use may also provide a pathway for utilizing agricultural and food wastes. Plant-derived adsorbents and their reported performance are discussed in the following section.

5. REMOVAL OF METHYLENE BLUE USING PLANT-DERIVED ADSORBENTS

5.1. Rice-Derived Materials

Rice straw and rice husk are agricultural by-products that have been investigated as low-cost adsorbents for MB. Jawad et al. used rice straw powder directly as a biosorbent and reported a maximum adsorption capacity of 158 mg/g [14]. MB adsorption occurred through electrostatic attraction, *n*–π interactions, and hydrogen bonding with functional groups on the rice straw surface. The point of zero charge (pH_{pzc}) of the rice straw biosorbent (RSB) was found to be 6.1, so the authors predicted that MB removal would be favorable at pH values higher than the measured pH_{pzc}. RSB carries negative charges and is expected to engage in electrostatic interactions with MB. However, it was observed that the adsorption capacity of RSB did not increase noticeably above pH 7.0. Rice-derived materials have also been incorporated into engineered composite adsorbents. Alver et al. prepared magnetic alginate/rice husk beads and reported a maximum experimental MB adsorption capacity of 274.9 mg/g [15]. The authors also conducted studies to assess the effect of pH on MB removal and found no significant changes within the pH range of 6–10.

The addition of magnetic properties to the adsorbents allowed them to be easily separated from the solution after adsorption. This feature demonstrates how composite formation can improve adsorbent recovery.

Rice-derived adsorbents have also been evaluated in continuous adsorption systems. Singh et al. modified rice straw powder with a 0.1 M sodium hydroxide solution and conducted continuous-flow sorption studies using a fixed-bed column with the rice-derived materials [16]. An MB adsorption capacity of 21.99 mg/g was reported using their system. Hummadi et al. compared untreated and torrefied rice husk and found that torrefaction increased the maximum equilibrium adsorption amount from 21.5 to 38.0 mg/g in batch experiments [17]. They also demonstrated MB removal using an inverse fluidized-bed column, which achieved 84% overall removal with torrefied rice husk.

5.2. Pine-Derived Materials

Pine-derived waste, including bark and pinecones, has been investigated as a low-cost adsorbent for MB removal. Saadallah et al. reported Algerian pine bark as an inexpensive, sustainable adsorbent [18]. The characterization of the adsorbent showed an ideal porous structure and functional groups for adsorption. The adsorption studies resulted in a maximum removal efficiency of 99.47% under optimized conditions and an adsorption capacity of 37.15 mg/g. Pinecones have also shown their potential to be promising adsorbents. Solmaz evaluated *Pinus nigra* Arn. Powder from Anatolian black pinecone (PC-*Pn*A) as a potential adsorbent for MB removal [19]. PC-*Pn*A showed a pH_{pzc} of 5.77 and a maximum MB adsorption capacity of 91.46 mg/g. More recently, Senra et al. studied pinecone residue (*Pinus taeda)* and found that the 60-mesh fraction, which consisted of smaller particles, is more suitable for removing MB than the 45- and 35-mesh fractions [20]. By optimizing contact time, initial dye concentration, and adsorbent dosage, the study achieved an experimental adsorption capacity of 255.63 mg/g.

5.3. Duckweed-Derived Materials

Duckweed has also been studied as an inexpensive adsorbent to remove MB from wastewater due to its availability, rapid growth, and oxygen-containing functional groups. Jeffrey et al. prepared an adsorbent from duckweed (*Lemna minor*) by washing, drying, grinding, and sieving the biomass without chemical treatment [21]. The material showed an experimental adsorption capacity of 200.80 mg/g at an initial MB concentration of 50 mg/L. Adsorption followed the pseudo-second-order kinetic model and was best described by the Temkin isotherm. Electrostatic interactions, hydrogen bonding, and π–π interactions were proposed as the primary adsorption mechanism of the duckweed-derived adsorbent. Duckweed powder can also be modified to improve its practical use as an MB adsorbent. In the study performed by Seng et al., Fe_3O_4 nanoparticles were co-precipitated onto duckweed powder in the presence of ascorbic acid as a reducing agent [22]. The prepared adsorbent exhibited an adsorption capacity of 78.59 mg/g and could be easily separated from the treated solution due to its magnetic properties. In addition, it retained more than 69% of its initial removal efficiency after five regeneration cycles, demonstrating its potential for repeated use.

5.4. Watermelon-Derived Materials

Watermelon waste has also been investigated as a low-cost raw material for producing adsorbents for MB removal. Shukla et al. converted watermelon rind into biochar by slow pyrolysis at 550 °C without any additional chemical treatment [23]. The resulting watermelon rind biochar (WM-BC) had surface functional groups that could interact with MB and porous structure, providing multiple adsorption sites with a pHpzc of 6.787. The highest removal efficiency was 84% at pH 11, and adsorption reached equilibrium after approximately 100 min. The adsorption data followed the Langmuir isotherm and pseudo-second-order kinetic model, with a maximum adsorption capacity of 115.61 mg/g. Further modification of watermelon waste can improve its adsorption performance and practical handling. Rajendran et al. prepared the magnetic nanocomposites activated carbon (MWMSC) from watermelon shell by incorporating Fe_3O_4 and subsequently activating the material at 650 °C [24]. MWMSC had a BET surface area of 155.35 m^2/g and exhibited an adsorption capacity of 303.30 mg/g for MB. It was demonstrated that MWMSC removed about 99% of MB from both a laboratory sample and a real wastewater sample. The adsorption data were well described by the Langmuir isotherm and pseudo-second-order kinetic model, while thermodynamic analysis indicated that the process was spontaneous and exothermic.

5.5. Spent Coffee Grounds

Spent coffee grounds are another common organic waste that can be converted into adsorbent materials for MB removal. One way to use spent coffee grounds is to convert them into biochar through pyrolysis. In the study reported by Nagarajan et al., spent coffee ground biochar was evaluated using response surface methodology to determine the effects of adsorbent dosage, contact time, solution pH, and temperature [25]. Adsorbent dosage and temperature were identified as the critical factors, and under optimized conditions, the MB removal efficiency reached 99.99%. The adsorption data followed the pseudo-second-order kinetic model and the Freundlich isotherm, suggesting favorable adsorption on a heterogeneous surface.

A different approach was reported by Flores et al., who incorporated spent coffee grounds (SCG) into calcium alginate beads (SCG_ALG) via ionic gelation without pyrolysis [26]. The resulting SCG_ALG adsorbent had a BET surface area of 115 m^2/g and showed a Langmuir maximum adsorption capacity of 1601.85 mg/g. Thermodynamic analysis indicated MB removal by SCG_ALG is a physisorption process. Also, electrostatic and π–π interactions were suggested as the key mechanism in the adsorption of MB. The material also maintained relatively high performance over three adsorption cycles and could be easily separated from the treated water.

5.6. Banana and Other Fruit Peels

Banana peels are another abundant source of fruit waste that can be converted into adsorbents for MB removal. Gong et al. reported MB removal using banana-peel biomass charcoal [27]. For the preparation of their adsorbent, banana peels were ground and activated with KOH and urea, then pyrolyzed at a maximum temperature of 700 °C. The prepared nitrogen-modified biomass charcoal from banana peel (N-BC) was further modified by the addition of Fe_3O_4 to yield the magnetic biochar (Fe_3O_4–N-BC). N-BC showed an adsorption capacity of 807.87 mg/g, while the Fe3O4 load slightly reduced this capacity. However, Fe_3O_4–N-BC exhibited magnetic properties, facilitating easier separation from water. Fe_3O_4–N-BC showed an adsorption capacity of 638.08 mg/g and retained approximately 80% of its initial adsorption capacity even after five adsorption cycles.

A simpler approach was reported by Praipipat et al., who incorporated powders derived from banana, pomelo, and mangosteen wastes into alginate beads and compared their ability to remove MB [28]. Mangosteen powder beads exhibited the highest removal efficiency, 98.92%, and an adsorption capacity of 6.59 mg/g at an initial MB concentration of 100 mg/L. In comparison, banana powder beads achieved 97.59% removal and a removal rate of 6.51 mg/g. It was also demonstrated that three types of powder beads could be reused according to the adsorption–desorption experiments performed.

5.7. Water Hyacinth-Derived Materials

Water hyacinth is an invasive aquatic plant that can also serve as a low-cost biomass source for MB adsorption. Ratchawet et al. studied water hyacinth stem powder (WHSP) prepared without chemical modification, as an adsorbent targeting MB [29]. WHSP showed an experimental MB adsorption capacity of 126.7 mg/g and a removal efficiency of 92.1%. The experiments showed rapid adsorption during the first 60 minutes, with equilibrium reached after approximately 180 min. The characterization of WHSP suggested the presence of hydroxyl and carboxyl groups, which contributed to MB adsorption. The thermodynamic study also supported the conclusion that MB adsorption is endothermic and spontaneous.

Shen et al. chemically modified water hyacinth with maleic anhydride to obtain the maleic anhydride-modified water hyacinth powder (MA-EC) [30]. The chemical modification introduced additional carboxyl groups onto the water hyacinth powder, increasing the adsorbent's negative surface charge. MB adsorption rapidly occurred during the first 20 min and reached equilibrium at around 30 min. MA-EC also demonstrated its reusability for MB removal, with an adsorption capacity of 215.56 mg/g after four adsorption–desorption cycles.

5.8. Peanut-Derived Materials

Peanut shells and husks can also be converted into adsorbents for MB removal via thermal or chemical modification. Hashem et al. synthesized carbonaceous biomass from peanut shells using either NaOH, $ZnCl_2$, or steam activation [31]. Among the prepared materials, NaOH-activated carbon produced at a 1:3

(carbon: NaOH) ratio exhibited the highest adsorption performance, with a maximum adsorption capacity of 303.03 mg/g at 28 °C. The material also reached equilibrium within approximately 10–15 min. The proposed mechanism for MB removal with NaOH-activated carbon includes π–π interactions, hydrogen bonding, and electrostatic interactions.

Aryee et al. developed a peanut-husk adsorbent modified with potassium permanganate (PNK) under relatively mild conditions [32]. The characterization of PNK indicated that the adsorbent contains functional groups that support its efficiency to remove MB, with a pH_{pzc} of 6.11. PNK had an MB adsorption capacity of up to approximately 270 mg/g in batch experiments. In a continuous-flow column, an adsorption capacity of 276.0 mg/g and a removal efficiency of 82.5% were reported. PNK also showed useful adsorption performance over five regeneration cycles.

Table 2. Summary of plant-derived absorbents for methylene blue removal and reported experimental conditions
**NR means not reported*

Ref.	Adsorbent	Preparation method	Maximum adsorption capacity (mg/g)	Removal efficiency (%)	Initial MB concentration (mg/L)	pH	Temperature	Contact time
[14]	Rice straw biosorbent (RSB)	Washed, dried, ground rice straw; used without major chemical modification	158	NR	20–100	2–12	303 K*	0–240 min
[15]	Magnetic alginate/rice husk beads	Rice husk incorporated into alginate; $CaCl_2$/Fe salts used to form magnetic composite beads	274.9	NR	NR	6–10 tested	NR	NR

[16]	Modified rice straw powder	Rice straw treated with 0.1 M NaOH and used in a fixed-bed column	21.99	NR	NR	NR	NR	Continuous-flow
[17]	Torrefied rice husk (TRH)	Rice husk thermally torrefied; tested in batch and inverse fluidized-bed systems	38.0†	84‡	NR	NR	NR	95 min‡
[18]	Algerian pine tree bark	Pine bark prepared as a porous biosorbent and used in batch adsorption	37.15	99.47	60–140 tested	8 optimum	55 °C optimum	2–240 min tested
[19]	Pinus nigra pinecone powder	Pinecones dried, ground and sieved into powder	91.46	NR	NR	pHpzc = 5.77; experimental pH varied	Varied	Varied
[20]	Pinus taeda pinecone residue	Pinecone residue ground and separated by particle size/mesh	255.63§	NR	>37.5 optimum region	NR	NR	>45 min optimum region

[21]	Duckweed powder	Duckweed washed, dried, ground and sieved; no chemical treatment	200.80	NR	50 for maximum capacity	10 optimum	NR	NR
[22]	Magnetic duckweed (MDW)	Fe_3O_4 nanoparticles co-precipitated onto dried duckweed using ascorbic acid	78.59	85.27	50	10	298–318 K studied	60 min
[23]	Watermelon rind biochar (WM-BC)	Slow pyrolysis of watermelon rind at 550 °C without additional chemical treatment	115.61	84¶	NR	11¶	NR	~100 min¶
[24]	Magnetic watermelon-shell activated carbon (MWMSC)	Watermelon-shell carbon activated and modified with Fe_3O_4; activation at ~650 °C	303.30	~99 in real/lab samples	NR	6 optimum	NR	60 min optimum
[25]	Spent coffee-ground	Spent coffee grounds converte	NR	99.99	NR	7 optimum	30 °C optimum	30 min optimum

	biochar (SCGB)	d to biochar by pyrolysis						
[26]	Spent coffee ground–alginate beads (SCG_ALG)	Spent coffee grounds incorporated into calcium alginate beads by ionic gelation	1601.85	NR	50 in thermodynamic tests	6 in thermodynamic tests	25–60 °C studied	60 min
[27]	Banana-peel N-biochar / Fe_3O_4–N-BC	KOH/urea activation and pyrolysis up to 700 °C; Fe_3O_4 subsequently incorporated	807.87 N-BC; ~640 Fe_3O_4–N-BC	NR	300 in adsorbent comparison; 50–400 studied	Natural pH for comparison; 3–12 studied	298 K for comparison	NR
[28]	Banana, pomelo and mangosteen peel alginate beads	Fruit-waste powders incorporated into alginate beads	6.59 (MPB)	98.92 (MPB)	100	NR	NR	NR
[29]	Water hyacinth stem powder (WHSP)	Stems dried, ground and used without chemical modification	126.7	92.1	NR	NR	NR	~180 min equilibrium
[30]	Maleic-anhydride-modified water hyacinth	Water hyacinth stems chemically modified	434.70	NR	1000 in pH experiment	8 used in reported high-performance condition	25 °C	~30 min equilibrium

	(MA-EC)	with maleic anhydride						
[31]	Peanut-shell activated carbon	Peanut shells carbonized and activated with NaOH, $ZnCl_2$ or steam; best NaOH ratio 1:3	303.03	>95	NR	10	NR	50 min
[32]	$KMnO_4$-modified peanut husk (PNK)	Peanut husk chemically modified using potassium permanganate	~270 batch; 276.0 column	82.5 column	NR	NR	Ambient conditions	NR

6. COMPARISON OF PLANT-DERIVED ADSORBENTS

Section 5 demonstrates the potential of a wide range of plant-derived wastes and biomass materials as MB removal adsorbents. However, their reported adsorption capacities vary considerably. Some relatively simple materials, such as untreated rice straw, pinecone powder, duckweed, and water hyacinth stem powder, showed notable adsorption capacities without extensive modification. Several materials underwent modification, such as pyrolysis, chemical activation, magnetic modification, or incorporation into composite materials, also showed high adsorption capacities. However, the extra processing steps should also be considered. Modification can also provide benefits beyond adsorption capacity. The incorporation of Fe_3O_4 into duckweed, watermelon-derived carbon, and banana peel biochar enables to easily recover the used adsorbents with a magnet. Similarly, incorporating plant-derived materials such as rice husk, spent coffee grounds, and fruit peels into alginate beads made the adsorbent to easily handle and separate from the treated water. Other approaches including chemical modification altered the surface properties of plant-based adsorbents such as water hyacinth and peanut husk, contributing to reusability or performance in continuous-flow systems. These factors are important because even an adsorbent with a high adsorption capacity may not be practical if it is difficult to recover or reuse. Regeneration studies

showed that several reviewed materials could be used for multiple adsorption cycles, although adsorption performance generally decreased with repeated use. The adsorption capacities reported in the literature should be carefully evaluated because different experimental conditions, such as MB concentration, adsorbent dosage, pH, temperature, and contact time, have been used. The reported maximum capacities were also obtained using different adsorption models and experimental approaches. Consequently, a higher absorption capacity does not necessarily mean that an absorbent is a better candidate for removing MB practically. Removal efficiency should also be interpreted separately from adsorption capacity. For example, some materials achieved removal efficiencies above 95% despite relatively low adsorption capacities due to the experimental conditions used. Overall, these studies show that adsorption capacity alone is not enough to determine whether a plant-derived adsorbent is suitable for practical applications. Preparation requirements, material recovery, reusability, stability, and experimental conditions should also be considered when comparing different adsorbents.

7. CONCLUSION AND FUTURE PERSPECTIVES

Plant-derived adsorbents have shown their potential to remove MB from wastewater, but several challenges still need to be addressed in the future.

1. Experimental conditions (i.e., differences in MB concentration, adsorbent dosage, pH, temperature, and contact time) often vary from one study to another and this makes it difficult to compare adsorption outcome directly
2. Many studies were performed under the controlled laboratory conditions. Actual wastewater may contain various salts, other dyes, organic compounds, and other contaminants, which could interfere with adsorption. Thus, studies using complex water samples or actual wastewater would help show how plant-derived adsorbents perform under practical conditions.
3. Since a majority of research uses batch experiments, testing with fixed-bed columns and other continuous-flow systems would be useful for evaluating larger-scale applications.

The synthetic procedure and applicable further modification of plant-derived adsorbents is another important factor. The preparation methods, such as pyrolysis, chemical and magnetic modification, and the formation of composites or beads can improve adsorption performance and they also make the adsorbents to recover or reuse easily. However, these methods may also require more energy, chemicals, and processing. Thus, the benefits of modification should be evaluated while considering the additional resources needed to prepare the adsorbent. Recovery, regeneration, and long-term stability also need further attention. Several adsorbents in the literature could be reused for multiple adsorption cycles, but their performance often decreased over time.

Future work needs to demonstrate the following:

1. How do these adsorbent materials perform over longer periods?
2. How effectively can they be regenerated?
3. How can MB-loaded adsorbents be safely handled or disposed of?

Overall, plant-derived materials and agricultural wastes have been considered as potential alternatives for MB removal from wastewater. This is because they are inexpensive, widely available, and able to adsorb MB effectively. Some can be used with little processing, while others perform better after chemical or physical modification. However, adsorption capacity is not a sole factor to determine whether an adsorbent is practical or not. Preparation, recovery, reusability, stability, cost, environmental impact, and performance in actual wastewater are also important when considering their future use.

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