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Two-Dimensional Nanomaterials for Organic Dye Removal from Industrial Wastewater: Performance, Mechanisms, and Prospects

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Abstract. Industrial wastewater poses a serious environmental challenge, as the discharge of large amounts of organic dyes threatens aquatic ecosystems and human health. Accordingly, certain two-dimensional (2D) nanomaterials, such as graphene, MXenes, and molybdenum disulfide, have demonstrated considerable potential for the efficient removal of organic dyes. Their high specific surface area, tunable interlayer spacing, and abundant surface functional groups offer significant advantages in dye adsorption and membrane separation applications. This review examines the adsorption efficiency, removal mechanisms, membrane separation performance, and recyclability of selected 2D nanomaterials for the treatment of industrial dye wastewater. Notably, particular emphasis is placed on the influence of material structural characteristics on dye removal. Furthermore, the effects of different environmental conditions on removal mechanisms, membrane fouling behavior, and long-term operational stability are also discussed. Additionally, environmental accumulation, ecological toxicity, eco-friendly synthesis, and sustainable development strategies are discussed. The paper highlights current research limitations and future directions, providing a theoretical foundation for the efficient and environmentally friendly application of 2D nanomaterials in water treatment.

Keywords: Two-dimensional nanomaterials, dye wastewater, adsorption removal, membrane separation, environmental applications

1. Introduction

Organic dye wastewater discharged from textile printing and dyeing industries often features stable structures, resistance to degradation, potential carcinogenicity, and long-term accumulation in water bodies. Traditional methods such as coagulation, adsorption, and biodegradation suffer from limited removal efficiency, insufficient selectivity, and risks of secondary pollution [1]. Two-dimensional (2D) nanomaterials, with their large specific surface area, tunable interlayer structures, and abundant active sites, have been widely applied in dye adsorption and membrane separation. For instance, a 2D Zn(II) coordination polymer (PCP-10) reported by Özcan et al. generated superoxide radicals under ultraviolet light induction, achieving a degradation rate of 91% along with excellent stability

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and recyclability [2]. However, practical applications face challenges, including inconsistent evaluation standards, limited validation in real wastewater, insufficient data on long-term stability and ecological impacts, and the need for greener synthesis techniques. Thus, this review summarizes recent advances in 2D nanomaterials for organic dye removal from industrial wastewater, with a focus on removal efficiency and underlying mechanisms. Notably, the relationships between structural characteristics and adsorption performance are investigated to identify the dominant mechanisms governing dye adsorption. Besides, recent advances in membrane separation are evaluated, particularly the effects of pore-size regulation on pollutant transport and strategies for membrane fouling mitigation and regeneration. Furthermore, potential ecological risks associated with 2D nanomaterials are assessed, together with strategies for green design and sustainable development. These findings highlight key challenges and future prospects for their application in industrial wastewater treatment.

2. Performance and mechanism of two-dimensional materials in organic dye adsorption

2.1. Main materials and adsorption efficiency

The structural characteristics and surface properties of different two-dimensional materials determine their ability to remove organic dyes, with specific surface area, interlayer structure, surface functional groups, and types of active sites being key factors influencing adsorption performance. Specific surface area determines the number of adsorption sites, while interlayer structure affects dye transport and site accessibility. The fabrication method improves removal efficiency by tuning pore structure, interlayer spacing, and surface chemistry.

GO, MXene, and MoS_2, as representative two-dimensional materials, exhibit distinct functional differences in removal efficiency. GO, with its abundant oxygen-containing functional groups and tunable interlayer structure, has a solid foundation for applications in dye adsorption and membrane separation, although its aqueous stability still needs improvement. Pressure-assisted regulation of interlayer arrangement can enhance dye molecule transport and adsorption efficiency [3]. In contrast, MXene features adjustable interlayer spacing and rich surface active sites, enabling effective removal of various dyes and demonstrating excellent water permeability. Its adsorption capacity can be further enhanced via composite modification [4]. Due to the lack of hydrophilic groups and strong structural rigidity, MoS_2 relies more on size-based sieving than electrostatic adsorption during dye removal. Besides, its unique semiconductor band structure enables efficient photocatalytic degradation, unlike the physical retention mechanisms of GO and MXene. Studies have shown that an ultrasound-assisted MoS_2/CeO_2 heterostructure (4:1) achieves 94% methylene blue degradation after 90 min of UV irradiation, demonstrating excellent photocatalytic activity [5].

2.2. Mechanism analysis and mode of action

The adsorption of organic dyes by two-dimensional nanomaterials is influenced collectively by the material's structural characteristics, surface chemistry, and solution environment, primarily achieved through interfacial interactions such as π–π stacking, electrostatic forces, hydrogen bonding, and van der Waals forces. Among these, π–π stacking plays a crucial role in the adsorption of aromatic dyes onto two-dimensional carbon-based materials. The sp^2-hybridized carbon structure on the surface of graphene oxide (GO) can form π–π interactions with aromatic rings in dye molecules, promoting their enrichment on the material surface and resulting in strong adsorption capacity for dyes containing multiple benzene rings [6]. Yang et al. demonstrated that GO exhibits a maximum

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adsorption capacity for Rhodamine B (RhB) as high as 107.230 mg/g. Meanwhile, solution pH is a key factor regulating electrostatic interactions. Since the GO surface carries a negative charge, optimal adsorption of RhB occurs at around pH = 7. When combined with polyacrylamide, electrostatic repulsion is reduced, yet the dye retention rate increases to 95.43%, surpassing the 85.03% achieved by ultrafiltration [3, 6]. In addition, hydrogen bonding and van der Waals forces further enhance the binding stability between dye molecules and the material surface, particularly in materials rich in polar groups or porous structures. For instance, the adsorption of RhB onto BPM-COF mainly results from the combined effects of hydrogen bonding, electrostatic interactions, and van der Waals forces [7]. Thus, during the design of two-dimensional materials, surface properties should match dye structure and solution conditions to enhance synergy among multiple mechanisms.

2.3. Recycling and performance optimization

For practical applications, adsorbent recyclability is an important consideration. Studies have shown that BPM-COFs synthesized in one step from BTCH and PMDA exhibit high adsorption capacity toward RhB, achieving an 80.22% removal efficiency at a dye concentration of 192.54 $mg \cdot g^{-1}$. After three desorption-adsorption cycles, the removal efficiency remained at 73.01%, indicating that the material maintains stable adsorption performance during repeated use. This excellent recyclability may be attributed to the ordered pore structure and strong covalent bonding of COFs, which prevent significant loss of active adsorption sites during desorption [7].

Beyond adsorption, conventional MoS_2 suffers from limited performance in PMS activation due to low radical generation efficiency and insufficient active sites, necessitating structural modification to enhance dye degradation capability [8]. Fan et al. prepared few-layer MoS_2 nanosheets ($SMoS_2$) with Li assistance and further introduced Co to construct a Co-$SMoS_2$ catalyst, increasing the CBZ degradation rate by 17-fold while maintaining good stability across pH 3–11 [8]. This modification enhances the catalyst's ability to activate PMS by increasing the exposure of active edge sites and facilitating electron transfer. Additionally, the few-layer structure shortens the mass transfer distance between pollutants and active sites, thereby improving the overall degradation efficiency.

3. Application of two-dimensional material membranes in industrial dye wastewater treatment

3.1. Pore size regulation and dye retention

Two-dimensional material membranes are typically composed of stacked nanosheets, with interlayer channels forming effective transport pores whose dimensions directly influence the sieving of dye molecules. Accordingly, membrane separation primarily relies on steric hindrance and electrostatic repulsion to retain dyes; thus, interlayer spacing and pore structure are key factors affecting separation performance. As pore size decreases, dye molecules find it harder to pass through, leading to higher retention rates but increased mass transfer resistance and reduced flux. Conversely, larger pores enhance mass transfer but may compromise separation selectivity.

Based on this relationship, researchers have optimized membrane performance by tuning interlayer structures and constructing composite separation layers. Jiang et al. achieved a RhB rejection rate of 98.7% by in situ fabricating a COF intermediate layer via the synergistic effects of Donnan exclusion and steric hindrance [9]. Meanwhile, Tian et al. added PDA-modified MoS_2 to PES membranes at 0–3.0 wt% loading. This reduced the molecular weight cutoff (MWCO) from 7402 to 5589 Da, thereby improving membrane selectivity, although at the expense of flux [10].

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Therefore, the design focus for two-dimensional material membranes lies in achieving a balance among pore size, retention capacity, and mass transfer efficiency, rather than merely maximizing retention.

3.2. Surface modification and flux variation

By modifying the membrane surface, dye separation performance can be regulated through changes in hydrophilicity, surface charge, pore structure, and water transport resistance. Tian et al. modified MoS_2 with PDA, utilizing the hydrophilic groups in PDA to enhance membrane surface wettability, reducing the water contact angle of MoS_2 from 79.6° to 55.5°. However, as the PDA concentration increased to 3.0 wt%, the membrane structure gradually became denser, resulting in a decrease in pure water flux to 42.0 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$. This indicates that moderate surface modification can improve membrane wettability and promote dye separation, whereas excessive modification may compress effective transport channels, increase mass transfer resistance, and lead to reduced flux [10].

In addition to mass transfer, surface-modified functional groups can alter interfacial interactions with dye molecules. Under strong electrostatic interactions, dye adsorption and retention are boosted; however, excessive interfacial interactions may cause dye accumulation on the membrane surface, leading to pore blockage and increased membrane fouling. Therefore, modification strategies must balance enhanced interfacial interactions with the preservation of mass transfer pathways to optimize membrane separation performance.

3.3. Membrane surface fouling and cleaning

Two-dimensional membranes in liquid separation processes are primarily affected by concentration polarization, interactions between contaminants and the membrane surface, and pore blockage. In particular, solute enrichment at the membrane surface forms a concentration polarization layer; at the same time, adsorption between dye molecules and membrane materials leads to pore fouling; while retained particles or macromolecules depositing on the surface form a cake layer, ultimately resulting in increased transmembrane pressure and flux decline [11, 12].

Accordingly, anti-fouling designs mainly aim to reduce contaminant adhesion at the membrane interface. For instance, the introduction of hydrophilic nanosheets or photocatalytic materials onto the membrane surface can create a hydration layer that reduces contaminant adsorption or promote photocatalytic degradation of organic pollutants, thereby minimizing irreversible fouling [13]. For already formed fouling layers, membrane performance must be restored through cleaning. Typically, cake layers formed by surface deposition can typically be removed using physical methods such as backwashing or ultrasound; however, contaminants embedded within pores require chemical cleaning, which may damage the membrane structure and compromise long-term stability [12].

4. Environmental impact and sustainable improvement of two-dimensional materials

4.1. Biological exposure and toxicity accumulation

In aquatic environments, the size, layer number, surface functional groups, and structural stability of 2D materials influence their transport, transformation, and biological exposure. These materials can undergo environmental transformations through processes like oxidation, structural reorganization, or particle release, and may subsequently be taken up by organisms. Lin et al. reported that

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nanoscale GO can be gradually degraded by splenic macrophages, although this process lasts approximately nine months, indicating a prolonged retention time in tissues. In contrast, larger-sized GO particles are cleared more slowly, potentially increasing the risk of inflammatory responses. Similarly, MoS_2 can transform into soluble molybdate salts in vivo, yet its degradation process may still induce inflammation [14]. The size and structural stability of two-dimensional materials directly affect their in vivo transformation behavior and toxicological effects.

In addition to intrinsic material properties, organic components in environmental media can alter the interfacial behavior of two-dimensional materials, thus influencing their mobility and biological exposure. Surface adsorption of biomolecules like proteins on these materials forms protein coronas, which in turn modify surface charge, dispersion state, and aggregation behavior. Cheng et al. found that BSA in wastewater can adsorb onto MXene nanosheets to form protein coronas, affecting their aggregation, sedimentation, and environmental fate [15]. Similar interactions may also occur in dye-containing wastewater systems, where microbial metabolites and natural organic matter alter the interface properties of materials, further impacting the environmental stability and ecological risks of two-dimensional materials.

4.2. Green synthesis and recycling utilization

The preparation of two-dimensional materials often involves the use of organic solvents and high-temperature treatments, so green synthesis primarily focuses on reducing solvent consumption and energy consumption. Ren et al. summarized methods for preparing ultrathin 2D materials using supercritical carbon dioxide (SCF), leveraging its high diffusivity and tunable solvating power to promote nanosheet exfoliation, thereby reducing the use of organic solvents in conventional liquid-phase exfoliation processes. Moreover, SCF combined with strategies such as ultrasound assistance, reverse micelle induction, and Pickering emulsion induction has been applied to the fabrication of materials such as graphene and MoS_2, enhancing exfoliation efficiency [16]. Compared to traditional high-temperature processing methods, flash Joule heating (FJH) shortens reaction time through rapid thermal cycling, significantly reducing energy consumption during material synthesis [17].

In addition to the preparation process, recycling and reuse of 2D materials in practical applications are also critical factors affecting their sustainability. Due to the small size and strong dispersion of nanosheets, direct separation and recovery pose significant challenges. Therefore, improving recyclability is typically achieved through morphology control or composite construction. Luong et al. fabricated millimeter-scale particles by self-assembly and cross-linking of GO with chitosan, which not only maintained a methylene blue adsorption capacity of 951.35 $mg \cdot g^{-1}$ but also enhanced material separability and recoverability while preserving cyclic adsorption performance [18]. Yang et al. incorporated fragmented GO or MXene structures into MXene composites, achieving a material recovery rate of approximately 95% while retaining good methylene blue adsorption capacity [19]. These studies demonstrate that macroscopic processing or composite structural design can improve the recyclability of 2D materials without compromising their adsorption performance.

5. Conclusion

This review examines the applications and optimization strategies of two-dimensional nanomaterials such as GO, MXene, and MoS_2 in treating industrial organic dye wastewater. Studies show that these 2D materials achieve efficient dye removal through high specific surface area, tunable interlayer structures, and active surface sites, with π–π interactions, electrostatic forces, and

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hydrogen bonding being the primary adsorption mechanisms. In membrane separation processes, the regulation of interlayer spacing and surface properties can improve dye retention; however, membrane fouling and flux decline still limit their long-term practical use. Furthermore, once released into the environment, the size, structural stability, and interfacial behavior of 2D materials influence their migration, transformation, and biological effects, necessitating further assessment of potential ecological risks. Regarding green synthesis and recycling, strategies such as supercritical CO_2-assisted exfoliation, flash Joule heating, and morphology control can reduce energy consumption during fabrication and improve recovery efficiency. Currently, the application of 2D materials faces challenges including insufficient validation in real wastewater, lack of standardized evaluation criteria, and limited data on long-term environmental impacts. Future research should integrate actual water conditions to better understand the relationship between material structural changes and environmental behaviors.

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