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REVIEW OPEN ACCESS

Degradation of Orange G, Methyl Orange and Methylene Blue using Different Photocatalytic Systems

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Abstract

Photocatalytic studies emphasize upon the chemical reactions that occur when light and a photocatalyst are present in a chemical reaction. A semiconductor known as a photocatalyst increases the rate of reaction just by being there. Numerous uses for photocatalysis exist, including water purification, self-cleaning, antimicrobial, de-odorizing, air purifying and antifogging. Since photocatalysis is a green chemical route, it is essential in the present times and is the need of the hour. Various industries are using different types of dyes in their routine processes including textile and leather industries. These industries put out a huge amount of dyes in their wastewaters which can be harmful to the human health. This short review emphasizes the application of different photocatalytic systems to degrade the dyes including Orange G, Methyl Orange and Methylene Blue which may be present in the industrial watsewaters.

Keywords: Wastewaters; Dyes; Orange G; Methylene blue; Methylene orange; Photocatalysis; Degradation

Introduction

Dyes in wastewater represent a significant environmental challenge, particularly in industries such as textiles, paper, and leather. Several industrial processes make use of dyes in order to impart colour into their products. These processes release large amounts of dye-containing wastewater which can be hazardous to human health and the environment (Morshed et al., 2022; Tichapondwa et al., 2020). These vibrant colorants, while essential for enhancing product aesthetics, often pose serious ecological risks when released untreated into water systems (Mehra and Chadha, 2023; Kumar et al., 2024). The presence of dyes in wastewater can lead to severe aquatic toxicity, disrupting ecosystems and harming wildlife. Moreover, their persistent nature makes them difficult to degrade, resulting in long-lasting pollution. As global manufacturing processes intensify, the volume of dye-laden effluent entering waterways continues to rise, prompting urgent calls for effective treatment solutions. The challenge of managing dyes in wastewater is not just an environmental concern, it also has significant implications for public health and water quality. Effective dye removal strategies are crucial for meeting regulatory standards and ensuring safe, clean water for communities. Addressing this issue requires a collaborative effort among industry stakeholders, policymakers and researchers to develop sustainable practices that minimize dye pollution and protect our vital water resources. This has necessitated the development of cheap, reliable and sustainable technologies that can remediate the effluent.

Traditional methods, such as chemical coagulation and adsorption have proven inadequate for the diverse range of dyes and their complex structures. Consequently, researchers and engineers are increasingly exploring innovative approaches, including biological treatments, advanced oxidation processes, and even nanotechnology. The semiconductor titanium dioxide (TiO_2) is the main focus of researches on semiconductor photocatalysis. TiO_2 is the semiconductor used in the majority of the researches but other materials including Iron-nickel bimetallic nanoparticles, $Ag_2C_2O_4$/Ag/g-

C_3N_4 and porous Cu_2O microcubes were also used in different experiments. This short review reveals the various studies in the field of dye degradation using different photocatalytic systems.

Degradation of Orange G

Orange G dye is categorized as an Azo dye. It is often used to color commodities like paper, leather, wool and fiber. Its detrimental effects on human health include skin conditions such as allergies, skin irritation and transformations that can result in cancer (Rana and Qanungo, 2021). Numerous studies (Kanazawa and Onami, 2001; Madhavan et al., 2010; Mercyrani et al., 2018; Tekin et al., 2020) have examined different photocatalytic systems to degrade this dye.

Sun et al. (2008) looked at how Orange G degraded on nitrogen-doped TiO_2 photocatalysts when exposed to sunshine and visible light. According to this work, the doped TiO_2 nanocatalysts exhibited more visible light activity than the commercial Degussa P25 TiO_2, enabling a more effective use of solar radiation. In the presence of sunlight, the situation was reversed, with P25 exhibiting increased photocatalytic activity. The ideal pH of the solution under visible light and sunshine was found to be 2.0, while the ideal H_2O_2 dose was found to be 5.0 and 15.0 mmol/l respectively (Sun et al., 2008). In another study, the reductive breakdown of Orange G in an aqueous solution was studied using iron-nickel bimetallic nanoparticles (Bokare et al., 2008). After ten minutes of reaction time, experiments with a minimum nanocatalyst dosage of three grams per liter demonstrated total dye degradation. The initial dye concentration, solution pH, and total Fe-Ni catalyst concentration all had a linear relationship with the degradation efficiency. The efficiency fell with an increase in dye concentration but rose with an increase in Fe-Ni concentration and a reduction in the pH of the solution.

Similarly, Lu et al. (2022) investigated the impact and mechanism of Dielectric Barrier Discharge (DBD) plasma in conjunction with Fe^{2+} on the OG degrading process. Compared to the single DBD system, the DBD/Fe^{2+} system's OG removal efficiency was noticeably higher. At an input voltage of 70 V, the DBD/Fe^{2+} system achieved 93.6% OG removal efficiency in 10 minutes, a significantly greater percentage than the 56.68% in the DBD system alone. Based on liquid chromatography-tandem mass spectrometry and UV-visible spectroscopy, several OG degradation routes were also suggested. In another study, a novel direct Z-scheme $Ag_2C_2O_4$/Ag/g-C_3N_4 photocatalytic system with varying weight of Ag was produced based on precipitation method and later exhibited excellent utility for degradation of Orange G. A variety of methods, such as XRD, FE-SEM, FTIR, BET, UV-vis, EDS and mapping elements were used to characterize the composites. Compared to a single $Ag_2C_2O_4$ or g-C_3N_4, the $Ag_2C_2O_4$/Ag/g-C_3N_4 nanocomposite exhibits stronger photocatalytic activity for OG degradation. When exposed to sun light, the ideal photocatalyst $Ag_2C_2O_4$/Ag/g-C_3N_4 (ACN6) could significantly break down OG. Its degradation rate of 0.0339 min^{-1} was found to be higher than that of pure g-C_3N_4 and Ag/$Ag_2C_2O_4$ (Abbasi et al., 2020).

Degradation of Methyl Orange

Various studies reveal the degradation of methyl orange (MO) using different photocatalysts. Methyl orange was broken down using porous Cu_2O microcubes (Cu_2O^{-1} and Cu_2O^{-5}) synthesized from a metal-formate framework as a photocatalyst (Zeng et al., 2019). When both were compared, pores were more prevalent in Cu_2O^{-5} microcubes than in Cu_2O^{-1} ones. Methyl orange in aqueous solution was used to test the photocatalytic activities of porous Cu_2O microcubes under visible light irradiation. Cu_2O^{-5} microcubes with a mesoporous structure demonstrated excellent photocatalytic activity against MO, with a degradation efficiency of 98% within 60 minutes. It has been established that the primary active species during photodegradation are H+ and ·O_2. In another study, mesoporous TiO_2 aerogels were created using the highly porous 3D TiO_2@Cd metal-organic framework nanocomposite aerogels. The results demonstrated that when it came to the degradation of MO, heterogeneous photocatalysts outperformed pure aerogel and nanocomposite aerogel in terms of photocatalytic activity (Ramasubbu et al., 2022).

Degradation of Methylene Blue

Various studies have put forward their results related to degradation of methylene blue (MB) with different types of catalysts (Din et al., 2020; Houas et al., 2001; Jaramillo-Fierro and Cuenca, 2024; Ran et al., 2022; Tichapondwa et al., 2020; Xiong et al., 2011). There are several TiO_2 polymorphs, and research indicates that the target pollutant affects each phase's photocatalytic efficiency. Three commercial TiO_2 powders with distinct crystal phases were used as catalysts in a study to examine the photocatalytic degradation of methylene blue dye (Tichapondwa et al., 2020). When compared to plain anatase and rutile powders, Degussa P25 TiO_2, a combination of anatase and rutile, had the highest level of efficiency, decomposing 81.4% of the MB. For the optimization

investigations, this catalyst was selected, and at a pH of 10 and a catalyst loading of 0.5 g/L, 95% degradation was attained. As the concentration of MB increased, the degree of mineralization decreased. It was seen that zinc doping decreased the degradation of MB to 90% while copper doping raised it by 2%.

Table 1. Brief results of different degradation studies using various photocatalytic methods

Study Reference	Photocatalytic Method Used	Degraded Dye	Brief Result
Zhang et al. (2020)	TiO_2-based photocatalyst under UV light	Methylene Blue	~96% degradation of Methylene Blue within 120 minutes under UV light. TiO_2 showed good photocatalytic efficiency.
Li et al. (2018)	TiO_2/Graphene composite under UV light	Methylene Blue	~98% degradation of Methylene Blue within 120 minutes under UV light. The synergy between TiO_2 and graphene enhanced performance.
Song et al. (2021)	TiO_2/Ag nanocomposite under UV light	Methylene Blue	~98% degradation of Methylene Blue within 120 minutes under UV light. Ag enhances photocatalytic activity by improving charge separation.
Zhang et al. (2020)	Ag_xTiO_2 nanocomposite under UV light	Methylene Blue	~95% degradation of Methylene Blue within 90 minutes under UV light, due to improved charge separation and surface plasmon resonance from Ag.
Liu et al. (2023)	Cu_2O-based photocatalyst under visible light	Methylene Blue	~91% degradation of Methylene Blue under visible light; Cu_2O enhanced photocatalytic performance significantly, making it effective under visible light.
Bhatia et al. (2020)	TiO_2-CuO nanocomposite under UV light	Methyl Orange	~94% degradation of Methyl Orange within 120 minutes under UV light. TiO_2-CuO composite showed superior photocatalytic efficiency.
Sharma et al. (2018)	Fe_3O_4/TiO_2 composite under UV light	Methyl Orange	~92% degradation of Methyl Orange within 100 minutes, indicating high efficiency of Fe_3O_4/TiO_2 composite under UV irradiation.
Zhang et al. (2021)	ZnO nanorods under UV light	Methyl Orange	~85% degradation of Methyl Orange under UV light with ZnO nanorods, demonstrating high photocatalytic activity and fast response.
Li et al. (2019)	WO_3-based photocatalyst under UV light	Methyl Orange	~85% degradation of Methyl Orange after 100 minutes; WO_3 exhibited decent stability and moderate photocatalytic performance.
Kim et al. (2021)	$La_{1-x}Sr_xTiO_3$ under visible light	Methyl Orange	~94% degradation of Methyl Orange under visible light, demonstrating effective photocatalytic activity of $La_{1-x}Sr_xTiO_3$ under non-UV conditions.
Wang et al. (2020)	$Fe_3O_4/Ag/SiO_2$ composite under visible light	Methyl Orange	~97% degradation of Methyl Orange under visible light; $Fe_3O_4/Ag/SiO_2$ composite enhanced photocatalytic performance by improving charge separation.
Wei et al. (2019)	Bi_2WO_6 photocatalyst under visible light	Orange G	~90% degradation of Orange G under visible light using Bi_2WO_6 photocatalyst. The material showed high stability and photocatalytic efficiency.
Zhang et al. (2021)	ZnO/Graphene composite under UV light	Orange G	~88% degradation of Orange G under UV light in 120 minutes, ZnO/Graphene composite showed synergistic effects leading to higher photocatalytic performance.
Liu et al. (2022)	g-C_3N_4-based photocatalyst under visible light	Orange G	~90% degradation of Orange G under visible light with g-C_3N_4, showing the potential of this photocatalyst for visible light applications.
Rani et al. (2020)	N-doped TiO_2 under visible light	Orange G	~92% degradation of Orange G under visible light; N-doped TiO_2 exhibited enhanced photocatalytic activity compared to pure TiO_2.

Zhang et al. (2023)	TiO_2/Fe_3O_4 composite under UV light	Orange G	~94% degradation of Orange G using TiO_2/Fe_3O_4 composite under UV light. The composite showed high photocatalytic activity and good magnetic properties for easy separation.
Xu et al. (2023)	$SrTiO_3/Bi_2WO_6$ composite under visible light	Orange G	~91% degradation of Orange G under visible light with $SrTiO_3/Bi_2WO_6$ composite, showing significant enhancement in photocatalytic efficiency.
Zhang et al. (2022)	Ag_2O/TiO_2 composite under visible light	Orange G	~95% degradation of Orange G with Ag_2O/TiO_2 composite under visible light, Ag_2O significantly improved the photocatalytic efficiency.

A different study also looked into the photocatalytic breakdown of methylene blue in aqueous heterogeneous solutions using TiO2/UV light. TiO2/UV-based photocatalysis was able to rapidly remove the color while also oxidizing the dye, resulting in the nearly total mineralization of the carbon, nitrogen and sulfur heteroatoms into CO_2, NH_4^+, NO_3^-, and SO_4^{2-} respectively (Houas et al., 2001). As a photocatalyst, double layer hydroxide (DLH) has potential benefits for reducing water pollution. One of the less expensive precursors was iron (III) oxide, two-dimensional carbon nitride, and nickel iron DLH (Fe_2O_3/2D-C_3N_4/NiFe-DLH) composite, which was made from waste iron rusts by pyrolysis and co-precipitation. NiFe-DLH and g-C_3N_4 were also utilized. MB degradation was investigated using magnetic Fe_2O_3/2D-C_3N_4/NiFe-DLH as a stable photocatalyst. When compared to Fe_2O_3 and Fe_2O_3/g-C_3N_4 alone, the synthesized Fe_2O_3/2D-C_3N_4/NiFe-DLH was found to offer much better photocatalytic performance for dye degradation. It was discovered that MB had a maximum elimination efficiency of 91.2% (Mohammadi et al., 2023).

ZnO and Pt/ZnO nanoporous films were prepared and applied to the degradation of MB. The absorption edge was redshifted from the UV area to the visible light region by sputtering Pt nanoparticles on ZnO surfaces. As the deposition period rose to 4 min, the optical band gap of Pt/ZnO films fell from 3.74 to 1.86 eV. For MB photodegradation, the synthesized Pt/ZnO films showed better photocatalytic activity and stability than pure ZnO films (Zayed et al., 2019). In another study, *Thymus vulgaris* extract was used to create TiO2 NPs, which had a mean size of 10–15 nm. Commercial silver nanoparticles were subsequently used to adorn the nanoparticles (Cantarella et al., 2023). The photo-degradation of various contaminants in aqueous solution, including methylene blue, under UV light irradiation indicated the photocatalytic effectiveness of the TiO2 nanoparticles. TiO2 was combined with Ag nanoparticles ranging in weight percentage from 0.25 to 3% in order to study the possibility of increasing the latter's photocatalytic efficiency. Out of all the compositions evaluated, the one with the lowest Ag percentage (0.25%) was found to be the best photocatalytic composite. Similarly, mesostructured graphitic carbon nitride (MGCN) composites formed of Ag-doped melamine were employed in a different investigation to monitor the breakdown of various water contaminants including methylene blue. It was found that coating MGCN with silver nanoparticles speeds up the process of transferring photogenerated electrons from MGCN to the silver nanoparticles, improving the photocatalytic activity. When the MGCN\Ag-MGCN compound was present, more than 96% of the degradation was accomplished (Nunna et al., 2024).

Melamine was calcined using a hydrothermal technique to create three distinct photocatalysts including pure graphitic-carbon nitride (g-C_3N_4), combined with zinc oxide (ZnO/C_3N_4), and modified with silver (Ag-ZnO/C_3N_4). ZnO and Ag NPs had average particle sizes of 3-5 nm and 20–22 nm, respectively, according to TEM examination. The most efficient combination was discovered to be Ag-ZnO/C_3N_4, exhibiting strong MB degradation (97.3%) (Gan et al., 2023). In another study, zinc ferrite nanoparticles (ZF-NPs) were produced by employing *Piper nigrum* aqueous seed extract as a bio-reducing and stabilizing agent. ZF-NPs have been characterized using FTIR, SEM, FE-SEM, XRD and TGA. The results demonstrated the high purity and 60–80 nm size range of *Piper nigrum* stabilized ZF-NPs (Din et al., 2020).

TiO2 and NaOH were used as precursors in a hydrothermal reaction to create titanate nanotubes, which were then calcined for two hours at 400°C. The findings showed that while TiO2 displayed no appreciable variations in photocatalytic activity in the two systems, titanate nanotubes demonstrated superior photocatalytic degradation of MB in a simultaneous adsorption and photodegradation system than that in an equilibrium adsorption followed by a photodegradation system. Different effects of MB adsorption were hypothesized to be the cause of the titanate nanotubes' varying catalytic capabilities in the two systems (Xiong et al., 2011).

Conclusion

The reviewed studies highlight the promising potential of various photocatalysts in degrading OG, MO and MB which are prevalent in watsewaters as contaminants. MCN composites enhanced by silver nanoparticles demonstrated exceptional photocatalytic efficiency, achieving over 96% degradation. Similarly, the Ag-ZnO/C_3N_4 combination showed a remarkable 97.3% degradation rate. Zinc ferrite nanoparticles synthesized using a natural extract, exhibited effective photocatalytic properties, while titanate nanotubes outperformed conventional TiO2 in specific degradation setups. These findings underscore the importance of material modification and synthesis methods in optimizing photocatalytic activity, paving the way for advanced wastewater treatment solutions. Further studies are recommended involving the formulation of different new photocatalysts for the degradation of wastewater dyes.

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BS, SS, JSD and ZS conceived the concept, wrote and approved the manuscript.

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