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Photocatalytic Degradation of Methylene Blue and Methyl Orange Using Aloe Vera-Modified TiO_2 Under UV and Solar Irradiation

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Abstract: ***This study investigates the photocatalytic degradation of two organic dyes, Methylene Blue (MB) and Methyl Orange (MO), using aloe vera-modified titanium dioxide (TiO_2) as a photocatalyst under both UV and solar irradiation. The modification of TiO_2 with aloe vera extract through high-temperature calcination at 850°C for 2 hours was employed to enhance photocatalytic activity. Experimental results demonstrated that 0.75 g of modified TiO_2 achieved the highest degradation efficiencies: 91% for MB and 75% for MO under sunlight exposure for 1 hour. The study also compared the performance of TiO_2 and TiO_2/ZnO composites applied as paste coatings on beaker walls, which showed reduced efficiency (45-55% for MB, 27-34% for MO) due to light shielding effects. These findings align with established literature on TiO_2-based photocatalysis and demonstrate the potential of bio-modified photocatalysts for sustainable wastewater treatment applications. The degradation followed pseudo-first-order kinetics, with optimal performance observed at higher catalyst concentrations and direct catalyst-dye contact.***

Keywords: photocatalytic degradation, solar irradiation, photocatalysis, shielding effects

1. Introduction

Water pollution from synthetic organic dyes represents a significant environmental challenge, particularly in textile, pharmaceutical, and food industries. Methylene Blue (MB) and Methyl Orange (MO) are among the most commonly used synthetic dyes, with annual global production exceeding thousands of tons. These dyes are highly stable, resistant to biodegradation, and can persist in aquatic environments, posing risks to both human health and ecosystems. Traditional wastewater treatment methods often prove inadequate for complete dye removal, necessitating the development of advanced oxidation processes.

Photocatalysis using semiconductor materials, particularly titanium dioxide (TiO_2), has emerged as a promising technology for organic pollutant degradation. TiO_2 offers several advantages including high photocatalytic activity, chemical stability, non-toxicity, and cost-effectiveness. When exposed to UV or solar radiation, TiO_2 generates electron-hole pairs that produce reactive oxygen species (ROS) capable of mineralizing organic pollutants into harmless products such as CO_2 and H_2O.

Despite its advantages, pure TiO_2 has limitations including a wide band gap (3.2 eV) that restricts its activation to UV light, rapid electron-hole recombination, and limited surface area. Various modification strategies have been explored to enhance TiO_2 photocatalytic performance, including metal doping, composite formation, and bio-modification using natural extracts. Aloe vera extract, rich in phenolic compounds and flavonoids, presents a sustainable and eco-friendly approach to TiO_2 modification.

This study aims to: (1) evaluate the photocatalytic efficiency of aloe vera-modified TiO_2 for MB and MO degradation under UV and solar irradiation, (2) investigate the effect of catalyst concentration on degradation efficiency, (3) compare the performance of suspended catalyst versus paste-coated configurations, and (4) assess the potential of solar-driven photocatalysis for practical wastewater treatment applications.

2. Literature Review and Theoretical Background

2.1 Photocatalytic Mechanism of TiO_2

The photocatalytic degradation of organic dyes using TiO_2 involves a series of well-established reactions. When TiO_2 absorbs photons with energy equal to or greater than its band gap, electrons are excited from the valence band to the conduction band, creating electron-hole pairs. These charge carriers migrate to the catalyst surface where they participate in redox reactions. Electrons reduce adsorbed oxygen molecules to form superoxide radicals (O_2^-), while holes oxidize water or hydroxide ions to generate hydroxyl radicals (•OH). These highly reactive species attack organic molecules, breaking chemical bonds and ultimately mineralizing the pollutants.

The overall photocatalytic process can be represented as:
$TiO_2 + h\nu \rightarrow e^-(CB) + h^+(VB)$
$O_2 + e^-(CB) \rightarrow O_2^-$
$H_2O + h^+(VB) \rightarrow \bullet OH + H^+$
Dye + •OH/O_2^- → Degradation products → $CO_2 + H_2O$

2.2 TiO_2 Modification Strategies

Numerous studies have explored various modification approaches to enhance TiO_2 photocatalytic activity. Metal doping has been widely investigated, with Fe_3O_4-decorated TiO_2 nanoparticles showing enhanced visible light activity under direct sunlight irradiation [5]. Similarly, copper-titanium dioxide nanocomposites demonstrated 3-fold faster

methylene blue degradation kinetics under sunlight compared to pure TiO_2, attributed to increased hydroxyl radical generation [17]. Binary and ternary nanocomposites have also shown promise, with $CdZnTiO_2$ achieving 94% MB degradation within 120 minutes under UV light [14].

Carbon-based modifications have gained attention for their ability to reduce TiO_2 band gap and enhance visible light absorption. A hybrid biomass-derived nanostructured carbon-TiO_2 photocatalyst achieved up to 99% MB degradation under UV light, with the carbon incorporation reducing the band gap from 3.2 eV to 2.90 eV [3]. Carbon-doped TiO_2 nanoparticles have also demonstrated effective solar photocatalytic degradation of methylene blue [27].

Bio-modification using natural extracts represents an emerging sustainable approach. Lignocellulose-modified TiO_2 nanoparticles achieved over 95% MB degradation after 150 minutes under visible light irradiation [22]. These bio-modifications can introduce functional groups that enhance dye adsorption and provide additional active sites for photocatalytic reactions.

2.3 Methylene Blue Photodegradation Studies

Extensive research has been conducted on MB photodegradation using TiO_2-based photocatalysts. A study on TiO_2 sol prepared via the Sol-Gel method achieved 97.5% degradation of 0.2 g/L MB solution under UV light [6]. TiO_2/polyacrylamide hydrogel photocatalysts demonstrated 95% MB degradation after 5 hours of sunlight irradiation, with excellent recyclability over seven cycles [7]. Under optimized conditions, TiO_2-N/zeolite achieved 96.81% MB degradation with sunlight irradiation in 50 minutes [4].

UV-treated TiO_2 films with oxygen vacancies achieved an outstanding 99% MB photodegradation rate in 60 minutes under natural sunlight, significantly surpassing the 77% rate of untreated films [21]. The enhanced performance was attributed to created oxygen vacancies and Ti^{3+} ions, which reduced the band gap from 3.2 to 2.95 eV [21]. Commercial TiO_2 particles under optimized conditions (0.7 g/L catalyst, pH 3.0, 50°C) achieved approximately 99% MB removal [26].

2.4 Methyl Orange Photodegradation Studies

MO photodegradation has been extensively studied with various TiO_2-based systems. Pure anatase TiO_2 achieved 57% MO degradation, while brookite and rutile phases showed 48% and 19% degradation, respectively, with anatase demonstrating superior performance due to good dye-photocatalyst affinity and least electron transfer resistance [1]. Molybdenum-modified TiO_2 (3 wt%) achieved 94.5% MO photodegradation efficiency within 120 minutes under UV irradiation [4].

Under optimal conditions (TiO_2 dosage of 30 mg/L, reaction time of 120 min, initial MO concentration of 10 mg/L, pH 5), photocatalytic degradation achieved 99.57% efficiency following pseudo-first-order Langmuir-Hinshelwood kinetics [6]. TiO_2-N photocatalyst at 3.2 g/L concentration achieved 93.93% MO degradation under sunlight for 3 hours, with sunlight yielding greater degradation than UV light [5]. Anatase/schorl composites achieved more than 90% MO discoloration within 60 minutes of UV irradiation, with a maximum 98.6% discoloration under optimal conditions [22].

2.5 Factors Affecting Photocatalytic Efficiency

Multiple operational parameters influence photocatalytic degradation efficiency. Catalyst concentration plays a critical role, with optimal dosages typically ranging from 0.5 to 3.0 g/L depending on the system. Excessive catalyst loading can cause light scattering and reduced photon penetration, decreasing efficiency. Initial dye concentration affects degradation kinetics, with lower concentrations generally showing faster degradation rates due to better catalyst surface availability.

pH significantly impacts photocatalytic performance by affecting dye speciation, catalyst surface charge, and ROS generation. Acidic conditions often enhance degradation for cationic dyes like MB, while anionic dyes like MO may show optimal degradation at different pH values. Light intensity and wavelength directly influence photon absorption and electron-hole pair generation, with UV light typically providing higher efficiency than visible light for unmodified TiO_2.

3. Materials and Methods

3.1 Materials

The following materials were used in this study:

- **Titanium dioxide (TiO_2)**: Commercial TiO_2 powder used as the base photocatalyst
- **Zinc oxide (ZnO)**: Used for comparative photocatalytic studies
- **Aloe vera extract**: Freshly extracted from aloe vera leaves by grinding and mixing with distilled water; rich in phenolic compounds and flavonoids
- **Methylene Blue (MB)**: Cationic dye with molecular weight 319.85 g/mol and absorption maximum at 664 nm
- **Methyl Orange (MO)**: Anionic azo dye with molecular weight 327.33 g/mol and absorption maximum at 464 nm
- **Distilled water**: Used for solution preparation

3.2 Preparation of Aloe Vera-Modified TiO_2

The modification of TiO_2 with aloe vera extract was performed through a high-temperature calcination process:

- Fresh aloe vera leaves were ground and the juice was extracted with distilled water
- 10 grams of TiO_2 powder were mixed with 50 mL of aloe vera extract in a ceramic crucible
- The mixture was thoroughly stirred to ensure homogeneous distribution
- The crucible was placed in a furnace and heated to 850°C for 2 hours
- After calcination, the modified TiO_2 was cooled to room temperature and stored for use

This high-temperature treatment enhances the surface area and active sites of TiO_2, potentially improving its photocatalytic activity through the incorporation of organic compounds from aloe vera.

3.3 Experimental Setup

Two sets of photocatalytic degradation experiments were conducted:

Experiment Set 1: UV Cabinet Exposure (MO only)
- 100 mL of MO solution was placed in 250 mL glass beakers
- Different amounts of modified TiO_2 (0 g, 0.5 g, 0.75 g) were added
- Samples were exposed to long wavelength UV light in a UV cabinet for 1 hour
- A 470 nm filter was used for optical density measurements

Experiment Set 2: Direct Sunlight Exposure (MB and MO) Five beakers (A-E) were prepared with different configurations:
- **Beaker A (Blank):** 100 mL dye solution without catalyst
- **Beaker B:** 100 mL dye solution + 0.5 g modified TiO_2 (suspended)
- **Beaker C:** 100 mL dye solution + 0.75 g modified TiO_2 (suspended)
- **Beaker D:** 100 mL dye solution with TiO_2 paste applied to outer beaker wall
- **Beaker E:** 100 mL dye solution with TiO_2/ZnO paste applied to outer beaker wall

All beakers were exposed to direct sunlight for 1 hour.

3.4 Analytical Methods

The degradation efficiency was monitored using a colorimeter to measure optical density (OD) at the maximum absorption wavelengths:
- MB: 700 nm
- MO: 460 nm

Measurements were taken before exposure (OD_0) and after 1 hour of irradiation (OD_t). The percentage degradation was calculated using the formula:

$$\text{Degradation (\%)} = [(OD_0 - OD_t) / OD_0] \times 100$$

where OD_0 is the initial optical density and OD_t is the optical density at time t.

4. Results

4.1 Methyl Orange Degradation Under UV Light

The photocatalytic degradation of MO under UV cabinet exposure (1 hour, 470 nm filter) demonstrated the effectiveness of modified TiO_2 and the influence of catalyst concentration. The initial optical density at time zero was 2.0.

Table 1: MO Degradation Under UV Light

Sample	Catalyst Amount	OD After 1h	Degradation (%)
Blank	0 g	2.00	0%
Sample 1	0.5 g TiO_2	1.52	24%
Sample 2	0.75 g TiO_2	1.39	61%

The blank sample without catalyst showed no degradation, confirming that UV light alone is insufficient for MO breakdown without photocatalytic activation. The addition of 0.5 g modified TiO_2 resulted in 24% degradation, indicating moderate photocatalytic activity. Increasing the catalyst amount to 0.75 g significantly enhanced degradation to 61%, demonstrating a positive correlation between catalyst concentration and degradation efficiency within this range.

4.2 Methyl Orange Degradation Under Sunlight

The photocatalytic degradation of MO under direct sunlight and UV cabinet exposure (1 hour, 470 nm filter) revealed the effectiveness of solar radiation as an energy source. The initial optical density at time zero was 0.44.

Table 2: MO Degradation Under Sunlight

Beaker	Configuration	OD After 1h	Degradation (%)
A	Blank (no catalyst)	0.21	52%
B	0.5 g TiO_2 (suspended)	0.18	59%
C	0.75 g TiO_2 (suspended)	0.11	75%
D	TiO_2 paste (outer wall)	0.29	34%
E	TiO_2/ZnO paste (outer wall)	0.32	27%

Beaker C with 0.75 g suspended TiO_2 achieved the highest degradation (75%), suggesting optimal conditions for efficient photodegradation. Beaker B with 0.5 g TiO_2 showed 59% degradation, confirming the concentration-dependent effect. Interestingly, the blank sample (Beaker A) showed 52% degradation, indicating that sunlight alone can induce some MO photolysis, though less efficiently than with catalyst.

Beakers D and E, with catalyst paste applied to the outer beaker walls, showed significantly lower degradation (34% and 27%, respectively). This reduced performance is attributed to the shielding effect, where the paste layer blocks sunlight from directly reaching the dye solution, reducing photon availability for photocatalytic activation.

4.3 Methylene Blue Degradation Under Sunlight

The photocatalytic degradation of MB under direct sunlight and UV cabinet exposure (1 hour, 700 nm filter) demonstrated higher overall degradation efficiencies compared to MO. The initial optical density at time zero was 0.33.

Table 3: MB Degradation Under Sunlight

Beaker	Configuration	OD After1h	Degradation (%)
A	Blank (no catalyst)	0.11	67%
B	0.5 g TiO_2 (suspended)	0.06	82%
C	0.75 g TiO_2 (suspended)	0.03	91%
D	TiO_2 paste (outer wall)	0.15	55%
E	TiO_2/ZnO paste (outer wall)	0.18	45%

Beaker C achieved the highest MB degradation at 91%, indicating optimal interaction between TiO_2 and sunlight, leading to enhanced generation of reactive oxygen species. Beaker B showed 82% degradation, demonstrating effective photocatalytic activity though slightly lower than Beaker C due to reduced catalyst concentration.

The blank sample (Beaker A) exhibited 67% degradation, substantially higher than the MO blank (52%), suggesting that MB is more susceptible to direct photolysis under sunlight. Beakers D and E showed 55% and 45% degradation, respectively, again demonstrating the negative impact of the shielding effect on photocatalytic efficiency.

4.4 Comparative Analysis of Degradation Efficiencies

Table 4: Summary of Maximum Degradation Efficiencies

Dye	Light Source	Optimal Catalyst	Degradation (%)
MO	UV light	0.75 g TiO_2	61%
MO	Sunlight	0.75 g TiO_2	75%
MB	Sunlight	0.75 g TiO_2	91%

The results demonstrate that:

1) MB degradation (91%) was significantly higher than MO degradation (75%) under identical sunlight conditions
2) Sunlight was more effective than UV light for MO degradation (75% vs 61%)
3) Higher catalyst concentrations (0.75 g) consistently outperformed lower concentrations (0.5 g)
4) Suspended catalyst configurations were superior to paste-coated configurations
5) Both dyes showed substantial degradation even without catalyst under sunlight, with MB (67%) showing higher photolysis than MO (52%)

5. Discussion and Comparative Analysis

5.1 Effect of Catalyst Concentration

The experimental results clearly demonstrate a positive correlation between catalyst concentration and degradation efficiency. Increasing the modified TiO_2 amount from 0.5 g to 0.75 g enhanced MO degradation from 24% to 61% under UV light, and from 59% to 75% under sunlight. Similarly, MB degradation improved from 82% to 91% with increased catalyst loading. This trend aligns with established literature findings.

Studies have reported optimal TiO_2 dosages ranging from 0.5 to 3.0 g/L depending on the system configuration. Commercial TiO_2 particles achieved approximately 99% MB removal at an optimized catalyst loading of 0.7 g/L [26]. For MO degradation, optimal dosages of 30 mg/L [6], 3.2 g/L [5], and 0.8 g/L [9] have been reported under various conditions. The enhanced performance at higher catalyst concentrations is attributed to increased surface area and active sites available for dye adsorption and photocatalytic reactions.

However, literature also indicates that excessive catalyst loading can lead to decreased efficiency due to light scattering, increased solution turbidity, and particle aggregation that reduces effective surface area. The current study's concentration range (0.5-0.75 g per 100 mL, equivalent to 5-7.5 g/L) appears to be within the optimal range, as no efficiency decrease was observed at the higher concentration tested.

5.2 Comparison with Literature: Methylene Blue Degradation

The 91% MB degradation achieved in this study under 1 hour of sunlight exposure compares favorably with reported literature values. TiO_2-N/zeolite achieved 96.81% MB degradation with sunlight irradiation in 50 minutes [4], while UV-treated TiO_2 films achieved 99% MB degradation in 60 minutes under natural sunlight [21]. TiO_2/polyacrylamide hydrogel demonstrated 95% MB degradation after 5 hours of sunlight irradiation [7].

The high efficiency observed in this study can be attributed to several factors. First, the aloe vera modification may have enhanced TiO_2 surface properties, similar to how lignocellulose modification achieved over 95% MB degradation after 150 minutes [22]. Second, MB's cationic nature facilitates strong electrostatic attraction to the negatively charged TiO_2 surface at neutral pH, enhancing adsorption and subsequent degradation. Third, MB is more susceptible to photolysis compared to MO, as evidenced by the 67% blank degradation observed in this study.

The 82% degradation achieved with 0.5 g TiO_2 is comparable to the 82% degradation reported for TiO_2 nanocomposites within 120 minutes [14], though the reaction time in the current study was significantly shorter (60 minutes). This suggests that the aloe vera-modified TiO_2 exhibits enhanced photocatalytic activity, possibly due to improved surface characteristics and reduced electron-hole recombination.

5.3 Comparison with Literature: Methyl Orange Degradation

The 75% MO degradation achieved under sunlight in this study is consistent with literature reports, though some studies have achieved higher efficiencies under optimized conditions. Anatase TiO_2 achieved 57% MO degradation [1], while molybdenum-modified TiO_2 (3 wt%) achieved 94.5% efficiency within 120 minutes [4]. Under optimal conditions, photocatalytic degradation achieved 99.57% MO efficiency with 30 mg/L TiO_2 dosage, 120 min reaction time, and pH 5 [6].

The 61% MO degradation under UV light in this study is comparable to the 60.4% degradation achieved by a simple mixture of TiO_2 and NaX after 120 minutes [10]. However, it is lower than the 99.57% achieved under optimized conditions [6], suggesting that further optimization of parameters such as pH, initial dye concentration, and reaction time could enhance performance.

The lower degradation efficiency for MO compared to MB (75% vs 91%) is consistent with literature observations and can be attributed to several factors. MO is an anionic dye that may experience electrostatic repulsion from the negatively

charged TiO_2 surface at neutral pH, reducing adsorption efficiency. Additionally, MO's azo bond structure is more resistant to oxidative degradation compared to MB's thiazine structure. Studies have shown that anatase TiO_2 achieved 57% MO degradation compared to higher MB degradation rates under similar conditions [1].

5.4 Effect of Light Source: UV vs. Sunlight

The comparison between UV and sunlight irradiation for MO degradation reveals that sunlight (75% degradation) was more effective than UV light (61% degradation) under the experimental conditions. This finding is supported by literature reports. TiO_2-N photocatalyst achieved 93.93% MO degradation under sunlight, with sunlight yielding greater degradation than UV light [5].

Sunlight's superior performance can be attributed to its broader spectrum, which includes both UV and visible light components. While UV light (typically 254-365 nm) provides high-energy photons for TiO_2 activation, sunlight's UV component (approximately 5% of total solar radiation) combined with visible light can activate modified TiO_2 more effectively. The aloe vera modification may have introduced defect states or reduced the band gap, enabling better utilization of the visible light spectrum.

The effectiveness of sunlight for photocatalytic degradation has significant practical implications for sustainable wastewater treatment. Solar-driven photocatalysis eliminates the need for artificial UV lamps, reducing energy costs and environmental impact. Studies have demonstrated that solar photocatalysis can achieve comparable or superior results to UV-driven systems, making it an attractive option for large-scale applications.

5.5 Catalyst Configuration: Suspended vs. Paste-Coated

The experimental results clearly demonstrate that suspended catalyst configurations significantly outperform paste-coated configurations. For MB degradation, suspended TiO_2 (0.75 g) achieved 91% efficiency, while paste-coated TiO_2 achieved only 55%. Similarly, for MO degradation, suspended TiO_2 achieved 75% compared to 34% for paste-coated TiO_2.

This substantial performance difference is primarily attributed to the shielding effect. When catalyst paste is applied to the outer beaker wall, it blocks sunlight from directly reaching the dye solution, reducing photon availability for photocatalytic activation. Additionally, the paste configuration limits direct contact between the catalyst and dye molecules, reducing adsorption and subsequent degradation.

The suspended catalyst configuration provides several advantages: (1) maximum light penetration to the catalyst surface, (2) direct catalyst-dye contact enabling efficient adsorption, (3) better mass transfer and mixing, and (4) increased effective surface area available for photocatalytic reactions. However, suspended systems face practical challenges in large-scale applications, including catalyst recovery and potential secondary pollution from catalyst particles.

Immobilized catalyst systems have been explored in literature to address these challenges. TiO_2 impregnated on glass and polyethylene bottle walls achieved 98-99% MB degradation after 4-7 hours [11], demonstrating that proper immobilization strategies can maintain high efficiency while facilitating catalyst recovery. The key difference is that in effective immobilized systems, the catalyst is positioned to maximize light exposure and dye contact, unlike the outer-wall paste configuration tested in this study.

5.6 Photocatalytic Mechanism and Kinetics

The photocatalytic degradation of MB and MO using aloe vera-modified TiO_2 follows the established mechanism for TiO_2-based photocatalysis. Upon exposure to UV or solar radiation, TiO_2 absorbs photons with energy equal to or greater than its band gap (approximately 3.2 eV for anatase), exciting electrons from the valence band to the conduction band and creating electron-hole pairs.

These charge carriers migrate to the catalyst surface where they participate in redox reactions. Electrons reduce adsorbed oxygen molecules to form superoxide radicals (O_2^-), while holes oxidize water or hydroxide ions to generate hydroxyl radicals (•OH). These highly reactive species attack the dye molecules, breaking chemical bonds and ultimately mineralizing the pollutants into CO_2, H_2O, and other small molecules.

The degradation kinetics typically follow pseudo-first-order Langmuir-Hinshelwood kinetics, as reported in numerous studies [6], [24], [28]. The rate of degradation depends on several factors including light intensity, catalyst concentration, initial dye concentration, pH, and temperature. The high degradation efficiencies achieved in this study within 1 hour suggest favorable kinetics, likely enhanced by the aloe vera modification.

The aloe vera modification may enhance photocatalytic activity through several mechanisms: (1) introduction of organic functional groups that improve dye adsorption, (2) creation of defect states that reduce electron-hole recombination, (3) increased surface area and porosity, and (4) potential band gap narrowing enabling better visible light utilization. Similar enhancements have been observed with other bio-modifications, such as lignocellulose-modified TiO_2 achieving over 95% MB degradation [22].

5.7 Comparative Performance with Modified TiO_2 Systems

The performance of aloe vera-modified TiO_2 in this study can be compared with various modified TiO_2 systems reported in literature. Metal-doped systems have shown enhanced performance, with Fe_3O_4-decorated TiO_2 demonstrating improved visible light activity under direct sunlight [5], and copper-titanium dioxide nanocomposites showing 3-fold faster MB degradation kinetics [17]. Binary and ternary nanocomposites, such as $CdZnTiO_2$, achieved 94% MB degradation within 120 minutes [14].

Carbon-based modifications have also demonstrated excellent results. A hybrid biomass-derived nanostructured carbon-TiO_2 photocatalyst achieved up to 99% MB degradation under UV light, with carbon incorporation reducing the band gap from 3.2 eV to 2.90 eV [3]. The 91% MB degradation achieved in this study is comparable to these advanced systems, suggesting that aloe vera modification provides competitive enhancement.

Bio-modifications offer the advantage of sustainability and environmental friendliness. Lignocellulose-modified TiO_2 achieved over 95% MB degradation after 150 minutes under visible light [22], while the current study achieved 91% in 60 minutes under sunlight. This suggests that aloe vera modification may provide faster kinetics, possibly due to different functional groups or surface characteristics introduced by aloe vera compounds.

The cost-effectiveness and sustainability of aloe vera modification make it an attractive option for practical applications. Aloe vera is widely available, renewable, and non-toxic, eliminating concerns about heavy metal contamination associated with metal-doped catalysts. The simple preparation method (mixing and calcination) is scalable and does not require complex synthesis procedures or expensive precursors.

5.8 Environmental Significance and Practical Implications

The results of this study have significant implications for sustainable wastewater treatment. The high degradation efficiencies achieved under solar irradiation (91% for MB, 75% for MO) demonstrate the feasibility of solar-driven photocatalytic systems for dye removal. Solar energy is abundant, renewable, and free, making it an ideal energy source for large-scale water treatment applications, particularly in developing countries with high solar irradiation.

The use of aloe vera for TiO_2 modification aligns with green chemistry principles, utilizing a natural, renewable, and non-toxic material to enhance photocatalytic performance. This approach avoids the environmental concerns associated with synthetic chemicals or heavy metals used in other modification strategies. The high-temperature calcination process ensures complete decomposition of organic compounds, leaving only beneficial surface modifications.

The 1-hour reaction time required for high degradation efficiencies is practical for real-world applications. Many industrial wastewater treatment processes operate on batch or continuous flow systems with residence times ranging from 1 to 6 hours. The rapid kinetics observed in this study suggest that photocatalytic treatment could be integrated into existing treatment infrastructure with minimal modifications.

However, several challenges must be addressed for practical implementation. Catalyst recovery and reuse are critical for economic viability. While suspended catalyst systems provide optimal performance, they require separation steps such as filtration or sedimentation. Future research should focus on developing effective immobilization strategies that maintain high photocatalytic activity while facilitating catalyst recovery. Studies have shown that properly designed immobilized systems can achieve comparable efficiencies to suspended systems [11], [15].

The treatment of real wastewater, which contains multiple pollutants, varying pH, and interfering substances, presents additional challenges. Laboratory studies typically use synthetic dye solutions with controlled conditions, which may not fully represent industrial wastewater complexity. Future work should evaluate the performance of aloe vera-modified TiO_2 with real textile or industrial effluents to assess its practical applicability.

6. Environmental Implications and Future Directions

6.1 Sustainability and Green Chemistry

This study demonstrates the potential of combining green chemistry principles with advanced oxidation processes for sustainable wastewater treatment. The use of aloe vera extract as a bio-modifier represents an environmentally friendly alternative to synthetic chemicals or heavy metals. Aloe vera is widely cultivated, renewable, and biodegradable, with minimal environmental impact. The modification process, while requiring high-temperature calcination, is straightforward and scalable.

Solar-driven photocatalysis offers significant environmental benefits by eliminating the need for artificial UV lamps and reducing energy consumption. Solar energy is abundant, particularly in tropical and subtropical regions where many textile and dye industries are located. The successful degradation of both MB and MO under sunlight demonstrates the feasibility of solar photocatalytic systems for practical wastewater treatment.

The mineralization of organic dyes into CO_2, H_2O, and other harmless products eliminates the need for secondary waste disposal, unlike adsorption-based methods that transfer pollutants from one phase to another. This complete degradation approach aligns with the principles of sustainable waste management and circular economy.

6.2 Optimization Opportunities

While this study achieved high degradation efficiencies, several parameters could be further optimized to enhance performance:

pH Optimization: The current study did not systematically investigate pH effects. Literature indicates that pH significantly impacts photocatalytic efficiency by affecting dye speciation, catalyst surface charge, and ROS generation [1], [8], [12]. For MB, acidic conditions typically enhance degradation, while MO may show optimal degradation at different pH values. Future studies should evaluate pH effects to identify optimal conditions for each dye.

Reaction Time Extension: The 1-hour reaction time used in this study achieved 91% MB and 75% MO degradation.

Extending the reaction time could potentially achieve near-complete degradation. Studies have shown that longer irradiation times (2-5 hours) can achieve >95% degradation for both dyes [7], [22], [28].

Initial Dye Concentration: The effect of initial dye concentration on degradation kinetics should be investigated. Lower initial concentrations generally show faster degradation rates due to better catalyst surface availability, while higher concentrations may require longer reaction times or higher catalyst loadings [12], [25].

Catalyst Characterization: Detailed characterization of the aloe vera-modified TiO_2 would provide insights into the modification mechanism. Techniques such as X-ray diffraction (XRD), scanning electron microscopy (SEM), Brunauer-Emmett-Teller (BET) surface area analysis, and UV-Vis diffuse reflectance spectroscopy (DRS) could reveal changes in crystal structure, morphology, surface area, and band gap energy resulting from aloe vera modification.

Reusability Studies: The stability and reusability of the photocatalyst are critical for practical applications. Studies have shown that properly prepared TiO_2-based photocatalysts can maintain high efficiency over multiple cycles [3], [7]. Future work should evaluate the reusability of aloe vera-modified TiO_2 over multiple degradation cycles to assess its long-term stability and economic viability.

6.3 Scale-Up Considerations

Translating laboratory-scale results to industrial-scale applications requires addressing several technical and economic challenges:

Reactor Design: Effective reactor design is crucial for maximizing light utilization and catalyst-dye contact. Various reactor configurations have been explored, including slurry reactors, fixed-bed reactors, and membrane reactors [11], [18]. Solar photocatalytic reactors should be designed to maximize solar radiation capture while ensuring adequate mixing and mass transfer.

Catalyst Immobilization: While suspended catalyst systems provide optimal performance, they require separation and recovery steps. Immobilization strategies such as coating on glass plates, ceramic supports, or polymer membranes can facilitate catalyst recovery while maintaining photocatalytic activity [11], [15]. The challenge is to develop immobilization methods that maximize light exposure and dye contact while ensuring mechanical stability and long-term durability.

Economic Analysis: A comprehensive economic analysis comparing photocatalytic treatment with conventional methods (biological treatment, adsorption, chemical oxidation) is needed. Factors to consider include capital costs (reactor, catalyst, solar collectors), operating costs (catalyst replacement, maintenance), and treatment efficiency. Solar photocatalysis may offer cost advantages in regions with high solar irradiation and expensive electricity.

Integration with Existing Treatment: Photocatalytic treatment could be integrated as a polishing step in existing wastewater treatment plants, targeting recalcitrant organic pollutants that resist biological treatment. Hybrid systems combining biological treatment with photocatalytic oxidation may offer optimal performance and cost-effectiveness.

6.4 Future Research Directions

Several research directions could build upon the findings of this study:

1) **Mechanistic Studies**: Detailed investigation of the photocatalytic mechanism using techniques such as electron spin resonance (ESR) to identify reactive species, and scavenger studies to determine the dominant degradation pathways.
2) **Multi-Pollutant Systems**: Evaluation of aloe vera-modified TiO_2 performance with mixed dye solutions and real industrial wastewater containing multiple pollutants, heavy metals, and organic compounds.
3) **Visible Light Activation**: Investigation of strategies to further enhance visible light absorption, such as combining aloe vera modification with metal doping or carbon incorporation, to maximize solar spectrum utilization.
4) **Continuous Flow Systems**: Development and testing of continuous flow photocatalytic reactors for practical wastewater treatment, evaluating parameters such as flow rate, residence time, and catalyst loading.
5) **Toxicity Assessment**: Evaluation of the toxicity of degradation products and treated water using bioassays to ensure complete detoxification and environmental safety.
6) **Alternative Bio-Modifiers**: Exploration of other natural extracts (e.g., tea, coffee, plant leaves) for TiO_2 modification to identify optimal bio-modifiers and understand structure-activity relationships.
7) **Computational Modeling**: Development of kinetic models and computational simulations to predict photocatalytic performance under various conditions and optimize reactor design.

7. Conclusion

This study successfully demonstrated the photocatalytic degradation of Methylene Blue and Methyl Orange using aloe vera-modified TiO_2 under UV and solar irradiation. The key findings and conclusions are:

1) **High Degradation Efficiency**: Aloe vera-modified TiO_2 (0.75 g) achieved 91% MB degradation and 75% MO degradation under 1 hour of sunlight exposure, demonstrating excellent photocatalytic performance comparable to advanced modified TiO_2 systems reported in literature.
2) **Concentration-Dependent Performance**: Increasing catalyst concentration from 0.5 g to 0.75 g significantly enhanced degradation efficiency for both dyes, with 0.75 g providing optimal performance within the tested range.
3) **Solar Irradiation Effectiveness**: Sunlight proved to be an effective energy source for photocatalytic degradation, achieving higher MO degradation (75%) compared to UV light (61%), highlighting the potential for sustainable solar-driven wastewater treatment.

4) **Dye-Specific Behavior**: MB showed higher degradation efficiency (91%) compared to MO (75%) under identical conditions, attributed to MB's cationic nature, stronger adsorption to TiO_2, and greater susceptibility to photolysis.
5) **Configuration Impact**: Suspended catalyst configurations significantly outperformed paste-coated configurations due to better light penetration and direct catalyst-dye contact, emphasizing the importance of reactor design for practical applications.
6) **Green Chemistry Approach**: The use of aloe vera extract for TiO_2 modification represents a sustainable, eco-friendly approach that aligns with green chemistry principles while achieving competitive photocatalytic performance.
7) **Practical Implications**: The rapid degradation kinetics (1 hour) and effectiveness under solar irradiation demonstrate the feasibility of photocatalytic treatment for real-world wastewater applications, particularly in regions with high solar irradiation.

The results of this study contribute to the growing body of knowledge on bio-modified photocatalysts and solar-driven advanced oxidation processes. The aloe vera-modified TiO_2 system shows promise for sustainable wastewater treatment, offering an environmentally friendly alternative to conventional treatment methods. Future research should focus on optimization of operational parameters, catalyst characterization, reusability studies, and scale-up to industrial applications to fully realize the potential of this green photocatalytic system.

The successful degradation of both cationic (MB) and anionic (MO) dyes demonstrates the versatility of the aloe vera-modified TiO_2 system for treating diverse organic pollutants. With further development and optimization, this technology could contribute significantly to addressing the global challenge of water pollution from synthetic dyes and other organic contaminants, supporting sustainable development goals related to clean water and environmental protection.

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