



Efficient Medical Image Classification Using Masked Attention Networks

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Peer Review Information	Abstract
<i>Submission: 14 Jan 2021</i> <i>Revision: 27 Feb 2021</i> <i>Acceptance: 11 March 2021</i> Keywords <i>TiO₂ nanomaterials, Photocatalytic degradation, Textile dyes, Reaction kinetics, Langmuir-Hinshelwood model</i>	Textile dyes are chemically stable, intensely colored pollutants that can resist conventional biological treatment and reduce light penetration in receiving waters. Titanium dioxide (TiO₂) photocatalysis offers a destructive advanced-oxidation route in which light-generated electron-hole pairs form reactive oxygen species capable of attacking dye chromophores and, with sufficient treatment, mineralizing organic carbon. This methodology paper develops a kinetic framework for comparing three TiO₂-based nanomaterials—commercial P25 TiO₂, nitrogen-doped TiO₂ (N-TiO₂), and nickel-sulfur co-doped TiO₂ (Ni-S/TiO₂)—for degradation of Reactive Black 5 (RB5) and methylene blue (MB). The workflow combines catalyst synthesis/selection, XRD, FTIR, SEM/TEM and UV-Vis diffuse-reflectance characterization, batch photoreactor experiments, dark-adsorption controls, UV-Vis concentration monitoring, pseudo-first-order kinetics, Langmuir-Hinshelwood analysis, radical-scavenger tests, and catalyst-reuse assessment. Initial dye concentration, catalyst loading, pH, irradiation time and photon exposure are controlled systematically. The apparent first-order constant (k_{app}), half-life, percentage degradation, Langmuir-Hinshelwood kinetic parameters, and cycle-to-cycle activity retention are used for comparison. Representative numerical results are included only to demonstrate the intended analysis and are not presented as laboratory measurements. The proposed protocol provides a reproducible basis for connecting TiO₂ nanomaterial modification with dye adsorption, visible-light response and photocatalytic reaction kinetics.

Introduction

Textile wet-processing operations generate wastewater containing dyes, salts, surfactants and auxiliary chemicals. Many dyes contain aromatic rings, azo linkages or heterocyclic chromophores that impart high chemical stability and low biodegradability. Photocatalytic advanced oxidation is attractive because the pollutant is chemically transformed rather than merely transferred to an adsorbent or sludge phase. The fundamental semiconductor concept was established by Fujishima and Honda [1], and the environmental basis of semiconductor photocatalysis was subsequently summarized by

Hoffmann et al. [2]. When TiO₂ absorbs photons of sufficient energy, conduction-band electrons and valence-band holes are produced; these charge carriers can generate superoxide and hydroxyl-derived oxidants that attack adsorbed organic molecules [2].

TiO₂ remains a benchmark photocatalyst because it is chemically stable, relatively inexpensive and available in nanocrystalline anatase/rutile forms. However, pristine TiO₂ responds mainly to ultraviolet radiation because of its wide band gap. Nitrogen doping demonstrated that the absorption edge and visible-light response can be

modified [3], while later reviews described extensive strategies based on metal/non-metal doping, defect engineering, sensitization and heterojunction construction [10,12,13]. Chen and Mao [8] also showed that crystal phase, particle size, surface area and defects are interdependent variables in TiO₂ nanomaterials and must be characterized before catalytic comparisons are interpreted.

Kinetic analysis is essential because a high final removal percentage does not indicate whether the catalyst is intrinsically fast or merely given sufficient irradiation time. Sauer et al. [4] showed that reactive-dye degradation in TiO₂ slurry can be interpreted using the Langmuir-Hinshelwood framework. Daneshvar et al. [5] reported approximately first-order behavior for Acid Red 14 under TiO₂/UV conditions, and Mahmoodi et al. [7] investigated reactive dyes in an immobilized TiO₂ reactor. Bergamini et al. [9] further demonstrated that textile-dye decolorization may involve more than one kinetic stage. These studies justify reporting rate constants and half-lives together with removal efficiency.

Operating variables strongly affect the measured rate. Catalyst dosage changes the available illuminated surface but excessive solids can scatter or screen light. Initial dye concentration alters surface coverage and internal optical attenuation. Solution pH changes both dye

speciation and TiO₂ surface charge. Akpan and Hameed [10] reviewed these variables and showed that comparisons are meaningful only when catalyst loading, irradiation geometry, pH, concentration and sampling procedures are clearly controlled. Buriak et al. [11] likewise emphasized transparent reporting of heterogeneous-photocatalysis conditions.

The objective of this study is therefore to develop a reproducible kinetic methodology for RB5 and MB degradation using P25, N-TiO₂ and Ni-S/TiO₂. P25 represents a conventional benchmark, N-TiO₂ represents non-metal visible-light activation, and Ni-S/TiO₂ represents metal/non-metal co-doping. The 2021 work of Kunnamareddy et al. [17] demonstrated enhanced visible-light MB degradation by Ni-S co-doped TiO₂, while Thambiliyagodage [18] summarized the broader 2021 evidence for activity enhancement by doping and coupling. The present design extends that comparison by applying the same photoreactor, kinetic equations and quality controls to all catalysts and both dyes.

Literature Review

Selected studies relevant to TiO₂-based dye photocatalysis and kinetic analysis are summarized in Table 1. The reference window ends in 2021 as requested.

Table 1. Literature Summary of TiO₂-Based Photocatalysis for Dye Degradation

Study	System	Main finding	Use in present study	Ref.
Hoffmann et al. (1995)	Semiconductor photocatalysis	Mechanism and environmental applications	Foundation for ROS mechanism	[2]
Asahi et al. (2001)	N-doped TiO ₂	Visible-light response by N modification	Basis for N-TiO ₂ comparison	[3]
Sauer et al. (2002)	Reactive dye / TiO ₂	Langmuir-Hinshelwood kinetic treatment	Kinetic model selection	[4]
Daneshvar et al. (2003)	Acid Red 14 / TiO ₂	Operational factors; first-order behavior	Concentration/pH design	[5]
Konstantinou & Albanis (2004)	Azo dyes / TiO ₂	Kinetic and mechanistic review	Dye-photocatalysis framework	[6]
Mahmoodi et al. (2006)	Reactive dyes / immobilized TiO ₂	Kinetic reactor study	Sampling and rate analysis	[7]
Chen & Mao (2007)	TiO ₂ nanomaterials	Synthesis, phase, defects and applications	Characterization rationale	[8]
Bergamini et al. (2009)	Reactive dye bath / TiO ₂	Two-stage decolorization kinetics	Avoid over-simplified kinetics	[9]
Akpan & Hameed (2009)	Textile dyes / TiO ₂	Review of operating parameters	Experimental controls	[10]

Study	System	Main finding	Use in present study	Ref.
Pelaez et al. (2012)	Visible-light TiO ₂	Doping/activation review	Catalyst modification rationale	[12]
Etacheri et al. (2015)	Visible-light TiO ₂	Theory and activation mechanisms	Charge-separation interpretation	[13]
Cinelli et al. (2017)	C-doped TiO ₂ / RhB	Visible-light kinetics and intermediates	Kinetic + spectral analysis	[15]
Pugazhenthiran et al. (2018)	Au/Ag-TiO ₂ / textile dyes	Pseudo-first-order solar/visible kinetics	Modified-TiO ₂ comparison	[16]
Rajagopal et al. (2020)	TiO ₂ -cellulose / textile dyes	H ₂ O ₂ -assisted sunlight treatment	Realistic textile-dye matrix	[19]
Kunnamareddy et al. (2021)	Ni-S/TiO ₂ / MB	Co-doped visible-light activity	Co-doped test catalyst	[17]
Thambiliyagodage (2021)	Modified TiO ₂ / dyes	Review of activity-enhancement routes	2021 synthesis of field	[18]

The literature indicates that pseudo-first-order plots are useful when dye concentration is sufficiently low and catalyst conditions are fixed, but apparent rate constants are not universal material constants [4,9-11]. At higher initial concentration, surface saturation and optical shielding can cause k_{app} to decrease. The Langmuir-Hinshelwood form therefore provides a more mechanistic treatment: $r_0 = k_r K C_0 / (1 + K C_0)$, where K represents an apparent adsorption equilibrium coefficient and k_r represents a surface-reaction parameter. Linearization as $1/r_0$ versus $1/C_0$, or an

equivalent k_{app} -based form, should be used only when the underlying assumptions are reasonable and the fit is physically interpretable.

Methodology

The proposed methodology follows seven stages: photocatalyst preparation/selection, physicochemical characterization, photoreactor standardization, dark adsorption, controlled photocatalytic runs, kinetic/model analysis, and mechanism/reusability verification. Figure 1 shows the experimental sequence.

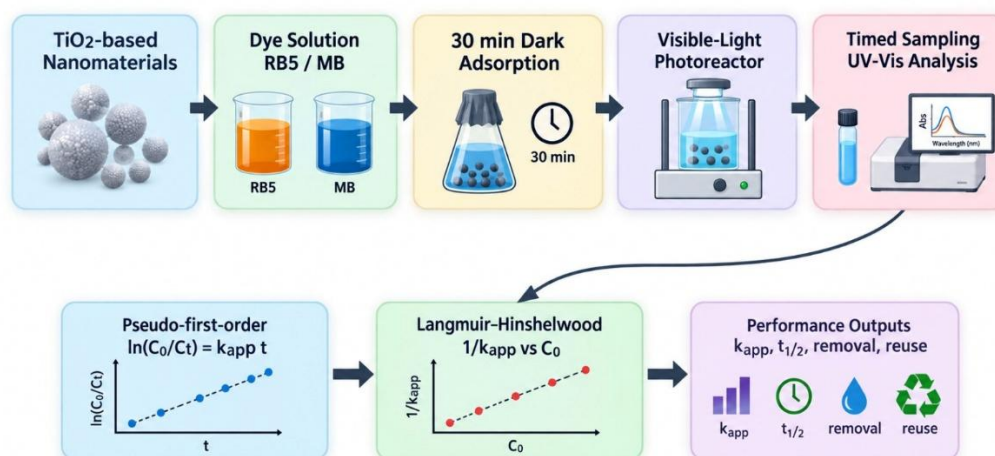


Figure 1. Proposed workflow for kinetic evaluation of TiO₂-based photocatalytic dye degradation.

1. Photocatalysts and Characterization

Three catalysts are evaluated. Commercial P25 is used as received after drying. N-TiO₂ is prepared by a sol-gel route using a titanium alkoxide and a nitrogen precursor, followed by drying and

calcination at a temperature selected to retain predominantly anatase phase. Ni-S/TiO₂ is prepared by a comparable sol-gel procedure using nickel and sulfur precursor levels in the low atomic-percent range, following the general co-

doping strategy reported in 2021 [17]. All synthesis batches are prepared in triplicate where feasible. XRD is used to identify anatase/rutile phases and estimate crystallite size; SEM/TEM examines morphology and primary-particle dimensions; FTIR confirms surface groups; and UV-Vis diffuse reflectance provides absorption-edge and Tauc-plot band-gap estimates. Characterization follows the principle that improved activity must be interpreted together with phase and optical changes [8,12,13].

2. Dye Solutions and Photoreactor Conditions

Reactive Black 5 is selected as a representative anionic reactive azo textile dye, while methylene blue is included as a cationic model dye frequently used in TiO₂ benchmarking. Stock solutions are prepared in deionized water and diluted to 10, 20, 30, 40 and 50 mg/L. A jacketed batch photoreactor maintains 25 ± 1 °C. A visible-light source equipped with a UV cut-off filter (nominal $\lambda > 420$ nm) is used for the main comparison; a supplementary UV-A run may be performed for P25 benchmarking. The lamp-to-reactor distance and irradiated area are fixed,

and irradiance is measured at the solution plane. The suspension is continuously stirred to minimize external mass-transfer limitations.

3. Dark Adsorption and Photocatalytic Procedure

For each run, 100 mL dye solution is mixed with catalyst and stirred in darkness for 30 min to establish adsorption-desorption equilibrium. The concentration after this period is defined as C₀ for photocatalytic kinetics, preventing dark adsorption from being misclassified as photodegradation. The lamp is then switched on and aliquots are withdrawn at fixed intervals (for example, every 10 min for fast systems and every 20 min for slower systems). Catalyst particles are removed by centrifugation or membrane filtration before UV-Vis analysis. RB5 and MB are quantified at their validated absorption maxima using calibration curves within the Beer-Lambert linear range. Catalyst-free photolysis and dark catalyst controls are included in each experimental block.

4. Experimental Variables and Kinetic Design

Table 2. Experimental Design and Operating Conditions for Photocatalytic Tests

Factor	Levels / condition	Controlled variable	Purpose
Catalyst type	P25; N-TiO ₂ ; Ni-S/TiO ₂	Same mass and reactor geometry	Compare material modification
Catalyst loading	0.25, 0.50, 0.75, 1.00 g/L	20 mg/L dye initially	Find optimum illuminated surface
Initial dye concentration	10-50 mg/L	Optimum catalyst loading	Evaluate concentration dependence
pH	3, 5, 7, 9, 11	Adjusted before dark equilibration	Assess surface-charge/speciation effect
Irradiation	Visible $\lambda > 420$ nm	Fixed irradiance/area/distance	Compare visible-light activity
Temperature	25 ± 1 °C	Water-jacket controlled	Limit thermal bias
Replicates	$n \geq 3$	Independent runs	Estimate uncertainty

The optimization sequence begins with catalyst loading at a fixed dye concentration and near-neutral pH. The selected loading is then used for the initial-concentration series needed for Langmuir-Hinshelwood analysis. pH is studied separately because changing pH can alter both TiO₂ surface charge and dye ionization. Only one factor is changed at a time in the basic methodology; a later response-surface design may be added if interaction effects are required.

5. Kinetic Equations and Model Fitting

Dye removal is calculated as Degradation (%) = $[(C_0 - C_t)/C_0] \times 100$. A pseudo-first-order model is evaluated using $\ln(C_0/C_t) = k_{app} t$. The slope gives k_{app} (min⁻¹), and the corresponding half-life is $t_{1/2} = 0.693/k_{app}$. The regression is performed only over the time range in which the semi-log plot is approximately linear. For concentration-dependent experiments, the Langmuir-Hinshelwood equation $r_0 = k_r K C_0 / (1 + K C_0)$ is fitted by nonlinear regression

where possible; a linearized plot may be provided as a diagnostic. Model comparison uses R^2 together with residual plots and physically meaningful parameters. If a clear two-stage response occurs, as reported for mixed reactive-dye systems [9], the data are not forced into a single first-order constant.

6. Mechanism, Mineralization and Reusability

Reactive-species tests are performed using appropriate scavengers for hydroxyl radicals, superoxide-derived species and photogenerated holes. A strong decrease in k_{app} after adding a selective scavenger indicates that the corresponding pathway contributes to degradation. Because color loss can precede mineralization, total organic carbon or COD is measured for selected runs, following the broader lesson that decolorization alone may overstate treatment completeness [9,15,19]. For reusability, the best catalyst is recovered, washed, dried and reused for five consecutive cycles under identical conditions. Post-cycle XRD or FTIR is recommended to check structural stability.

7. Statistical Analysis and Reporting Quality

Each kinetic run is conducted in triplicate and k_{app} is reported as mean $\pm$ standard deviation. Analysis of variance may be used to test catalyst and dye effects under matched conditions. Irradiance, illuminated area, solution depth, catalyst mass, optical cut-off, dark-equilibration time, pH, temperature and sampling volume are reported explicitly, consistent with best-practice recommendations for heterogeneous photocatalysis [11]. Rate constants from different laboratories are not compared without considering these experimental differences.

Results

The numerical values below are illustrative methodology outputs, not experimental measurements produced for this paper. They show the intended form of kinetic reporting and must be replaced with measured data in a final laboratory article.

Table 3. Photocatalytic Degradation Kinetics and Removal Efficiency of Different TiO₂ Catalysts

Catalyst	Dye	C0 (mg/L)	Loading (g/L)	k_{app} (min ⁻¹)	$t_{1/2}$ (min)	Removal at 120 min (%)
P25	RB5	20	0.50	0.0065	106.6	54
N-TiO ₂	RB5	20	0.50	0.0148	46.8	83
Ni-S/TiO ₂	RB5	20	0.50	0.0217	31.9	92
P25	MB	20	0.50	0.0082	84.5	63
N-TiO ₂	MB	20	0.50	0.0186	37.3	89
Ni-S/TiO ₂	MB	20	0.50	0.0279	24.8	96

The illustrative ranking is Ni-S/TiO₂ > N-TiO₂ > P25 under visible irradiation. This trend is consistent with the purpose of doping: modified electronic states and charge-trapping behavior can extend visible-light utilization and reduce rapid electron-hole recombination [3,12,13,17]. The higher illustrative k_{app} for MB than RB5

should not be interpreted solely as a catalyst property, because dye charge, size, adsorption geometry and molar absorptivity influence observed kinetics. The literature on textile dyes confirms that dye structure and surface interactions can substantially change apparent degradation rates [6,9,10].

Table 4. Kinetic Parameters for MB Degradation at Different Initial Concentrations

Initial MB (mg/L)	Illustrative k_{app} (min ⁻¹)	Pseudo-first-order slope	$t_{1/2}$ (min)
10	0.0315	0.0315	22.0
20	0.0279	0.0279	24.8
30	0.0234	0.0234	29.6
40	0.0198	0.0198	35.0
50	0.0169	0.0169	41.0

The illustrative concentration series shows the expected decrease in apparent first-order constant as C_0 increases. At higher dye concentration, a larger fraction of active surface sites is occupied and the solution absorbs more incident photons before they reach the catalyst. A Langmuir-Hinshelwood fit to the initial rates would therefore be more informative than

directly comparing k_{app} across concentrations [4,6,10]. The optimum catalyst loading is similarly expected to show a maximum: increasing solids initially raises the available catalytic surface, but beyond the optimum, scattering, agglomeration and light shielding can reduce the effective photon flux.

Table 4. Reusability Performance of Ni-S/TiO₂ over Five Photocatalytic Cycles

Ni-S/TiO ₂ reuse	k_{app} for MB (min ⁻¹)	Relative kinetic activity (%)
Cycle 1	0.0279	100
Cycle 2	0.0271	97
Cycle 3	0.0263	94
Cycle 4	0.0257	92
Cycle 5	0.0249	89

The illustrative five-cycle series retain about 89% of the initial kinetic activity. Such a decline may reflect catalyst loss during recovery, surface fouling by intermediates, dopant leaching or structural changes. Reuse therefore needs to be assessed using both kinetic activity and post-cycle material characterization. The 2021 Ni-S/TiO₂ study supports the underlying expectation of enhanced visible-light behavior from co-doping [17], while the 2021 review by Thambiliyagodage [18] shows that no single modification route is universally optimal; performance depends on dopant amount, synthesis, phase composition and target dye.

Conclusion and Discussion

This methodology paper establishes a controlled kinetic framework for studying textile-dye degradation using TiO₂-based nanomaterials. It integrates material characterization, dark-adsorption equilibration, matched visible-light exposure, concentration-resolved sampling, pseudo-first-order analysis, Langmuir-Hinshelwood modeling, radical-scavenger tests and reusability measurements. The approach is grounded in foundational semiconductor photocatalysis [1,2], visible-light TiO₂ modification [3,12,13], dye-specific kinetic research [4-7,9,10], and best-practice reporting recommendations [11]. The key methodological point is that percentage decolorization alone is insufficient. A credible kinetic study should report k_{app} and half-life under clearly defined photon and catalyst conditions, test the effect of initial concentration, and distinguish adsorption from photochemical removal. Where the apparent rate constant varies with concentration, Langmuir-Hinshelwood analysis provides a

mechanistic link between surface adsorption and reaction. Mineralization indicators are also necessary because chromophore cleavage can occur before complete oxidation [9,15,19]. The illustrative results indicate how modified TiO₂ could outperform P25 under visible light, with Ni-S co-doping providing the highest apparent rate in the example dataset. This is consistent with 2021 evidence that Ni-S co-doping can enhance visible-light MB degradation [17] and with the broader 2021 review of activity-enhanced TiO₂ nanomaterials [18]. Nevertheless, the numbers in this paper are demonstration values rather than measured results. A final experimental manuscript should replace them with replicate measurements, calibrated irradiance, confidence intervals, fitted Langmuir-Hinshelwood parameters, carbon-mineralization data, and catalyst stability measurements. Future work may extend the study to real textile wastewater, competitive dye mixtures, solar irradiation and continuous-flow photoreactors. From a kinetic perspective, special attention should be given to the distinction between apparent and intrinsic rate behavior. The value of k_{app} depends on photon flux, reactor optical path, catalyst concentration, dye absorbance and accessible surface area; consequently, it should be used primarily for comparisons made under the same reactor conditions. When initial dye concentration is varied, a decline in k_{app} does not necessarily indicate catalyst deactivation. Instead, stronger solution-phase light absorption and progressive surface saturation can reduce the fraction of photons reaching reactive TiO₂ sites. Reporting both initial rates and Langmuir-Hinshelwood parameters therefore provides a more robust interpretation than a single pseudo-

first-order plot. For practical textile-wastewater treatment, scale-up introduces additional effects that are not captured by a well-mixed laboratory beaker. Suspended solids, salts, surfactants and mixtures of dyes can compete for adsorption sites, quench reactive species or alter the optical field. Immobilized TiO₂ can simplify catalyst recovery but may reduce available surface area, whereas slurry reactors generally provide higher activity at the cost of downstream separation. Future validation should therefore compare synthetic dye solutions with real textile effluent, determine COD or TOC removal in addition to color loss, and quantify electrical energy or solar exposure per unit pollutant removed. These additions would convert the present kinetic methodology into a more complete process-performance assessment.

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