



Challenge Journal of CONCRETE RESEARCH LETTERS

Research Article

Effectiveness of TiO₂ coating on concrete surfaces for photocatalytic degradation of solutions

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ABSTRACT

In this study, the photocatalytic performance of TiO₂-coated concrete surfaces was systematically investigated with respect to coating configuration and catalyst loading using methylene blue (MB) and Rhodamine B (Rh-B) as model organic pollutants. TiO₂ was applied onto concrete surfaces at different concentrations (5, 10, 15, and 20 wt%) in both single-layer and double-layer configurations, and the degradation behavior of the dyes under UV irradiation was evaluated. The results revealed that increasing TiO₂ content significantly enhanced photocatalytic activity in both coating configurations. However, double-layer coatings consistently outperformed single-layer systems, particularly at higher TiO₂ loadings. The maximum degradation efficiencies reached 52.12% for MB and 77.4% for Rh-B under UV irradiation, demonstrating the effectiveness of TiO₂-coated surfaces in removing organic pollutants. Kinetic analysis indicated that the photocatalytic degradation process followed pseudo-first-order kinetics, with higher apparent rate constants (K_{app}) observed in double-layer systems. This enhancement was attributed to increased availability of active sites, improved light utilization, and more efficient charge-carrier dynamics. Mechanistically, the generation of reactive oxygen species (ROS), such as hydroxyl and superoxide radicals, along with improved charge separation facilitated by surface hydroxyl groups and oxygen vacancies, played a key role in enhancing photocatalytic performance. Overall, the findings demonstrate that optimizing both TiO₂ loading and coating architecture provides an effective strategy for improving the photocatalytic efficiency of cement-based materials. The proposed double-layer TiO₂ coating system offers significant potential for developing self-cleaning, environmentally responsive construction materials for urban applications.

ARTICLE INFO

Article history:

Received – March 3, 2026
Revision requested – March 30, 2026
Revision received – April 17, 2026
Accepted – April 24, 2026

Keywords:

Titanium dioxide
Photocatalytic degradation
Self-cleaning concrete
Coating configuration
Catalyst loading
UV irradiation



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Citation: Şamdan F, Şamdan C, Günaltı E, Ünal S, Canbaz M (2026). Effectiveness of TiO₂ coating on concrete surfaces for photocatalytic degradation of solutions. *Challenge Journal of Concrete Research Letters*, 17(3), 250–261.

1. Introduction

Rapid urbanization and industrial growth have led to a significant increase in environmental pollution, especially in densely populated areas, where emissions from transportation, industrial processes, and construction activities accumulate on building surfaces. These pollutants, including nitrogen oxides (NO_x), volatile organic compounds (VOCs), and particulate matter, not only

worsen environmental quality but also pose serious risks to human health and the durability and appearance of building materials (Wei et al. 2023b). The durability of cementitious materials is likewise sensitive to chemically aggressive environmental exposure (Al-Safi et al. 2025). Conventional pollutant removal strategies include mechanical surface cleaning (Pacheco-Torgal 2014), chemical washing with detergents or oxidizing agents (Chen and Poon 2009; Fujishima et al. 2008), and

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physicochemical processes such as adsorption (Rashed 2013) and filtration (Shannon et al. 2008). However, these approaches are often energy-intensive, require repeated application, and may generate secondary pollution, thereby underscoring the need for more sustainable and passive solutions. This broader sustainability objective is consistent with current construction-material research emphasizing resource efficiency, waste valorization, and reduced environmental impact (Mahmud et al. 2025; Turan et al. 2025; Ünal and Canbaz 2025b, 2026; Wee Chit et al. 2025). In this context, photocatalytic technologies have emerged as a promising approach for environmental cleanup, enabling the breakdown of organic and inorganic pollutants through light-triggered reactions without the need for additional chemicals (Arif et al. 2026). As part of advanced oxidation processes (AOPs), photocatalysis relies on the generation of highly reactive species that convert pollutants into less harmful or inert substances. This makes it particularly suitable for long-term, low-maintenance applications in the built environment (Zhang et al. 2024).

In recent years, photocatalytic materials have been increasingly integrated into functional surfaces to impart self-cleaning properties, particularly in environments where persistent pollutant accumulation poses significant environmental and aesthetic challenges. In this context, photocatalysis is recognized as an effective and sustainable strategy within advanced oxidation processes (AOPs), enabling the degradation of resistant organic contaminants that are often not efficiently removed by conventional treatment methods (Shamim et al. 2026). The self-cleaning concept refers to a material's ability to autonomously decompose and remove surface-bound pollutants under UV irradiation, thereby minimizing the need for external cleaning interventions. This approach is particularly relevant for cementitious materials exposed to urban pollution, where the accumulation of organic and inorganic contaminants can adversely affect both surface appearance and long-term performance. Therefore, incorporating photocatalytic materials such as TiO_2 into construction materials has emerged as a promising strategy for developing multifunctional systems capable of environmental remediation and passive surface maintenance (Paiu et al. 2025).

The self-cleaning behavior of TiO_2 -coated surfaces is generally attributed to two complementary mechanisms: photocatalytic degradation and photo-induced hydrophilicity. Under UV irradiation, TiO_2 generates reactive oxygen species that oxidize and decompose organic contaminants adsorbed on the surface (Singh et al. 2026). Simultaneously, UV exposure induces a transition toward a superhydrophilic surface state, reducing the water contact angle and enabling water to spread uniformly across the surface (Kumar et al. 2025). This process facilitates the removal of degradation products and residual contaminants through a washing effect, preventing their accumulation on the material surface. The synergistic action of these two mechanisms significantly enhances the long-term self-cleaning efficiency of TiO_2 -based systems (Shareef et al. 2026).

Titanium dioxide (TiO_2) is one of the most extensively studied photocatalysts due to its chemical stability, non-

toxicity, cost-effectiveness, and strong oxidative capability under ultraviolet (UV) irradiation (Zhang et al. 2024; Liang et al. 2025). When TiO_2 is exposed to photons with energy equal to or greater than its band gap, electrons are excited from the valence band to the conduction band, resulting in the formation of electron-hole (e^-/h^+) pairs that initiate a series of redox reactions on the catalyst surface (Wang et al. 2026). These photogenerated charge carriers subsequently interact with adsorbed oxygen and water molecules, leading to the formation of various reactive oxygen species (ROS), including hydroxyl radicals ($\bullet\text{OH}$), superoxide radicals ($\bullet\text{O}_2^-$), singlet oxygen ($^1\text{O}_2$), and hydrogen peroxide (H_2O_2). These species play a critical role in degrading organic pollutants. Among them, hydroxyl radicals are highly reactive and non-selective, whereas non-radical species, such as singlet oxygen, possess longer lifetimes and can promote more selective oxidation pathways under specific conditions (Wang et al. 2026; Qi et al. 2025). The generation, distribution, and reactivity of these ROS are strongly governed by the physicochemical properties of the photocatalyst, including its band structure, charge-carrier dynamics, and surface characteristics, which ultimately determine the overall photocatalytic efficiency (Liang et al. 2025; Majeed et al. 2025).

In recent years, the integration of TiO_2 into cementitious materials has attracted significant attention as an effective strategy for developing multifunctional construction materials with self-cleaning and air-purifying capabilities. These systems enable the degradation of atmospheric pollutants, such as nitrogen oxides (NO_x), and organic contaminants directly on building surfaces, thereby improving urban air quality (Bahşi et al. 2022; Yan et al. 2025). The photocatalytic performance of such materials is primarily governed by material-specific parameters, including TiO_2 dosage, particle size, dispersion, and the microstructural characteristics of the cement matrix, which control the exposure of active sites and the accessibility of pollutants (Mostafa et al. 2023). The dosage-dependent influence of nano- TiO_2 on porosity and mechanical response has also been demonstrated in cementitious composites (Srivastava et al. 2025). Furthermore, although increasing TiO_2 content generally enhances photocatalytic activity, an optimum threshold is often observed, beyond which performance may decline due to particle agglomeration, light scattering, and reduced UV penetration (Adebanjo et al. 2024). In addition to dosage, the spatial distribution of TiO_2 within the cementitious matrix and the porosity of the material play a critical role in facilitating mass transfer and enhancing pollutant-surface interactions (Zhang et al. 2024; Hamidi and Aslani 2019). These findings emphasize that optimizing both material composition and microstructural design is essential to achieving efficient, sustainable photocatalytic performance in cement-based systems.

In addition to TiO_2 dosage and material composition, the method of photocatalyst incorporation and the thickness of the applied layer are crucial factors in determining the overall photocatalytic performance of cementitious systems. Previous studies have shown that surface-based applications, in which the photocatalyst is concen-

trated near or on the exposed surface, provide significantly higher utilization efficiency than bulk incorporation methods, primarily due to improved light accessibility and direct contact with pollutants (Si et al. 2021; Seremak et al. 2023). Furthermore, the design of multi-layer systems has attracted increasing attention, as double-layer configurations with a TiO₂-rich top layer have been reported to enhance photocatalytic activity by maximizing active surface exposure while maintaining the structural integrity of the substrate (Jin et al. 2023). In this context, coating thickness is also a critical parameter influencing photocatalytic efficiency. While increasing the thickness of TiO₂-based films or coatings can enhance photocatalytic activity by providing more active sites, excessive thickness can reduce light penetration and hinder efficient charge transfer, thereby limiting overall performance (Gómez-Monsivais et al. 2026). These findings indicate that achieving an optimal balance between coating thickness, catalyst distribution, and layer configuration is essential for maximizing photocatalytic efficiency. Despite these advances, a systematic comparison of coating configurations (e.g., single-versus double-layer systems) and varying TiO₂ dosages under controlled, comparable conditions remains limited in the existing literature, particularly for cement-based photocatalytic surfaces. In light of the above considerations, this study aims to provide a systematic, application-oriented evaluation of TiO₂-based photocatalytic coatings applied to cementitious surfaces by examining the combined effects of coating configuration and TiO₂ dosage under controlled experimental conditions. Unlike many previous studies that focus on either material composition or overall photocatalytic performance, this work adopts a comparative approach, investigating single- and double-layer coating systems with varying TiO₂ contents. Furthermore, a cavity-type experimental configuration was employed to confine the pollutant solution within a defined volume in direct contact with the coated surface, enabling a more controlled assessment of mass transfer, interfacial interactions, and diffusion limitations. This approach allows a more realistic representation of practical application conditions while maintaining experimental consistency. The photocatalytic performance of the developed systems was evaluated using methylene blue (MB) and Rhodamine B (Rh-B) degradation, and the results were analyzed through pseudo-first-order kinetic modeling to elucidate the influence of coating design and TiO₂ loading on degradation behavior.

2. Materials and Methods

2.1. Materials

In this study, a pozzolanic cement conforming to TS EN 197-1 standard, classified as CEM IV/B (P-Q) 32.5 N, was used for specimen production. The cement consisted of Portland clinker, trass, calcined pozzolan, and gypsum, and its chemical and physical properties are presented in Table 1. In this study, type F fly ash was used. The chemical properties of the fly ash produced at the Tunçbilek Thermal Power Plant are given in Table 2. Natural aggregates were used as fine and coarse fractions. The fine aggregate (0–4 mm) and coarse aggregate (4–22.4 mm) were obtained from the Sakarya River located in Bilecik, Türkiye. The physical properties and granulometric distribution of the aggregates are given in Table 3. A polycarboxylate ether-based high-range water-reducing admixture (BASF MasterGlenium 51) was used as a superplasticizer to improve workability. The properties of the superplasticizer are summarized in Table 4. Tap water was used for mixing, and its chemical composition is provided in Table 5. The concrete mix proportions and water-to-cement (w/c) ratio are presented in Table 6, with a w/c ratio of 0.35 used in all mixtures. Cubic specimens measuring 15 × 15 × 15 cm were produced. To simulate a confined reaction environment, hollow cavities 5 cm in diameter and 10 cm high were created within the specimens during casting. All mixtures were prepared in accordance with the procedures specified in TS EN 196-1. The mortar mixtures were prepared using a laboratory pan mixer to ensure uniform and homogeneous mixing of all constituents, including cement, aggregates, water, and admixtures. After casting, the specimens were cured in water at 20 ± 2 °C for 28 days. Following curing, the specimens were dried under laboratory conditions. A total of 48 specimens were prepared for the experimental study. Each condition was evaluated in triplicate to ensure repeatability. The upper surfaces of the specimens were partially treated with MB and Rh-B solutions, while the internal cavities were filled with the same solutions to enable controlled photocatalytic testing. The specimen configuration was illustrated in Fig. 1(a). The TiO₂ used in this study was supplied by Detsan Chemical Company. The photocatalyst had a specific surface area of 85 ± 10 m²/g, an average particle size of 15–25 nm, and anatase phase purity ≥ 99%.

Table 1. Physical and chemical properties of cement.

Physical properties			Chemical content (%)			
Density (g/cm ³)	2.85		CaO	32.0–42.0	MgO	1.3–2.0
Blaine (m ² /g)	4800–5300		SiO ₂	33.0–37.0	Na ₂ O	0.8–1.2
Compressive Strength (MPa)	2.day 9.5–13.0	28.day 34.0–38.0	Al ₂ O ₃	8.0–11.0	SO ₃	2.7–3.2
Setting time (min)	Initial 180–220	Final 230–280	Fe ₂ O ₃	2.6–3.0	K ₂ O	0.8–1.10
			LOI	3.0–5.0	Cl	0.02–0.05

Table 2. Chemical properties of fly ash.

Chemical content (%)			
SiO ₂	58.59	CaO	4.43
Al ₂ O ₃	21.89	MgO	1.41
Fe ₂ O ₃	9.31	LOI	0.59
S+F+A	89.79	Pozzolanic	89.40

Table 3. Physical properties of aggregates.

Sieve size (mm)	22.4	16	11.2	8	4	2	1	0.5	0.25	0.125
Granulometry (%)	100.0	88.4	73.8	63.5	44.6	32.8	24.7	16.3	11.2	7.4
Unit weight (kg/dm ³)	0–4 mm			1.44						
	4–22.4 mm			1.56						

Table 4. Properties of NGSP.

Density (kg/dm ³)	Alkaline (%)	Chlorine (%)	Structure
1.082–1.142	< 3	< 0.1	poly carboxylic ether

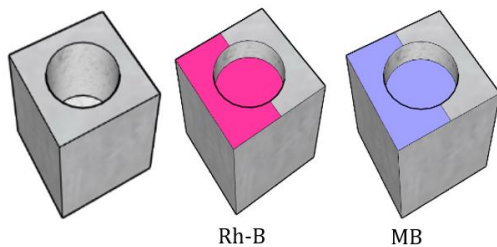
Table 5. Chemical analysis of tap water.

pH	7.28	SO ₄ [*]	70	Cl [*]	43
Hardness (Fr°)	32	NO ₃ [*]	7.5	NH ₄ [*]	<0.05
Turbidity (NTU)	0.10	Al [*]	<0.012	Mn [*]	<0.0007
Conductivity (µS/cm)	664	Fe [*]	<0.0075	*mg/L	

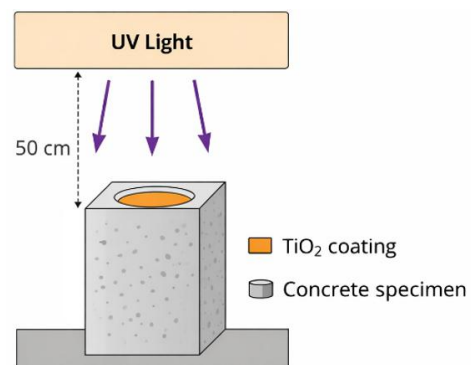
Table 6. Mix proportions.

Cement (kg/m ³)	water /cement	fine aggregate /cement	coarse aggregate /cement	NGSP / cement
300	0.35	2.96	3.68	0.01

(a)



(b)

**Fig. 1.** (a) Schematic representation of the concrete specimens and coating configurations; (b) Schematic illustration of the UV-assisted photocatalytic experimental setup.

2.2. Photocatalytic test

The photocatalytic efficiency of the TiO₂-coated concrete samples was assessed by measuring the breakdown of MB and Rh-B solutions upon exposure to ultra-

violet (UV) light. The experiments were structured to maintain controlled interaction between the pollutant solution and the photocatalytic surface. Before irradiation, solutions of MB and Rh-B were prepared at specific concentrations and placed into cavities created in the

concrete samples. In addition to the cavity setup, the upper surfaces of the specimens were exposed to MB and Rh-B solutions, while the internal cavities were filled with the corresponding dye solutions to ensure consistent contact between the pollutant and the photocatalytic surface and to enable the simultaneous evaluation of surface-based and confined reaction scenarios. A UV light source was utilized to trigger photocatalytic reactions. The lamp emitted light in the UVA (315–400 nm) and UVB (280–315 nm) spectra, with power outputs of 13.6 W and 3.0 W, respectively. The light source was set approximately 50 cm above the specimen surface to ensure uniform irradiation. The irradiance at the sample surface was approximately 0.5–0.8 W/m² at a distance of 50 cm, based on the lamp specifications and the inverse-square law governing irradiance as a function of distance. All tests were performed under laboratory conditions at room temperature, in an environment shielded from natural light to prevent external interference. A schematic representation of the experimental setup, including the UV light source and specimen positioning, is shown in Fig. 1(b). At predetermined time intervals (0, 12, 24, 36, and 48 hours), samples were collected from the cavity region to monitor the degradation process. The concentration changes of MB and Rh-B were monitored using a Thermo Scientific GENESYS 150 UV-Vis spectrophotometer. Absorbance (A) measurements were performed at the characteristic wavelengths of 664 nm for MB and 553 nm for Rh-B. Quartz cuvettes (10 mm path length) were used for all measurements. Distilled water was employed as the blank solution for baseline correction. Each measurement was conducted in triplicate to ensure repeatability and reliability of the results. Calibration curves were established for both dyes prior to analysis, and concentrations were determined from the corresponding absorbance–concentration relationships.

The effectiveness of photocatalytic degradation was determined by the reduction in concentration relative to the initial value. To quantitatively evaluate the reaction kinetics, the degradation data were examined using a pseudo-first-order kinetic model, which is widely used in heterogeneous photocatalytic systems. The kinetic rate constants were derived from the linear relationship between $\ln(C_0/C_t)$ and irradiation time, where C_0 denotes the initial concentration, and C_t denotes the concentration at a given time. A total of 48 independent specimens were prepared and tested, including four TiO₂ concentrations (5, 10, 15, and 20 wt%), two coating configurations (single-layer and double-layer), two dye systems (MB and Rh-B), and three replicates per condition to ensure experimental repeatability. Error analysis was conducted based on the standard deviation of triplicate measurements ($n = 3$). The low variability observed in the data indicates a high level of experimental precision and reliability.

2.3. Preparation of TiO₂ coatings

TiO₂ coatings were applied to the surfaces of the cured concrete samples using a manual spray deposition method. Before applying the coatings, the specimens' surfaces were cleaned and dried in a laboratory setting to remove dust and loosely bound particles, ensuring

proper adhesion between the coating and the substrate. TiO₂ suspensions were prepared by mixing the photocatalyst powder in distilled water at different weight percentages (5, 10, 15, and 20 wt%). The selected TiO₂ loading range (5–20 wt%) was determined based on literature values for photocatalytic coatings applied to cementitious materials. This range was chosen to ensure sufficient photocatalytic activity while avoiding excessive particle agglomeration, light shielding, and reduced UV penetration at higher loadings. Additionally, this range allows the identification of an optimal catalyst dosage and provides a practical balance between performance and material applicability (Hamidi and Aslani 2019; Adebajo et al. 2024).

The suspensions were mixed thoroughly using magnetic stirring before application. The coating process employed a handheld spray device with a small reservoir and a standard nozzle. The TiO₂ suspension was sprayed onto the specimen surfaces from approximately 20 cm away to achieve uniform coverage. The spraying was performed manually under controlled laboratory conditions, with consistent application time and the same preparation methods for all specimens. Two different coating setups were tested: single-layer and double-layer systems. In the single-layer setup, one spray application was applied to the concrete surface. In the double-layer setup, the first layer was applied and left to dry at ambient laboratory conditions (25 ± 2 °C) for about 2 hours before the second layer was applied. After completing the coating process, all specimens were kept under the same laboratory conditions (25 ± 2 °C) for 24 hours.

3. Result and Discussion

The photocatalytic performance of TiO₂-coated concrete surfaces was evaluated by measuring the degradation of MB and Rh-B under UV irradiation (Figs. 2 and 3). A continuous decrease in absorbance values at the characteristic wavelengths of MB and Rh-B was observed over time, indicating effective photocatalytic degradation of the dye molecules. This reduction in absorbance reflects the progressive breakdown of various structures within the dye molecules, confirming the catalytic activity of TiO₂ under UV irradiation.

The observed degradation behavior can be attributed to the generation of reactive oxygen species (ROS), such as hydroxyl radicals ($\bullet\text{OH}$) and superoxide radicals ($\bullet\text{O}_2^-$), which are produced upon UV-induced excitation of TiO₂ (Singh and Soundappan 2026). These highly reactive species attack the dye molecules adsorbed to the concrete surface, leading to bond cleavage and subsequent transformation into smaller, less harmful compounds (Chong et al. 2010). The consistent decrease in absorbance over time during irradiation demonstrates that the photocatalytic process is sustained and effective under the applied experimental conditions. Furthermore, the results confirm that the TiO₂-coated surfaces exhibit clear self-cleaning functionality, with degradation of organic pollutants occurring directly at the material surface without the need for external chemical agents. This behavior is consistent with previous studies on photocatalytic construction materials (Chen and Poon 2009).

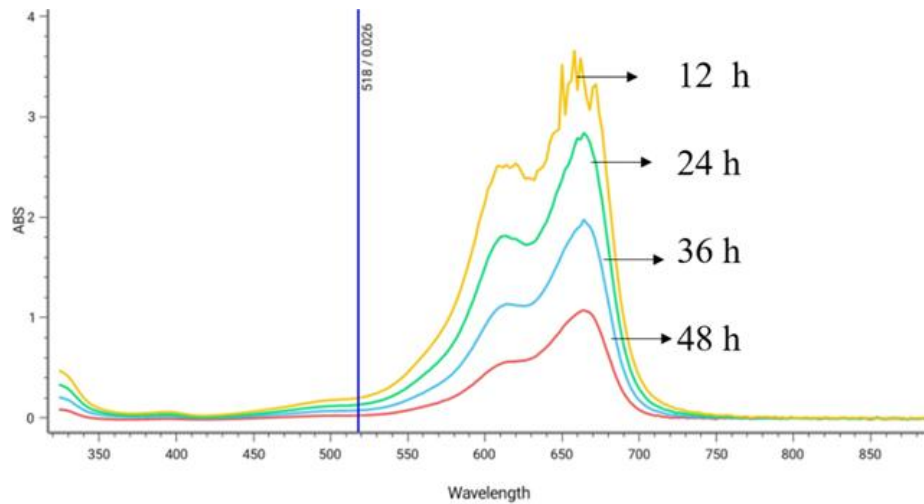


Fig. 2. The effect of exposure time on the photocatalytic activity of double layers of TiO₂-coated self-cleaning concrete (MB).

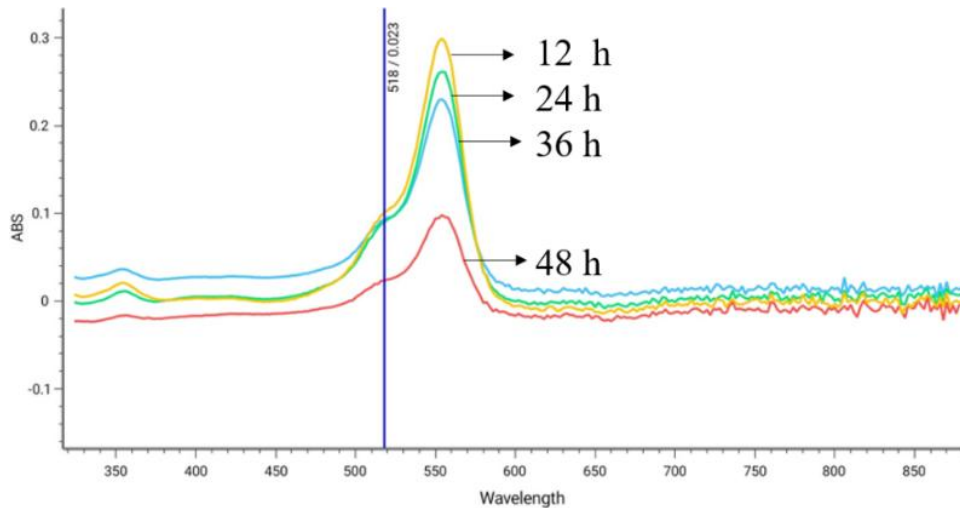


Fig. 3. The effect of exposure time on the photocatalytic activity of double layers of TiO₂-coated self-cleaning concrete (Rh-B).

The normalized concentration profiles (C_t/C_0) of MB and Rh-B in contact with single- and double-layer TiO₂-coated concrete surfaces are presented in Fig. 4. A clear decreasing trend in C_t/C_0 values was observed over time for all samples, indicating progressive photocatalytic degradation of both dyes. However, the extent of degradation strongly depended on both TiO₂ dosage and coating configuration.

For single-layer applications, increasing the TiO₂ content from 5% to 20 wt% resulted in a noticeable reduction in normalized concentrations for both MB and Rh-B, demonstrating that higher catalyst loading enhances photocatalytic activity by increasing the availability of active sites. In contrast, double-layer systems exhibited significantly lower final C_t/C_0 values than single-layer systems at the same TiO₂ dosage. This trend is consistent with previous studies, which show that increasing catalyst loading improves degradation efficiency up to an optimum level due to increased active sites and enhanced generation of reactive oxygen species (ROS) (Atalı et al. 2024; Bendjama and Hamdaoui 2025). This enhancement is attributed to higher surface exposure of TiO₂, im-

proved light absorption, and more efficient charge-carrier utilization, leading to increased ROS generation and faster degradation kinetics (Bendjama and Hamdaoui 2025; Ahouari et al. 2024). The difference between single- and double-layer configurations was more pronounced at higher TiO₂ loadings, suggesting that coating architecture plays a critical role in determining photocatalytic efficiency. In double-layer systems, the TiO₂-rich top layer likely provides improved exposure to UV irradiation and more effective interaction with pollutant molecules, leading to accelerated degradation kinetics. Moreover, photocatalytic degradation of organic dyes generally follows pseudo-first-order kinetics, with the apparent rate constant increasing as catalyst accessibility and surface activity improve (Sharma et al. 2023; Gao et al. 2024). Additionally, the improved distribution of TiO₂ at the surface enhances mass transfer and reduces diffusion limitations, further contributing to the observed performance improvement. Such enhancements facilitate the transport of pollutants to active sites and improve overall reaction efficiency in heterogeneous photocatalytic systems (Bendjama and Hamdaoui 2025).

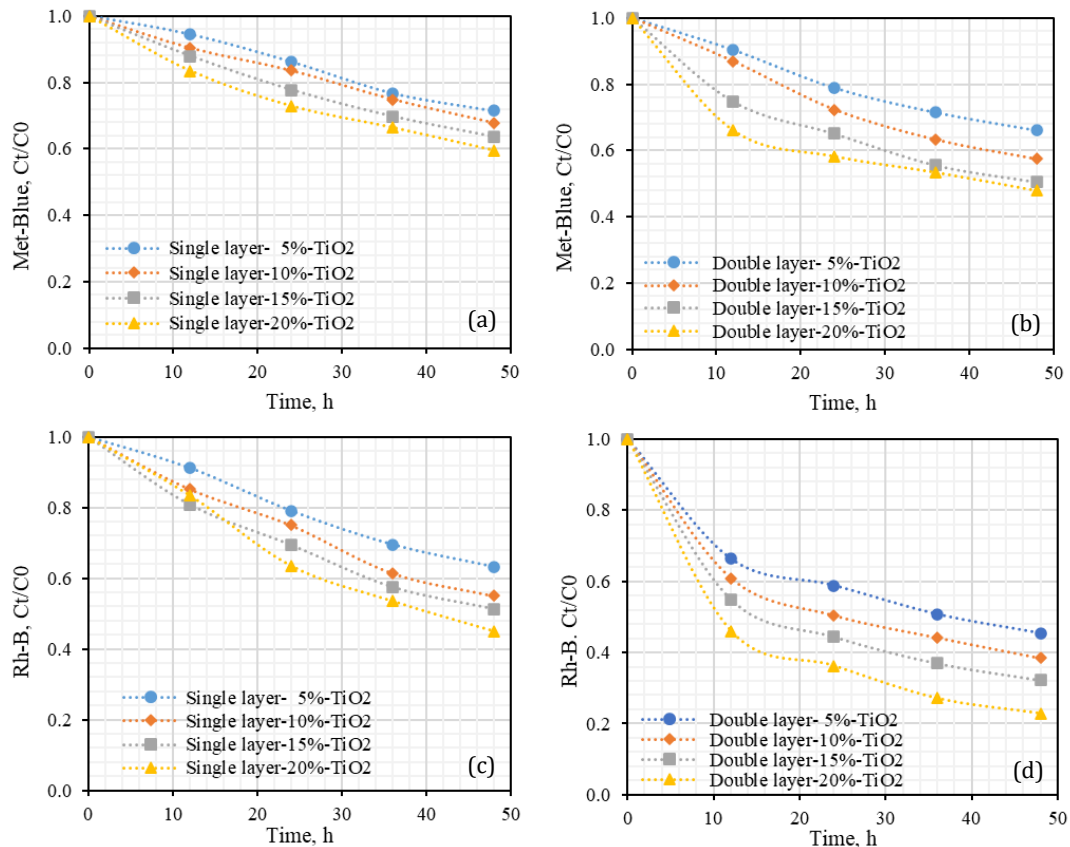


Fig. 4. Effect of single- and double-layer 5, 10, 15, and 20% TiO₂-coated self-cleaning concrete on the normalized concentration (C_t/C_0) concentration of:

(a) Single-layer TiO₂ in MB; (b) Double-layer TiO₂ in MB; (c) Single-layer TiO₂ in Rh-B; (d) Double-layer TiO₂ in Rh-B.

As shown in Fig. 4(a,b), for MB, the normalized concentration decreased from 1.00 to 0.71 for the 5% single-layer TiO₂-coated surface and further to 0.60 for the 20% single-layer coating, confirming the positive effect of increasing TiO₂ dosage. In the double-layer configuration, the normalized concentration decreased more significantly, from 1.00 to 0.60 at 5% TiO₂ and to 0.40 at 20% TiO₂, demonstrating enhanced photocatalytic performance due to increased surface-active sites and improved catalyst distribution.

A similar trend was observed for Rh-B, with the normalized concentration decreasing from 0.63 to 0.45 as the TiO₂ content increased in single-layer coatings (Fig. 4(c)). In double-layer systems (Fig. 4(d)), the normalized concentration decreased more sharply, reaching 0.45 at 5% TiO₂ and 0.23 at 20% TiO₂. These results clearly indicate that both increasing TiO₂ loading and adopting a double-layer coating strategy significantly enhance photocatalytic degradation efficiency. A similar increase in self-cleaning performance with increasing TiO₂ content has also been reported under artificial-light exposure (Ünal and Canbaz 2025a). Overall, the results demonstrate that both TiO₂ dosage and coating configuration are key parameters governing photocatalytic efficiency, with double-layer coatings at higher TiO₂ concentrations exhibiting the most effective degradation behavior for both MB and Rh-B solutions. This behavior indicates an optimal TiO₂ loading, beyond which further increases do not yield proportional improvements in photocatalytic performance. Furthermore, the differences observed between MB and Rh-B degradation behaviors can be at-

tributed to variations in molecular structure, adsorption characteristics, and interaction with reactive oxygen species.

Fig. 5(a-d) shows the degradation efficiency of MB and Rh-B solutions in contact with single-layer and double-layer TiO₂-coated surfaces at different concentrations under UV irradiation. The degradation efficiency is calculated by Eq. (1), where A_0 and A_t represent the initial and residual dye absorbance at time 0 and t , respectively.

$$\text{Degradation efficiency (\%)} = \frac{(A_0 - A_t)}{A_0} \times 100 \quad (1)$$

Figs. 5(c) and 5(d) were used to evaluate the photocatalytic degradation of concrete surfaces coated with single- and double-layer TiO₂ against Rh-B. As observed in the experimental results, the degradation efficiency of both dyes increased progressively with irradiation time, a typical behavior of photocatalytic systems driven by the continuous generation of reactive species under UV exposure (Saefumillah et al. 2025). The results showed that the double-layer TiO₂-coated surface had significantly higher degradation efficiency than the single-layer application. The 20% double-layered surface achieved 77.4% Rh-B degradation within 48 hours, while the single-layered surface achieved only 55% photocatalytic degradation efficiency. Similar results were obtained with MB. The double-layer coating and the high-TiO₂-surface showed the highest MB photocatalytic degradation after 48 hours. On the 20% single-layer-coated surface, MB degradation was 40.50%, whereas on the double-layer surface, the efficiency was 52.12%.

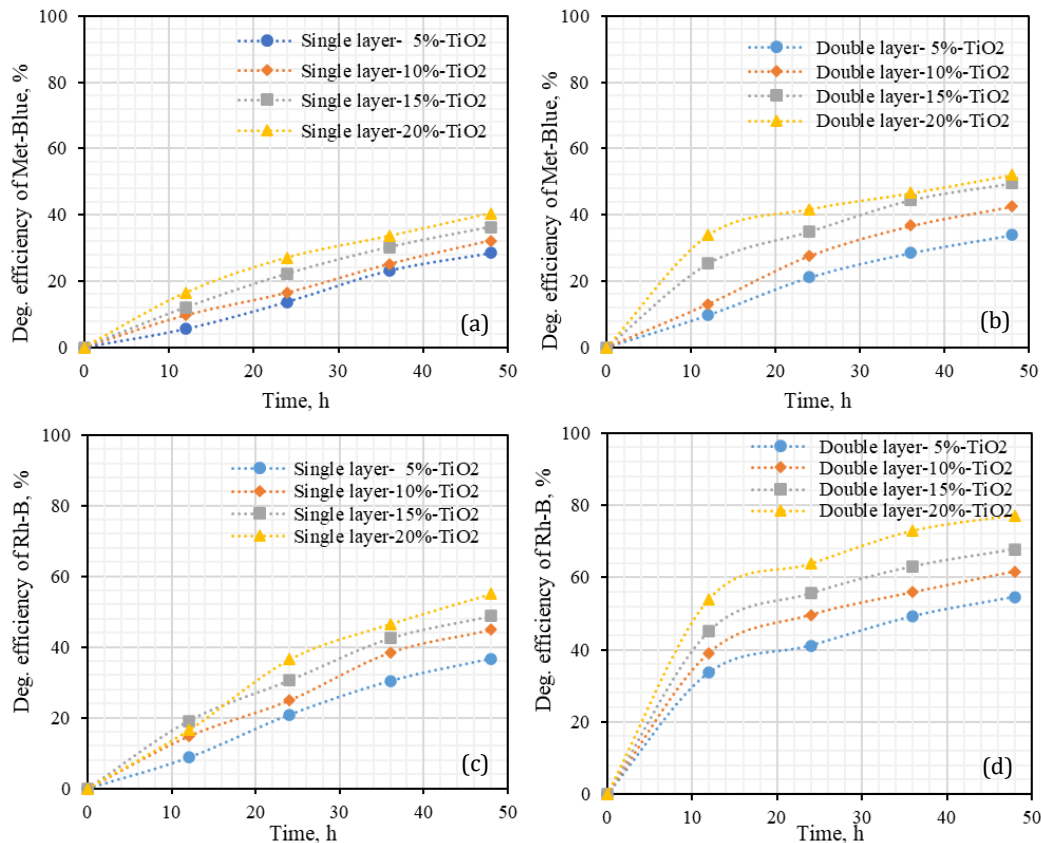


Fig. 5. The degradation efficiency of MB and Rh-B solutions with single- and double-layer 5, 10, 15, and 20% TiO₂-coated self-cleaning concrete: (a) Single-layer TiO₂ in MB; (b) Double-layer TiO₂ in MB; (c) Single-layer TiO₂ in Rh-B; (d) Double-layer TiO₂ in Rh-B.

This improvement is due to the increased number of active photocatalytic sites and better surface exposure in double-layer systems, which enhance light absorption and produce more reactive oxygen species (ROS), ultimately speeding up the degradation reaction (Adaileh et al. 2025; Saefumillah et al. 2025). Under UV light, TiO₂ produces active oxygen species that react with Rh-B molecules adsorbed on the cement surface, thereby mediating photocatalytic degradation. These reactive species, especially hydroxyl radicals ($\bullet\text{OH}$), superoxide radicals ($\text{O}_2\bullet^-$), and photogenerated holes (h^+), control the photocatalytic degradation process as photogenerated charge carriers react with adsorbed oxygen and water to produce highly oxidative species capable of breaking down complex dye molecules into smaller intermediates and ultimately mineralized products. (Ramesh et al. 2024).

The degradation mechanism of Rh-B involves a series of photoinduced reactions, including chromophore cleavage, N-deethylation/demethylation, and subsequent mineralization into simpler inorganic compounds, as extensively reported for TiO₂-based photocatalytic systems. (Sleiman et al. 2007). A similar degradation pathway can be assumed for MB, in which ROS attack the dye's conjugated structure, leading to stepwise degradation and eventual mineralization (Saefumillah et al. 2025).

Increasing the concentration and number of layers of TiO₂ molecules coated on the surface results in more active TiO₂ molecules per unit surface under UV light, forming more oxygen species and degrading more MB

and Rh-B. However, it is well known that excessive catalyst loading can lead to particle agglomeration and light scattering, which can hinder effective photon penetration and reduce photocatalytic efficiency beyond an optimum level, as morphological characteristics and interfacial interactions significantly influence charge transfer and catalytic performance (Yuqin Wei et al. 2023a).

$$\ln \frac{C_0}{C_t} = \ln \frac{A_0}{A_t} = K_{\text{app}} \cdot t \quad (2)$$

The rate constant (K_{app}) for the photocatalytic reaction was evaluated using a pseudo-first-order kinetic model, as described by Eq. (2). This model is widely applied in heterogeneous photocatalysis and reflects the proportional relationship between degradation rate and pollutant concentration at low concentrations (Kadem et al. 2025). The kinetic model is expressed as $\ln(C_0/C_t) = K_{\text{app}} \cdot t$, where C_0 and C_t represent the initial and instantaneous concentrations, respectively, and K_{app} is the apparent rate constant. C_t and A_t represent the pollutant concentration and absorbance at time t , respectively, while C_0 and A_0 denote the initial concentration and absorbance at the beginning of the experiment. The kinetic properties are displayed in Table 7. These parameters were used to evaluate the photocatalytic degradation behavior and kinetic performance of TiO₂-coated concrete surfaces. Figure 6 shows a strong linear correlation between $\ln(C_0/C_t)$ and time for both pollutants, confirming that the photocatalytic degradation process follows pseudo-first-order kinetics, a common behavior in TiO₂-

based photocatalytic systems at low pollutant concentrations.

Error analysis was conducted based on the standard deviation of triplicate measurements ($n = 3$). The exper-

imental error associated with concentration measurements was relatively low, with standard deviations typically ranging from ± 0.3 to ± 0.8 ppm, indicating high precision in the UV-Vis measurements.

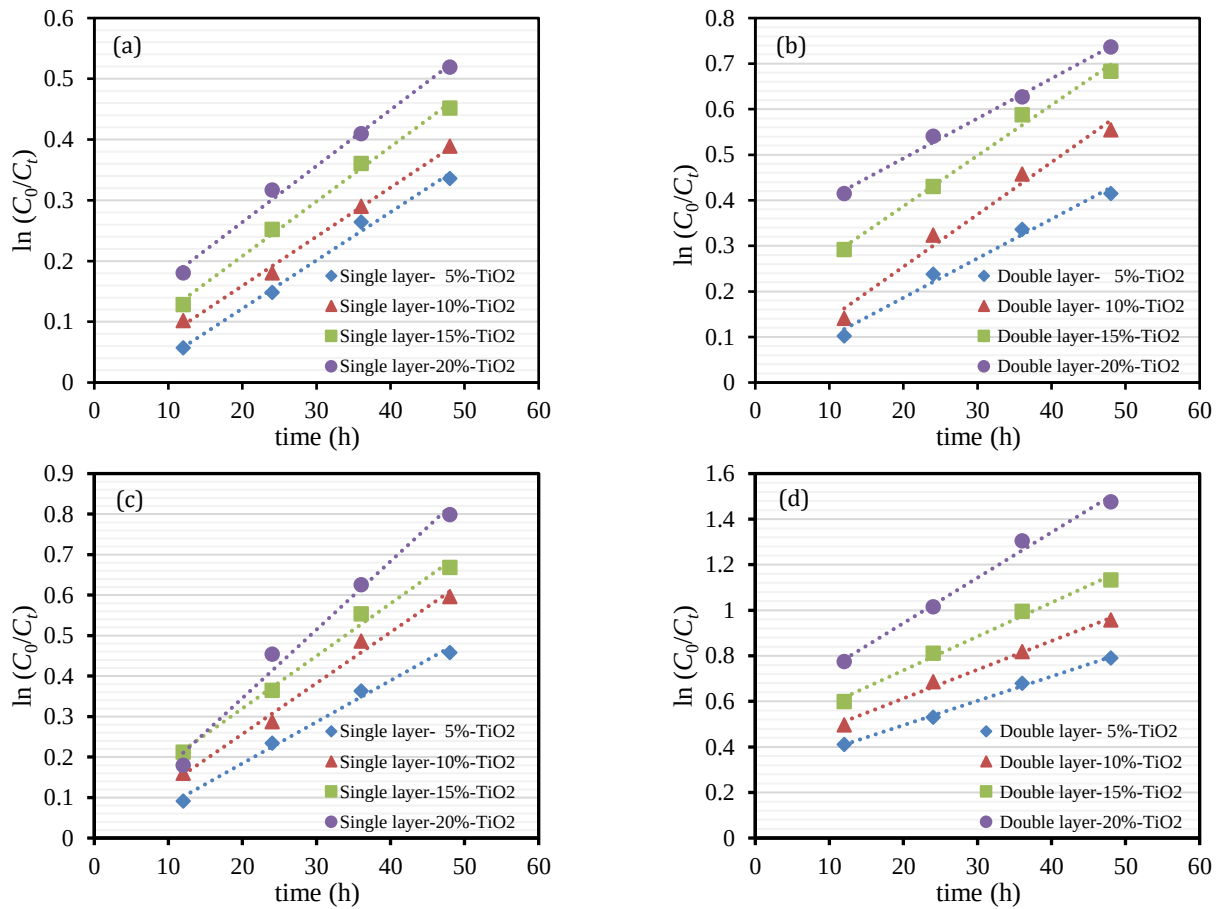


Fig. 6. The pseudo-first-order kinetics model graphs

for single- and double-layer TiO₂-coated self-cleaning concrete at 5, 10, 15, and 20%:

(a) Single-layer TiO₂ in MB; (b) Double-layer TiO₂ in MB; (c) Single-layer TiO₂ in Rh-B; (d) Double-layer TiO₂ in Rh-B.

Fig. 6 shows a strong linear correlation between $\ln(C_0/C_t)$ and time for both pollutants, indicating that the photocatalytic degradation follows pseudo-first-order kinetics, a common behavior in TiO₂-based photocatalytic systems at low pollutant concentrations (Sharma et al. 2023; Kadem et al. 2025). The photocatalytic reaction of coated surfaces against MB showed increases in the K_{app} value of 16.46% and 34.88% for single- and double-layer applications, respectively, when the TiO₂ concentration in the coatings was increased from 5% to 20% wt%. The rate of photocatalytic degradation of concrete surfaces against Rh-B was significantly influenced by TiO₂ concentration. An increase in TiO₂ concentration from 5% to 20% wt% resulted in degradation rate increases of 85.98% and 65.69% for single- and double-layer applications, respectively. These results clearly show that increasing the TiO₂ loading increases the apparent reaction rate by increasing the number of active sites and enhancing surface-pollutant interactions (Adaileh et al. 2025). These surface hydroxyl groups play a crucial role in photocatalytic reactions by acting as active sites for the generation of hydroxyl radicals ($\bullet OH$), while photogenerated electrons reduce oxygen to form

superoxide radicals ($O_2^{\bullet -}$). These reactive oxygen species are primarily responsible for attacking dye molecules, inducing their oxidative degradation, and promoting their mineralization (Bhattacharyya et al. 2025). More TiO₂ particles increase the density of surface hydroxyl groups, which serve as active sites for photocatalytic reactions, facilitating the generation of reactive oxygen species. These hydroxyl groups enhance interfacial charge transfer processes and promote the formation of highly oxidative species responsible for dye degradation. Additionally, surface defects and oxygen vacancies improve charge separation efficiency by trapping photo-generated electrons and suppressing electron-hole recombination (Yubin Wei et al. 2023b). Consequently, the lifetimes and availability of charge carriers are extended, leading to higher apparent rate constants (K_{app}) and enhanced photocatalytic degradation performance, particularly in double-layer systems with higher TiO₂ loading (Xing et al. 2023). These findings clearly demonstrate that the synergistic effect of increased TiO₂ loading and optimized coating architecture significantly enhances both the kinetics and overall efficiency of photocatalytic degradation processes.

Similarly, the standard deviation of the apparent rate constants (K_{app}) was ± 0.0001 to ± 0.0005 h^{-1} , demonstrating low variability and high consistency among replicate measurements. The statistical reliability of the ki-

netic analysis was confirmed by low standard deviation values and high correlation coefficients ($R^2 \approx 0.98$ – 0.99), indicating strong agreement with the pseudo-first-order kinetic model.

Table 7. Pseudo-first-order kinetic model constants (K_{app}) of single- and double-layer 5, 10, 15, and 20 wt% TiO_2 -coated self-cleaning concrete for MB and Rh-B. Values are reported as mean $\pm$ standard deviation ($n = 3$).

	MB		Rh-B	
	$K_{app} (h^{-1})$	R^2	$K_{app} (h^{-1})$	R^2
Single coated				
%5 TiO_2	0.0080 ± 0.0001	0.993	0.0103 ± 0.0002	0.993
%10 TiO_2	0.0081 ± 0.0001	0.996	0.0126 ± 0.0002	0.988
%15 TiO_2	0.0091 ± 0.0001	0.996	0.0130 ± 0.0002	0.992
%20 TiO_2	0.0093 ± 0.0002	0.995	0.0169 ± 0.0003	0.985
Double coated				
%5 TiO_2	0.0086 ± 0.0001	0.985	0.0107 ± 0.0001	0.997
%10 TiO_2	0.0115 ± 0.0001	0.98	0.0126 ± 0.0003	0.992
%15 TiO_2	0.0111 ± 0.0001	0.991	0.0149 ± 0.0002	0.992
%20 TiO_2	0.0088 ± 0.0001	0.995	0.0200 ± 0.0005	0.991

K_{app} values were calculated from the linear regression of $\ln(C_0/C_t)$ versus time. All measurements were performed in triplicate ($n = 3$), and the results are presented as mean $\pm$ standard deviation to ensure repeatability and statistical reliability. The low standard deviation values and high R^2 coefficients confirm the statistical reliability and repeatability of the kinetic analysis.

4. Conclusions

In this study, the photocatalytic performance of TiO_2 -coated concrete surfaces was systematically evaluated with respect to coating configuration and catalyst loading using MB and Rh-B as model pollutants. The results clearly demonstrated that both TiO_2 dosage and coating architecture play a decisive role in governing photocatalytic efficiency and degradation kinetics.

The normalized concentration profiles and degradation efficiency results revealed that increasing TiO_2 content from 5% to 20 wt% significantly enhanced photocatalytic performance. In particular, double-layer coatings consistently exhibited superior activity compared to single-layer systems, achieving up to 77.4% degradation for Rh-B and 52.12% for MB after 48 h of UV irradiation. This improvement was attributed to the higher surface availability of TiO_2 , improved light utilization, and enhanced interaction between the catalyst surface and pollutant molecules.

Kinetic analysis confirmed that the degradation process follows pseudo-first-order kinetics, with higher apparent rate constants (K_{app}) observed at increased TiO_2 loadings and in double-layer configurations. The enhancement in reaction rates was directly associated with increased active site density and improved charge-carrier dynamics.

From a mechanistic perspective, the improved performance of TiO_2 -coated surfaces was attributed to the generation of reactive oxygen species (ROS), such as hydroxyl and superoxide radicals, and to enhanced charge separation facilitated by surface hydroxyl groups and oxygen vacancies. These factors collectively contributed to reduced electron-hole recombination and increased availability of reactive species responsible for pollutant degradation.

This study primarily focuses on the photocatalytic performance of TiO_2 -coated concrete surfaces rather than the mechanical or physical characterization of the concrete substrate. Therefore, the results should be interpreted in the context of surface functionality.

Overall, the findings demonstrate that optimizing both TiO_2 loading and coating configuration provides an effective strategy for improving the photocatalytic efficiency of cement-based materials. These findings are consistent with previous studies (Hamidi and Aslani 2019; Maury-Ramírez et al. 2024) demonstrating that increasing TiO_2 loading and optimizing coating configuration significantly enhance photocatalytic efficiency by improving active-site accessibility, light utilization, and charge-carrier dynamics in cementitious materials. The developed double-layer TiO_2 coating system shows strong potential for practical applications in self-cleaning and environmentally responsive construction materials, particularly for the removal of organic pollutants in urban environments.

Future studies should focus on optimizing TiO_2 loading, evaluating coating thickness, and investigating long-term durability, adhesion strength, and environmental stability under real service conditions to enable practical implementation of photocatalytic concrete systems.

Acknowledgements

None declared.

Funding

The authors received no financial support for the research, authorship, and/or publication of this manuscript.

Conflict of Interest

The authors declare no potential conflicts of interest with respect to the research, authorship, and/or publication of this manuscript.

Data Availability

The datasets generated and/or analyzed during the current study are not publicly available but are available from the corresponding author upon reasonable request.

AI Assistance

No AI-based tools were used in the preparation of this manuscript.

Author Contributions

All authors made substantial contributions to the conception and design of the study, acquisition of data, analysis and interpretation of data; drafted or critically revised the manuscript for important intellectual content; and approved the final version to be published.

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