

## Effect of SiO<sub>2</sub> derived natural bamboo leaf ash on paving blocks to reduce pollutants

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### Abstract

The annual increase in traffic volume contributes approximately 71% of nitrogen oxide (NO<sub>x</sub>) emissions and 15% of sulfur oxide (SO<sub>x</sub>) emissions, along with other air pollutants. Paving blocks with a TiO<sub>2</sub>-SiO<sub>2</sub> photocatalyst layer offer a promising solution for mitigating these pollutants by enhancing the adsorption and degradation of organic compounds. Since, conventional SiO<sub>2</sub> sources are typically derived from nonrenewable materials, bamboo leaf ash presents a renewable and environmentally friendly alternative. This study aimed to identify the optimal TiO<sub>2</sub>-SiO<sub>2</sub> composition and evaluate the adsorption efficiency and physical properties of paving blocks incorporating SiO<sub>2</sub> derived from bamboo leaf ash. TiO<sub>2</sub>-SiO<sub>2</sub> composites were synthesized with mass ratios of 2:0.5 (K1), 2:1 (K2), 0.5:2 (K3), 1:2 (K4), and 2:2 (K5). Paving blocks were fabricated by mixing cement, sand, water, and the TiO<sub>2</sub>-SiO<sub>2</sub> composite in a ratio of 1:4:0.5:1, then molded into 20 × 10 × 6 cm blocks. Each block featured a 5 mm photocatalyst layer atop a 55 mm base layer. Characterization included scanning electron microscopy (SEM), compressive strength testing, water absorption analysis, and UV-Vis spectrophotometry. The optimal performance was found in sample K5 (TiO<sub>2</sub>:SiO<sub>2</sub> = 2:2), which demonstrated a pollutant adsorption value of 0.086 ppm, water absorption of 8.47%, and a compressive strength of 14.58 MPa. These results indicate a uniform distribution of the TiO<sub>2</sub>-SiO<sub>2</sub> composite, enhancing the efficiency of pollutant removal. The paving blocks meet the SNI 03-0691-1996 Quality Standard C, confirming their suitability for practical application.

**Keywords:** Bamboo leaf ash, Photocatalyst, TiO<sub>2</sub>-SiO<sub>2</sub> composite, Paving blocks, Pollutants

### 1. Introduction

Reducing toxic pollutants such as sulfur oxides (SO<sub>x</sub>) and nitrogen oxides (NO<sub>x</sub>) is a critical step in mitigating air pollution. These pollutants pose serious health risks, including respiratory and cardiovascular diseases [1]. Prolonged exposure has also been linked to increased risks of stroke, cancer, and premature death [2]. Beyond human health, SO<sub>x</sub> and NO<sub>x</sub> contribute to climate change and environmental degradation [3].

Air pollution is a global issue, affecting countries such as Brazil [4], India [5], China, United States [6], North Macedonia [7], and Indonesia [8]. One major source of these pollutants is the increasing volume of vehicular traffic, particularly in metropolitan areas [9]. In Indonesia, urban traffic continues to rise annually [10], contributing approximately 71% of NO<sub>x</sub> emissions and 15% of SO<sub>x</sub> and other particulate matter [11, 12].

One promising solution is the use of paving blocks coated with photocatalyst materials [13]. These coatings can absorb pollutants onto their surfaces, where, upon exposure to light, photocatalytic reactions oxidize the pollutant, which are then washed away by rainwater [14]. Paving blocks, composed of cement, fine aggregate, and water, are widely used in construction due to their environmental benefit [15].

Photocatalysts are materials that utilize light radiation to initiate chemical reactions. Titanium dioxide (TiO<sub>2</sub>) is one of the most commonly used photocatalysts due to its photostability, strong UV absorption, non-toxicity, chemical inertness, and efficiency in degrading organic compounds [16, 17]. Studies have shown that TiO<sub>2</sub> coatings can significantly reduce air pollutants. For example, TiO<sub>2</sub>-coated asphalt has demonstrated NO<sub>x</sub> removal efficiencies ranging from 31% to 55%, and SO<sub>2</sub> removal efficiencies from 4% to 20% [18, 19]. Photocatalytic concrete has achieved up to 60% NO<sub>x</sub> reduction under certain conditions [20]. A review reported NO<sub>x</sub> reduction of up to 60% in Milan, Italy, 20% in Paris, France, and between 16% and 90% in various U.S. locations [17].

The addition of SiO<sub>2</sub> can enhance the effectiveness of the TiO<sub>2</sub>-based photocatalyst layer by increasing acidity and transparency. This increased acidity promotes the absorption of free radicals and facilitates the degradation of organic compounds [21]. Researchers have utilized SiO<sub>2</sub>-rich materials in the manufacture of concrete paving blocks, including soda-lime glass (70 – 75% SiO<sub>2</sub>) [22] and sand (85 – 95% silica) [23, 24]. A study investigated the impact of using recycled glass rich in SiO<sub>2</sub> on the photocatalytic capacity of concrete paving blocks [25], while another study examined the effect of cathode ray tube glass and TiO<sub>2</sub> as substitutes for fine aggregate and cement [26]. These studies demonstrate that SiO<sub>2</sub> has been widely and effectively used as a supporting compound in paving block manufacturing, though most sources are glass-based.

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Alternative SiO<sub>2</sub> sources, such as silica fume and waste glass, are increasingly being used in pavement materials to enhance sustainability while enabling photocatalytic functionality. An investigation of the use of binary biomass ash and silica fume in low-carbon concrete for eco-friendly paving blocks. In this context, SiO<sub>2</sub> acts as a stabilizing matrix for the photocatalyst, helping to prevent nanoparticle agglomeration and improving light absorption for pollutant degradation. However, its effectiveness depends on precise dosing in order to balance performance benefits with potential handling challenges [27].

Recycled glass from beverage bottles or industrial waste is also considered a viable alternative source of SiO<sub>2</sub>. In photocatalytic applications, it provides a chemically stable, SiO<sub>2</sub>-rich matrix that supports long-term photocatalytic activity. However, a key limitation is the reduction in photocatalytic efficiency – by up to one-fifth – after several months of curing, primarily due to surface contamination [28].

Other sources of SiO<sub>2</sub> can be derived from organic materials, offering significantly more sustainable alternatives due to their renewability, lower energy requirements, reduced carbon footprint, and contribution to waste reduction. SiO<sub>2</sub>-rich compounds from organic sources can be obtained from the ash of burnt bamboo leaves. The addition of SiO<sub>2</sub> from bamboo leaf ash has been shown to improve the mechanical properties of paving blocks [29]. The novelty of this research lies in the use of bamboo leaf ash as a sustainable alternative source of SiO<sub>2</sub> to enhance the photocatalytic activity of TiO<sub>2</sub> in paving blocks.

Building on this background, the present study investigates the effect of a TiO<sub>2</sub>-SiO<sub>2</sub> composite photocatalyst coating derived from bamboo leaf combustion ash on the performance of paving blocks. The research aims to evaluate the quality and effectiveness of these paving blocks as a contributing material for reducing toxic pollutants (SO<sub>x</sub> and NO<sub>x</sub>) in highway environments. Paving blocks with a photocatalytic coating are expected to contribute to air pollutant reduction, aligning with United Nations Sustainable Development Goal (SDG) 11, which focuses on mitigating the impact of urbanization by improving air quality. In addition, the findings of this study have the potential to benefit various engineering fields by promoting the use of sustainable materials and mitigating air pollution and its associated health and environmental impacts.

## 2. Materials and methods

The materials used in this study included TiO<sub>2</sub> powder (purity approximately 99.5%), locally sourced bamboo leaves, Polyethylene Glycol (PEG) 6000, methylene blue, fine sand (with a fineness modulus of 2.35), Portland cement, and water. Sand granulometry was determined manually using a sieve shaker in the laboratory, and the results are presented in Table 1.

The equipment used included a concrete mixer, furnace, paving block mold (20 × 10 × 6 cm), cement scoop, sand sieve, bucket, scraper, small scoop, 150 mL porcelain cup, stainless pot, weighing scales, plate, compressive strength testing instrument, UV-Vis spectrophotometer, and scanning electron microscope (SEM).

This research was conducted over a five-month period at the Laboratory of Materials and Concrete Engineering, Department of Civil Engineering; the Chemical Engineering Laboratory; the Ecology Laboratory, Department of Chemical Engineering; and the Integrated Laboratory at Universitas Sumatera Utara, Medan, Indonesia.

**Table 1** Granulometry of Sand

Sieve No.	Sieve Size (mm)	Retained Weight (%)
No. 4	4.76	4.50
No. 8	2.38	11.95
No. 16	1.19	19.30
No. 30	0.6	38.70
No. 50	0.3	61.75
No. 100	0.15	99.25
Pan	-	100

### 2.1 Synthesis of TiO<sub>2</sub>-SiO<sub>2</sub> photocatalyst layer on paving blocks

Bamboo leaves were purchased, thoroughly washed in a container of water, sun-dried, and then pulverized. The TiO<sub>2</sub>-SiO<sub>2</sub> composite was prepared using various mass ratios of TiO<sub>2</sub> and SiO<sub>2</sub>: 2:0.5 (K1), 2:1 (K2), 0.5:2 (K3), 1:2 (K4), and 2:2 (K5). PEG 6000 was used as a binder at a fixed ratio of 5:8 of TiO<sub>2</sub>-SiO<sub>2</sub> composites and PEG 6000 mixtures. Each solid mixture was weighed and calcined in a furnace at 500 °C for 2 hours. After calcination, the bamboo leaf ash was found to contain 72% – 76% SiO<sub>2</sub>, as the major component [30, 31].

Paving blocks were produced by mixing cement, sand, water, and the TiO<sub>2</sub>-SiO<sub>2</sub> composite in a ratio of 1:4:0.5:1, using PEG 6000 as a binder. A concrete mixer was used for uniform blending. The mixture was then molded using conventional paving block molds. For the photocatalytic paving blocks, the structure was divided into two layers: a 55 mm thick base (normal) layer and a 5 mm thick composite layer. The base layer was prepared using the same ratio as the normal paving block but with a reduced mass.

Eight test samples were prepared for each variation, with each paving block measuring 20 × 10 × 6 cm. Standard normal paving block variations served as control samples for evaluating the effect of the photocatalytic layers. These normal variable blocks (K0) were produced without the TiO<sub>2</sub>-SiO<sub>2</sub> composite coating and are illustrated in Figure 1.

### 2.2 Paving block testing

Scanning electron microscopy (SEM) (SEM TM 3000, Technical Implementation Unit of Integrated Research Laboratory, Universitas Sumatera Utara, Medan, Indonesia) was conducted on the photocatalytic layer to assess the homogeneity of the TiO<sub>2</sub>-SiO<sub>2</sub> composite distribution. Additionally, UV-Vis spectrophotometry testing (Shimadzu UV-1800, the Ecology Laboratory, Department of Chemical Engineering, Universitas Sumatera Utara, Medan, Indonesia) was performed to evaluate the optimal composition and effectiveness of the TiO<sub>2</sub>-SiO<sub>2</sub> composite in reducing air pollutants such as SO<sub>x</sub> and NO<sub>x</sub>.

Compressive strength testing was performed using a compression testing machine (ELE International, maximum 2000 kN), in accordance with SNI 1974-1990/SNI 1974-2011 standards. Water absorption testing was also conducted using a soaking tub to evaluate the quality and durability of the paving blocks. All samples were cured for 21 days after molding and before undergoing testing.



**Figure 1** Visual comparison of paving blocks (a) Normal variation and (b) Addition of  $\text{TiO}_2\text{-SiO}_2$  composite variation

### 3. Results and discussions

#### 3.1 Paving block testing

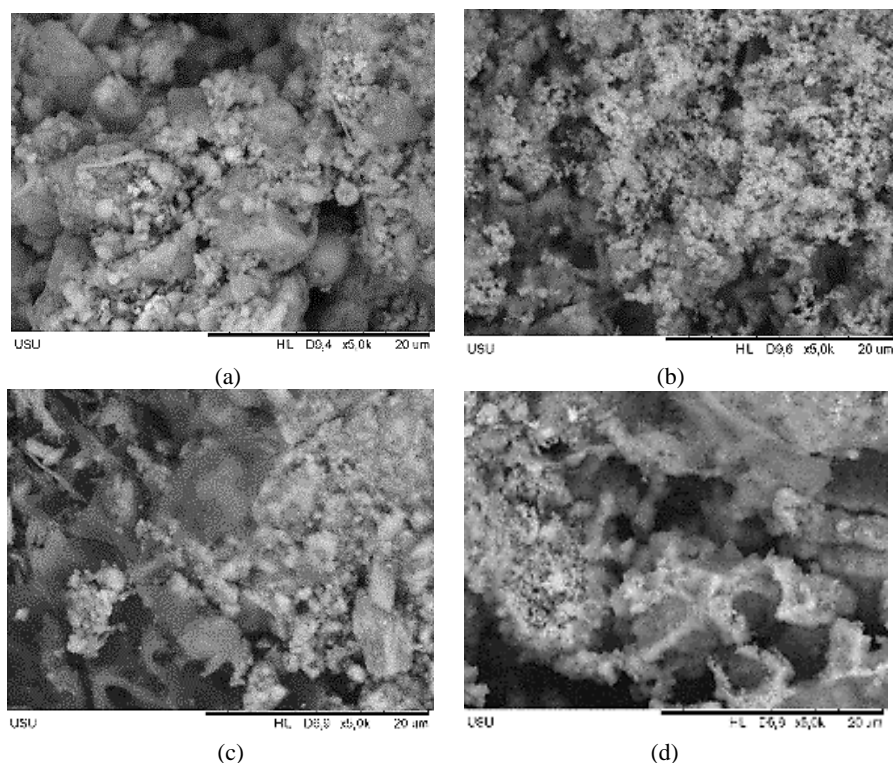
SEM analysis at 5000 $\times$  magnification was performed on the K1 to K5 test specimens to observe the morphological structure resulting from the addition of  $\text{SiO}_2$  and the use of PEG 6000 as a binder in the  $\text{TiO}_2$  photocatalyst layer. The results are presented in Figure 2.

The addition of  $\text{SiO}_2$  to the photocatalyst layer, using PEG 6000 as a binder, contributes to the formation of agglomerate structures, observed as chunk-shaped  $\text{TiO}_2\text{-SiO}_2$  clusters attached to the PEG 6000 surface [32]. SEM analysis of the sample revealed the presence of white powder agglomerates, indicating successful composite formation. These agglomerates are formed during the synthesis process, where PEG 6000 decomposes and facilitates the binding of  $\text{TiO}_2$  and  $\text{SiO}_2$ , resulting in a stable composite structure.

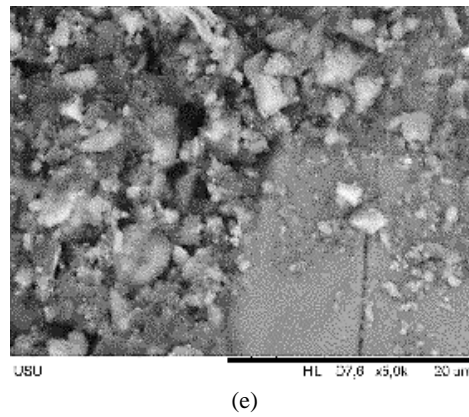
PEG 6000 act as a dispersing agent that helps prevent agglomeration during the synthesis of  $\text{TiO}_2\text{-SiO}_2$  composites. It enhances uniform particle distribution, which is vital for maintaining high photocatalytic activity [33]. Additionally, PEG 6000 enhances photocatalytic efficiency by increasing porosity, surface area, and the interaction between the composite and environmental pollutants within the paving blocks. These improvements make the material more effective at degrading environmental pollutants while supporting sustainable construction practices [34].

In samples K1 and K2, which have a higher  $\text{TiO}_2$  composition, the  $\text{TiO}_2\text{-SiO}_2$  agglomerates are relatively evenly distributed. In contrast, samples K3 and K4, with lower  $\text{TiO}_2$  content, show less uniform distribution of the agglomerates. Sample K5, with a balanced  $\text{TiO}_2$  and  $\text{SiO}_2$  ratio, exhibits a relatively even distribution of  $\text{TiO}_2\text{-SiO}_2$  bonds.

It has been reported that agglomeration can hinder particle dispersion within the cement matrix, weakening stress distribution due to the formation of low-strength agglomerates [35]. Additionally, agglomeration increases particle size, which reduces the overall surface area available for photocatalytic reactions [36]. Overall, the addition of  $\text{SiO}_2$  with PEG 6000 as a binding agent successfully facilitated the formation of  $\text{TiO}_2\text{-SiO}_2$  composite agglomerates.



**Figure 2** SEM analysis results of paving block samples (a) K1, (b) K2, (c) K3, (d) K4, and (e) K5



(e)

**Figure 2 (continued)** SEM analysis results of paving block samples (a) K1, (b) K2, (c) K3, (d) K4, and (e) K5

### 3.2. Test results of paving block compressive strength

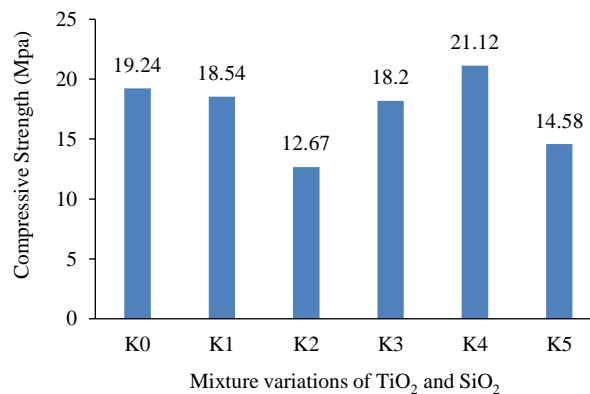
Figure 3 presents the results of the compressive strength tests. The average compressive strength values for each paving block variation were 19.24 MPa, 18.54 MPa, 12.67 MPa, 18.20 MPa, 21.12 MPa, and 14.58 MPa. The highest compressive strength was recorded in the 1 TiO<sub>2</sub>:2 SiO<sub>2</sub> variation (21.12 MPa), while the lowest compressive strength was observed in the 2 TiO<sub>2</sub>:1 SiO<sub>2</sub> variation (12.67 MPa). All variations met the minimum requirements for compressive strength for Quality C paving blocks, as specified in SNI 03-0691-1996 for [37].

The percentage decrease in compressive strength for each photocatalyst composite variation, relative to the normal sample, showed decreases of 7%, 66%, and 10%, an increase of 19%, and a final decrease of 47%. These results indicate that as the TiO<sub>2</sub> ratio increases, the compressive strength of the paving blocks tends to decrease. This trend aligns with previous research on the photocatalytic and mechanical properties of TiO<sub>2</sub>-modified concrete [38].

A higher SiO<sub>2</sub> content combined with a lower TiO<sub>2</sub> ratio tends to increase the compressive strength of paving blocks. Excessive TiO<sub>2</sub> can create weak points within the matrix due to poor particle interaction and bonding. While TiO<sub>2</sub> contributes to the formation of calcium silicate hydrate (C-S-H) – a key compound for strength development in concrete [39] – its overuse may disrupt the matrix structure.

In contrast, higher SiO<sub>2</sub> content improves compressive strength by improving hydration dynamics. SiO<sub>2</sub> reacts with Ca(OH)<sub>2</sub> to form additional C-S-H, thereby strengthening the concrete matrix. However, excessive SiO<sub>2</sub> can also lead to the formation of air cavities during hydration, which reduce density and compromise structural integrity, ultimately lowering compressive strength [40].

Therefore, maintaining an appropriate balance between SiO<sub>2</sub> and TiO<sub>2</sub> is crucial. The results of this study align with previous research on the properties of concrete pavements containing waste aggregates and nano-SiO<sub>2</sub> [41].



**Figure 3** Compressive strength test results of paving block mixtures with different TiO<sub>2</sub>-SiO<sub>2</sub> variations

### 3.3. Test results of water absorption of paving blocks

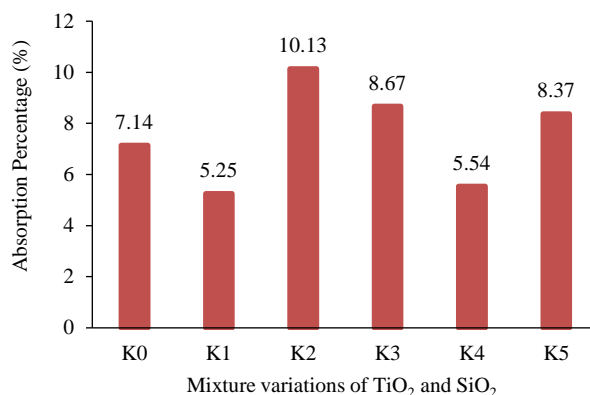
Water absorption testing was performed by soaking the test specimens for 24 hours to determine their wet weight, followed by oven-drying at 115 °C for another 24 hours to obtain their dry weight. The water absorption results for each variation (Figure 4) were 7.14%, 5.25%, 10.13%, 8.67%, 5.54%, and 8.47% respectively. These water absorption values fluctuate randomly.

As paving blocks absorb water, they also have the potential to absorb pollutants. The interconnected pore structure within the block facilitates the movement of contaminants into the material. Blocks with higher porosity may absorb more pollutants due to their increased surface area and pore volume. However, excessive water absorption can lead to long-term deterioration, as pollutants may trigger chemical reactions that weaken the structural integrity of the blocks [42, 43].

The pore structure of paving blocks is defined by the size and connectivity of capillary pores. During the hydration process, a network of pores forms, allowing water absorption. This absorption is influenced by both the volume and size of interconnected pores, which can vary depending on the materials used in the block's composition. For instance, incorporating materials such as coconut shell ash or palm oil eco-processed pozzolan can modify the pore structure, thereby affecting water absorption rates. Higher proportions of these materials often result in increased porosity and, consequently, higher water absorption values [44, 45].

High water absorption is often associated with lower compressive strength due to the presence of larger voids that weaken the material [46]. Among the paving block variations, K0, K1, K3, K4, and K5 met the maximum water absorption limits specified in SNI 03-0691-1996 Quality C [37]. However, the K2 variation did not meet these requirements. This discrepancy may be attributed to compaction issues during the molding process.

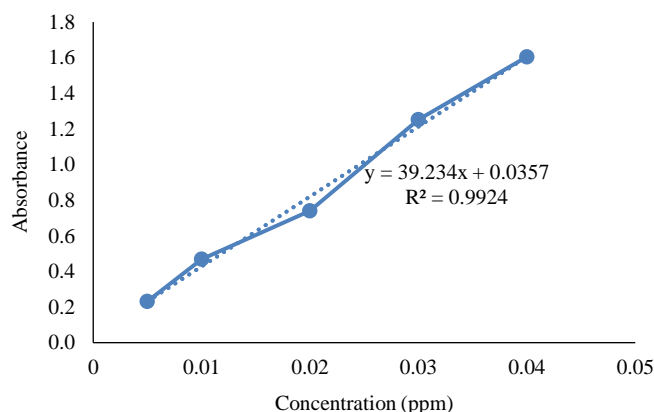
The density of the material used in the test specimens dramatically affects their water absorption capacity. Lower-density paving blocks tend to absorb more water. In contrast, higher-density paving blocks have more tightly bound particles, which reduces the size of the air cavities within the material. As a result, it becomes more difficult for water to penetrate and fill these smaller cavities.



**Figure 4** Water absorption test results of paving block mixtures with different TiO<sub>2</sub>-SiO<sub>2</sub> variations

### 3.4. UV-Vis spectrophotometer testing results

The solution was prepared by dissolving SO<sub>x</sub> and NO<sub>x</sub> pollutants in a methylene blue solvent. The resulting stock solution had a pollutant concentration of 0.040 ppm and was subsequently diluted to concentrations of 0.030 ppm, 0.020 ppm, 0.010 ppm, and 0.005 ppm. These five solutions were analyzed using UV-Vis spectrophotometry within the 600 – 800 nm wavelength range to obtain absorbance values, the results are shown in Figure 5.



**Figure 5** Plot results of the absorbance data of solution

Based on Figure 5, a linear equation can be used to determine the pollutant levels in the sample. Table 2 presents the calculated amounts of pollutants successfully absorbed by the paving block.

**Table 2** Sample level calculation results

Ratio of TiO <sub>2</sub> to SiO <sub>2</sub>	Absorbance	Concentration (ppm)
K0	1.079	0.027
K1	3.316	0.084
K2	3.141	0.079
K3	3.210	0.081
K4	2.975	0.075
K5	3.404	0.086

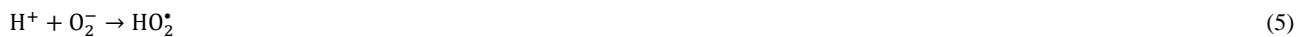
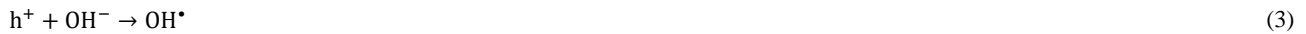
The application of the TiO<sub>2</sub>-SiO<sub>2</sub> composite photocatalytic layer enhances the effectiveness of pollutant absorption in paving blocks. A significant difference is observed between the normal (K0) and the treated samples (K1 – K5). Among the treated paving blocks, K5 shows the highest pollutant removal, with a concentration of 0.086 ppm or 45.71%, achieved using a TiO<sub>2</sub>: SiO<sub>2</sub> ratio of 2:2. This balanced ratio improves the photocatalyst's efficiency in both adsorbing and degrading pollutants.



The lowest pollutant removal was observed in sample K4, which used a TiO<sub>2</sub>: SiO<sub>2</sub> ratio of 1:2. This reduced performance is attributed to the lower TiO<sub>2</sub> content, which limits the photocatalytic activity of the composite layer. While SiO<sub>2</sub> contributes to structural support and surface area, an insufficient amount of TiO<sub>2</sub> hinders the photocatalytic reaction.

These results can be compared to previous studies. For example, a study reported that a cerium nitrate (Ce)–TiO<sub>2</sub> photocatalyst achieved up to 25% NO degradation using a 0.2% Ce content [47]. Similarly, nitrogen-doped TiO<sub>2</sub> (N-TiO<sub>2</sub>, 6%) incorporated into pavement material achieved ~23% NO degradation [48]. In another investigation, TiO<sub>2</sub> was applied using a fluid technique—spraying a TiO<sub>2</sub>-containing water solution or binder emulsion onto asphalt pavement—and achieved a maximum degradation efficiency of 20% for SO<sub>2</sub> pollutants [19].

The photocatalytic process is primarily activated by light irradiation. Reactions 1 – 5 illustrate the sequential photocatalytic mechanisms based on the electron structure of TiO<sub>2</sub> [49]:



The mechanism involving the reactive species O<sub>2</sub><sup>−</sup> and OH<sup>•</sup>, produced in reactions 2 – 4, leads to their interaction with NO<sub>x</sub> pollutants. The proposed reaction pathway for this process is outlined as follows [50]:



The superoxide ion O<sub>2</sub><sup>−</sup>, formed in Equation (5), generates hydroperoxyl radicals (HO<sub>2</sub><sup>•</sup>), which then react with NO according to Equations (7) and (8), ultimately producing HNO<sub>3</sub>. Similarly, based on Equations (6) and (7), OH<sup>•</sup> produced during the photocatalytic reaction interact with NO<sub>x</sub> species – including NO, and NO<sub>2</sub> – leading to the formation of HNO<sub>3</sub>. This HNO<sub>3</sub> is soluble in water and can be easily removed from the surface of the catalyst by environment factors such as rain.

In the case of SO<sub>x</sub> pollutants, the reaction mechanism proceeds through Equations 11 – 13, as follows:



As shown in reactions 11 – 13, sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) is formed through the oxidation of SO<sub>2</sub> to SO<sub>3</sub>, which then reacts with atmospheric H<sub>2</sub>O to produce H<sub>2</sub>SO<sub>4</sub>. The resulting H<sub>2</sub>SO<sub>4</sub> is water-soluble and can be easily washed away from the surface of the photocatalytic paving block by environmental factors such as rain.

#### 4. Conclusions

- The synthesis of TiO<sub>2</sub>-SiO<sub>2</sub> composites resulted in a fine, whitish-gray powder with a lighter final weight. Paving blocks incorporating the composite can be visually distinguished from normal blocks by the presence of a 5 mm thick photocatalyst layer with a whitish color on the top surface. Based on the overall testing, the best performance was observed in variation K5, which used a TiO<sub>2</sub>:SiO<sub>2</sub> ratio of 2:2. This variant demonstrated:
  - Pollutant sorption of 0.086 ppm
  - Uniform distribution of the TiO<sub>2</sub>-SiO<sub>2</sub> composite across photocatalyst surface
  - Water absorption of 8.47%
  - Compressive strength value of 14.58 MPa
- SEM analysis confirmed that the agglomeration structure was evenly distributed in variations with equal or higher TiO<sub>2</sub> content relative to SiO<sub>2</sub>, contributing to improved photocatalytic performance.
- The study successfully integrated SiO<sub>2</sub> derived from bamboo leaf ash with TiO<sub>2</sub> using PEG 6000, resulting in a well-distributed composite suitable for photocatalytic applications in paving blocks.
- The developed TiO<sub>2</sub>-SiO<sub>2</sub> composite paving blocks show promising potential for pollution reduction in urban environments. However, future research is needed to evaluate their long-term durability under varying environmental conditions, including exposure to UV radiation, moisture, and temperature fluctuations. Additionally, further studies should explore the scalability of these composite paving block for practical applications, considering factors such as cost, production feasibility, and large-scale environmental benefits.

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