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Advancing photocatalytic concrete technologies in design, performance and application for a sustainable future

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ABSTRACT

Photocatalytic concrete technology is gaining attention in sustainable building and infrastructure for its crucial role in catalyzing the decomposition of harmful air pollutants and improving air quality. It incorporates photocatalysts such as Titanium dioxide (TiO_2) and Zinc oxide (ZnO) to purify the air and offer self-cleaning capabilities. This review examines the pollutant removal capabilities of photocatalytic concrete, analyses the factors influencing its efficacy, explores different preparation methods and mechanical properties, and includes a life cycle assessment (LCA) to evaluate its environmental impact. Cement-based materials, serving as a carrier for photocatalysts, exhibit varying effects based on the type of photocatalysts, especially different types of TiO_2 and ZnO crystals. Analysis of preparation methods, including mixing, spraying and impregnation, emphasizes the imperative need for research aimed at improving the active lifespan and bonding strength of the coating to the substrate of concrete. The discussion covers strategies for enhancing photocatalyst performance through surface modification, addressing the associated technical and future challenges. Innovative methods such as the use of recycled glass to increase nitrogen oxides removal rates and the incorporation of porous materials such as zeolites to increase the photocatalytic efficiency of sulfur dioxide (SO_2) and carbon dioxide (CO_2) were evaluated. The TiO_2 nanoparticle fraction significantly influences the hydration and overall performance of cement-based materials, with an optimal range of 4–10 wt% of the cement mass recommended. LCA analyses indicate the need for exploring more environmentally friendly design options to enhance the application of photocatalytic technology in concrete infrastructure such as building facades, roads, tunnels and other infrastructures.

1. Introduction

1.1. Air pollution problems on a global scale

The continuous growth of the global economy and accelerated transport development since the turn of the century have contributed to increased air pollution, despite improvements in productivity and living standards. Particulate matter (PM) is a principal air pollutant, comprising primary PM directly emitted from engine combustion and secondary PM formed by chemical reactions between gaseous precursors such as nitrogen oxides (NO_x) and sulphur dioxide (SO_2).

NO_x plays a key role in chemical reactions in the atmosphere and are important precursors for the formation of particulate matter (e.g. PM2.5 and PM10). These consist of solid or liquid particles up to 2.5 μm or 10 μm in diameter and are essential for the formation of particulate pollution. In short, the presence of NO_x and the conversion processes directly influence the production of PM10 [1]. NO_x is a group of compounds composed of nitrogen and oxygen, specifically nitrogen monoxide (NO), nitrogen dioxide (NO_2) and nitrous oxide (N_2O). In engine emissions, NO_x mainly refers to NO and NO_2, with NO having the highest proportion. These emissions have a significant impact on the environment and human health. However, considering the health effects of air pollution, NO_x deserves special attention. While some NO_x may have limited environmental or medical benefits, its main detrimental effect is the formation of acidic particles after emission, which are dispersed by wet and dry deposition and damage the environment. NO_x, particularly NO and N_2O, are a major source of air pollution and their reaction products, such as acid rain and photochemical smog, pose

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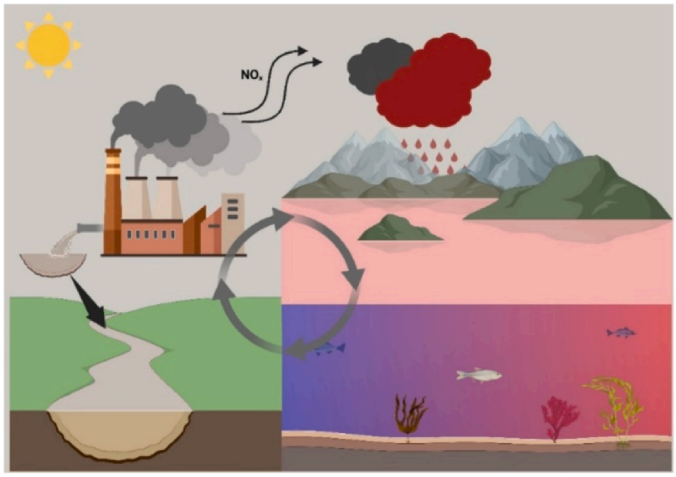


Fig. 1. Environmental impacts caused by NO_X air pollution.

a significant threat to the environment and human health. In addition, pollutants such as carbon monoxide (CO), HC (hydrocarbons), NO_2, PM, SO_2 and Pb contribute to environmental pollution, as depicted in Fig. 1.

NO_x can directly irritate the human respiratory system, leading to asthma or breathing difficulties [1,2]. According to STATISTA data [3–5], as shown in Fig. 2(a), the United States, Canada and Australia account for the majority of 90% of global NO_x emissions in 2022. To some extent, this has led to certain irreversible environmental issues, such as soil acidification. Research shows that NO formed during fuel combustion mainly includes two types [6]: (i) Prompt NO: when fuel combustion is incomplete, CH molecules in the fuel react with N_2 in the oxidant to form intermediate products, which then act as 'bridge mediums' and react with free O and OH radicals in the flame to form NO, as shown in Eqs. (1) to (3); (ii) Fuel NO [7]: Usually during the combustion of fossil fuels, nitrogen-containing compounds under oxidizing conditions form intermediates, which then react with oxygen in the air to produce NO, as shown in Eqs. (4) and (5).

$$CH + N_2 \rightarrow H + NCN \quad (1)$$

$$CH_2 + N_2 \rightarrow HCN + NH \quad (2)$$

$$C_2 + H_2 \rightarrow 2CH \quad (3)$$

$$CN + O \rightarrow C + NO \quad (4)$$

$$NH + O_2 \rightarrow NO + OH \quad (5)$$

With population growth and accelerated urbanization, there has been a significant increase in the volume of solid waste [9], which often

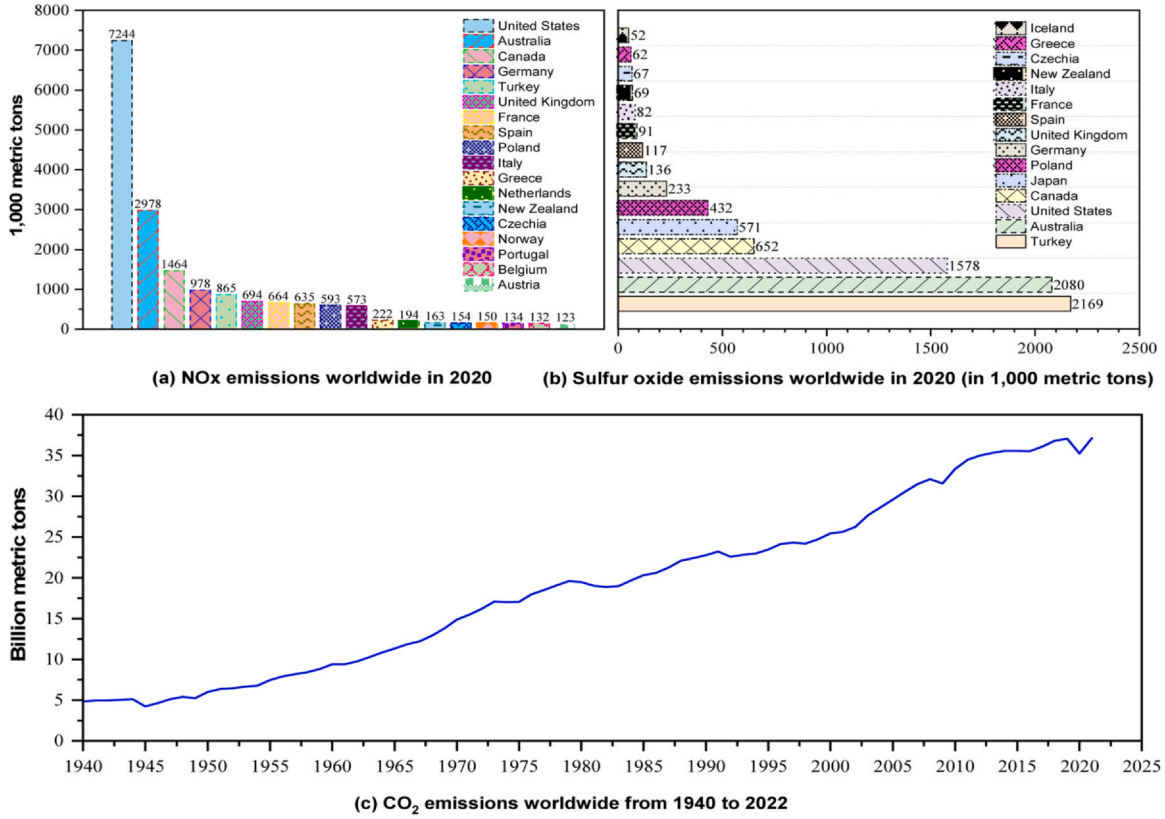


Fig. 2. Global emissions of (a) NO_x, (b) SO_x and (c) CO_2 [8].

contains sulphur compounds. Incineration is effective in relieving pressure on landfills by reducing the volume of waste, treating hazardous substances, and generating electricity or heat. However, the treatment of sulphur-containing waste releases hydrogen sulphide (H_2S) and Sulfur Oxides (SO_x), the main sources of which are organic waste, sludge, biomass, ores and fossil fuels. These substances decompose at high temperatures to form SO_2 and H_2S, contributing to air pollution as revealed in Eqs. (6–8) [8,10]. In Fig. 2(b), Turkey, Australia and the United States are among the top three countries in terms of sulphide emissions. In particular, SO_2 can lead to acid rain that damages water bodies, soils, ecosystems and crops, while H_2S is a toxic gas that is harmful to humans.

$$H_2S + 1.5O_2 \rightarrow SO_2 + H_2O(G) \quad (6)$$

$$SO_2 + 0.5O_2 \rightarrow SO_3 \quad (7)$$

$$SO_3 \rightarrow SO_2 + 0.5O_2 \quad (8)$$

The reduction of CO_2 (a key greenhouse gas) has been a long-standing issue for the international community. Carbon emissions from human activities arise mainly from energy production and use, industrial production, deforestation, and land-use change. Industrial processes emit large quantities of CO_2 in the combustion of coal, oil, and natural gas. At the same time, deforestation leads to the loss of carbon stocks, and the conversion of land to urbanization and agricultural land reduces carbon sequestration. As shown in Fig. 2(c), global carbon emissions have increased steadily with human technological progress since 1940. However, these human activities increase the CO_2 concentration in the atmosphere, leading to global warming and climate change. Consequently, urgent measures are essential to mitigate climate change effects, including reducing carbon emissions, adopting clean energy, protecting forests, and enhancing land management.

1.2. Definition of Photocatalysis

Photocatalysis is a widely adopted technology for tackling air pollution due to its distinctive capability to utilize sunlight for catalyzing chemical reactions on surfaces. This process effectively breaks down harmful pollutants into harmless compounds, contributing to the purification of the air and reducing the concentration of pollutants, including NOx and VOCs (volatile organic compounds) [11–14]. Photocatalysis goes beyond enhancing air quality; it possesses self-cleaning properties, thereby reducing the maintenance costs associated with urban infrastructure. Photocatalysis is widely acknowledged as a sustainable and energy-efficient solution. Its potential to improve urban air quality and contribute to healthier environments has led to its increasing adoption, highlighting its promise as a tool in the fight against air pollution.

Photocatalysis is a chemical reaction process that utilizes light energy to excite semiconductor materials, leading to the generation of electron-hole pairs and promoting chemical reactions. These semiconductors are typically wide bandgap semiconductors, such as TiO_2 and zinc oxide (ZnO). As shown in Fig. 3, when the incident photons have sufficient energy (i.e. higher than the absorption threshold of the semiconductor), the valence band electrons of the semiconductor undergo interband transition, jumping from the valence band to the conduction band. This elevates the electrons to a higher energy level, creating an electron (e^-) - hole pair (h^+). Subsequently, the electrons move in the conduction band, while the holes move in the valence band. They migrate in different ways within the semiconductor. Once the electrons and holes reach the surface of the semiconductor, they can participate in chemical reactions. Electrons can reduce surrounding substances, while holes can oxidize other materials. These reactions can reduce gases, such as CO_2, NO_x and SO_x, and degrade organic pollutants including benzene. Ultimately, the photocatalytic reaction could produce harmless products such as water and CO_2 and be used for water purification or energy production such as hydrogen [15–17].

1.3. Characteristics of photocatalytic materials

The activity of photocatalytic materials is affected by their own properties, such as particle size, surface area, morphology, phase composition, band gap energy, magnetization strength, surface defects, adsorption properties and photocatalytic reactivity. In semiconductor materials, the diffusion efficiency of electron-hole pairs is a crucial factor in determining the photocatalytic activity. Despite the relatively low number of nonequilibrium carriers, they can generate significant concentration gradients during their migration. The effective photocatalytic reaction process depends on the diffusion of these carriers to the semiconductor surface and their interaction with surface electrons. Although the diffusion length of the material remains constant, the key to achieving effective suppression of electron-hole complexation and enhancing their migration efficiency lies in the precise control of the particle size of the material. If the particle size of the material is smaller than the diffusion length of the nonequilibrium carriers, the carriers can be transported more efficiently rather than compounding within a particle. As the particle size of the material decreases, the carriers traverse shorter paths within the material, thereby reducing the likelihood of recombination and enhancing migration efficiency. This, in turn, increases the concentration of ions diffused to the surface, thereby improving the catalytic performance[18,19].

The pH of the solution plays a crucial role in regulating catalytic activity by external factors. This parameter influences the ionization state of the reactants and the rate of the catalytic surface reaction by adjusting the surface charge of the semiconductor material, subsequently altering the material adsorption capacity on the substrate. It is worth noting that the adsorption process is one of the fundamental prerequisites for catalytic reactions to occur. The effective adsorption capacity of a catalyst on the substrate molecules is a key factor in determining whether the catalytic reaction can be carried out successfully. If the surface charge of the catalyst is opposite to that of the substrate molecules, the electrostatic interaction between the two can increase the adsorption efficiency, thereby promoting catalytic activity. Conversely, if the catalyst and substrate molecules have the same charge, the adsorption capacity can be weakened, further reducing the catalytic efficiency. Previous studies have shown [20,21] that the solution pH can change the surface potential of TiO_2 and thus its adsorption degradation properties. Secondly, pH can affect the semiconductor energy band positions and thus change the hole oxidation capacity. In general, lower pH values favor the adsorption of organic compounds, while higher pH values can provide more hydroxide ions to react with the holes to form OH radicals, thus increasing the degradation rate of organic matter [22,23].

Meanwhile, the impact of reaction temperature should not be underestimated. Barakat et al. [24] found that an increase in temperature to 55 °C enhances the degradation effect for Ag-doped TiO_2 nanoparticles, but the optimal temperature is 25 °C for the nanofiber form. The specific temperature effect observed in the nanofiber form indicates that a rise in temperature leads to rapid degradation of dye molecules, as it increases the kinetic energy of the molecules, causing them to escape from the active film around the photocatalyst. Kim et al. [25] examined the effect of different organic hole scavengers (ethylenediaminetetraacetic acid (EDTA), methanol and formic acid) on the H_2 production rate of bare and platinum coated TiO_2 (Pt/TiO_2) suspensions under different light wavelengths (UV and visible light). Similarly, Chen et al. [26] found that the catalytic activity of TiO_2 and Pd/TiO_2 increased with temperature from 0 to 50 °C. However, at 70 °C, the performance of Pd/TiO_2 diminished, and TiO_2 showed a slight decline. Cu/TiO_2 exhibited higher activity at room temperature, underscoring the significant influence of reaction temperature and the type of auxiliary catalyst on catalytic activity. It was observed that temperatures above 70 °C led to increased recombination of carriers. Consequently, the research suggests that an optimal temperature range for effective photodegradation of organic substances lies within 50–80 °C.

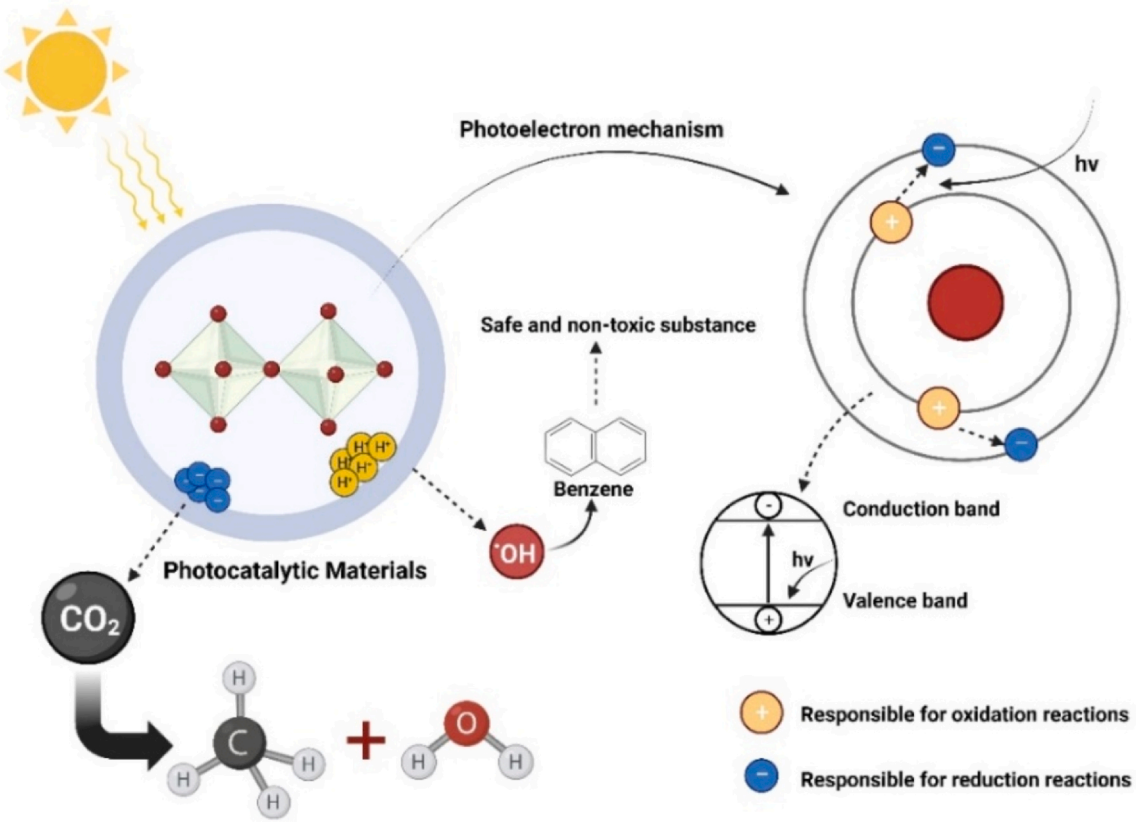


Fig. 3. Mechanisms of photocatalytic reaction [15–17].

1.4. Photocatalytic concrete

Besides using administrative and economic strategies to combat air pollution from greenhouse gases, it is vital to seek out comprehensive solutions. Photocatalytic concrete technology serves as an innovative solution. The photocatalytic concrete can purify the air and clean itself using sunlight [27,28]. In recent studies [29–32], the integration of photocatalytic technology with concrete materials has demonstrated excellent efficacy, which includes properties such as self-cleaning, the ability to effectively eliminate harmful gases and the reduction of road exhaust pollution.

In the 1990 s, the first practical application of photocatalytic concrete took place on a busy street in Tokyo, where photocatalytic concrete panels were installed that were coated with a layer of TiO_2. This innovative building product not only possessed photocatalytic functions but antimicrobial properties, marking the first practical application of a photocatalytic building product [33]. Subsequent projects and studies in Japan and other countries have further extended the application of photocatalytic concrete to a variety of urban infrastructure elements such as roads, pavements, building facades and public spaces. These early projects paved the way for the continued development and commercial application of photocatalytic concrete technology in architecture and urban planning.

Ballari et al. [34] laid photocatalytic concrete blocks on Castorweg Street in Hengelo to evaluate the degradation effect of these photocatalytic blocks on nitrogen compounds, as shown in Fig. 4(a). The results demonstrated that concrete pavements incorporating photocatalysts significantly reduced NOx concentrations by 19% over the entire day and by 28% during afternoon hours, compared to pavements without photocatalysts. Notably, under ideal weather conditions, i.e. high radiation and low relative humidity, the reduction in NO_x concentrations could reach 45%. Hüsken et al. [35] evaluated the performance and air purification properties of concrete products containing photocatalytic TiO_2. Under optimal laboratory conditions, some products had an average NO degradation rate of about 40%. Cardellicchio [36] conducted a durability study on the Roman Jubilee Church in Fig. 4(b), which utilized some of the earliest ultra-white reinforced concrete and self-cleaning photocatalytic cement including TiO_2. The results indicated that the hydrophilic light-induced effect of TiO_2 could maintain the youthfulness of the walls by cleaning volcanic ash dirt, thereby extending their lifespan. However, it is important to note that surface wear on the convex parts of the concrete could significantly increase the mechanical bonding between the concrete and rain dust (rougher surfaces provide more microscopic contact points, which helps to increase mechanical interlock between the two materials, thus improving bond strength), reducing the material hydrophilicity.

Although the de-pollution effects of photocatalytic construction and building materials are evident, unresolved issues exist in applications. Maintaining the long-term photocatalytic performance of materials such as TiO_2 on the surface of building materials is challenging. Aging of building materials may lead to a decrease in photocatalytic activity, thus reducing the effectiveness of the materials. Maury-Ramirez et al. [37], they performed accelerated weathering tests on the photocatalytic properties of autoclaved aerated concrete. The results of the study showed that, after weathering, the coating thickness of TiO_2 nanoparticles applied on the surface of autoclaved aerated concrete by dip-coating technique and a novel vacuum saturation method was reduced by more than 98%, while the content of TiO_2 was also reduced by more than 99% and 93%, respectively. However, despite the significant reduction in coating thickness and TiO_2 content, the residual TiO_2 particles in the autoclaved aerated concrete matrix were still able to act as effective active sites, resulting in a photocatalytic activity that showed only a slight decrease compared to the original performance. Other studies [38–40] pointed out that environmental stresses, such as degradation and peeling of coating adhesives and adsorption of pollutants or reaction products of the photocatalytic process, could lead to the detachment of TiO_2 particles and reduction in coating thickness.

(a) Castorweg Street [34] (b) Roman Jubilee Church [36]

Fig. 4. Photocatalytic concrete in practice.

The maintenance of TiO_2 materials may be required for sustained pollution reduction effects, necessitating regular cleaning or replacement. Furthermore, while the preparation of photocatalytic concrete holds promise, critical issues must be addressed. Ensuring the uniform distribution of photocatalytic materials in the concrete matrix is essential for effective performance. Managing the impact of photocatalytic particles on structural integrity and mechanical performance poses a significant challenge. Balancing photocatalytic activity with fundamental properties such as strength, workability and setting time is crucial. Lastly, the addition of photocatalytic materials increases overall production costs, presenting a key challenge in achieving an optimal balance between cost and performance for practical applications.

Comprehensive reviews in the category of photocatalytic concrete are limited. This work aims to fill this gap by providing an extensive review of the latest developments in the preparation and application of photocatalytic concrete for degrading air pollutants. The article delves into the preparation methods, material characteristics and life cycle assessment for this technology. By offering a thorough resource, this review seeks to support informed decision-making, inspire future research, advocate for environmentally friendly building practices, and promote the widespread adoption of photocatalytic concrete technology to address air pollution and enhance the sustainability of urban environments.

2. Preparation process for photocatalytic concrete

2.1. Selection of photocatalysts

TiO_2, especially in its nano form (Anatase and rutile crystalline structures), is the most widely used photocatalyst due to excellent photocatalytic performance, robustness, and compatibility with building materials. It is often used in concrete to prepare photocatalytic concrete [41,42] without significantly impacting the performance of the products. TiO_2 exhibits high reactive activity under sunlight or ultraviolet light, effectively decomposing pollutants in the air and organic pollutants on the surface of concrete. In 1979, Inoue et al. [43] first confirmed the degradation theory of CO_2 in photocatalytic principles, clarifying the kinetics of photocatalytic reactions through the charge transfer theory of semiconductor materials (TiO_2, ZnO, etc.), as shown in Eqs. (9–14). Gas-liquid mixtures such as CH_4, CH_3OH, etc. are produced, and these mixtures are again absorbed by the photocatalytic material. In the process, light-produced holes can react with these reduction products, causing them to be re-oxidized to CO_2.This re-oxidation process leads to the re-formation of CO_2, thus reducing the overall CO_2 reduction rate. Moreover, the relatively small specific surface area and instability of the cement substrate in photocatalytic reactions limit its capacity to offer the necessary active sites for effective pollutant degradation, thus diminishing the overall efficiency of the photocatalytic process.

Previous studies [44,45] have shown that whether TiO_2 is applied by surface coating or mixed to prepare photocatalytic concrete, its photocatalytic activity decreases over time. From our perspectives, we propose several key factors contributing to these challenges: (i) the uneven distribution of TiO_2 particles in the cement matrix, leading to regions with low or no photocatalytic activity and (ii) exposed TiO_2 primarily responding to ultraviolet (UV) light which constitutes only a small part of the solar spectrum. The hydration products largely encapsulate TiO_2 particles, reducing light penetration and significantly diminishing photocatalytic activity. Furthermore, previous research indicates that carbonation results in a reduction in the photocatalytic activity of cementitious materials containing TiO_2 [41]. Consequently, current research is primarily focused on identifying novel photocatalysts and exploring modified forms of TiO_2 to address these challenges effectively.

$$\text{Conductive Materials } (TiO_2 \text{or ZnO, etc}) + hv \rightarrow h^+ + e^- \quad (9)$$

$$H_2O + h^+ \rightarrow OH + H^+ \quad (10)$$

$$CO_2 + 2H^+ + 2e^- \rightarrow HCOOH \quad (11)$$

$$HCOOH + 2H^+ + 2e^- \rightarrow HCHO + H_2O \quad (12)$$

$$HCHO + 2H^+ + 2e^- \rightarrow CH_3OH \quad (13)$$

$$CH_3OH + 2H^+ + 2e^- \rightarrow CH_4 + H_2O \quad (14)$$

Nava-Nú ñez et al. [46] used bismuth oxyhalides BiOX (X = Cl, Br and I) photocatalysts in cement-based materials, delving into their impact on the intrinsic properties of the cementitious materials. These catalysts belong to a desirable new category of layered materials due to their unique layered structure, suitable bandgap structure, as well as their chemical stability and good cost-effectiveness. The results showed that cement-based samples containing BiOX had a higher pollution removal rate than those containing TiO_2, mainly due to the presence of oxygen vacancies and the strong oxidation potential in the catalyst. Kaja et al. [47] observed that silica fume enhances the NO_2 adsorption capacity of TiO_2 cementitious materials. This is primarily because silica fume introduces a greater volume of C-S-H gel, which possesses a higher specific surface area, into the matrix. This contrasts with the relatively smaller specific surface area of the calcium silicate hydrate crystals typically found in cement mortar. The high-specific surface area gel structure facilitates the elimination of NO_2. Furthermore, the study pointed out that the introduction of silica fumes led to higher capillary

porosity rates, which could prolong the residence time of NO_2, thereby enhancing the elimination rate.

Balbuena et al. [48] successfully synthesized a new type of a Fe_2O_3/TiO_2 nanocomposite via a plasma-assisted pathway. This composite exhibited excellent performance in photocatalysis, benefiting from the beneficial synergistic effect between Fe_2O_3 and TiO_2. In particular, the material significantly improved the removal activity of NO_x, demonstrating a capacity to selectively convert it into nitrates with a selectivity exceeding 60%. This research provides a promising direction for the development of efficient photocatalysts, especially in the field of air pollution control.

Shen et al. [49] developed an innovative photocatalytic carbonation coating for concrete, utilizing silicate and carbonate minerals (including C_3S, C_2S, C_3S_2 and CS) and introducing TiO_2, to address the decline in photocatalytic performance of traditional TiO_2 cementitious materials during the carbonation process. The results showed that the uniform distribution of TiO_2 particles on the mineral matrix of acid salts helps increase the number of photocatalytic active sites, thereby enhancing the overall catalytic activity. Consequently, this study established that the carbonated C_2S materials modified with TiO_2 exhibit superior NO removal performance, offering a viable direction for the development of innovative construction materials.

Cementitious materials are widely acknowledged as excellent carriers for photocatalysts, offering advantages such as a suitable pore structure, stability, adhesion, processability, low cost and durability [50]. TiO_2-based photocatalytic cementitious materials have demonstrated outstanding performance in air purification and self-cleaning properties [51–53]. However, building materials based on TiO_2 may have drawbacks that could result in the degradation of concrete surface properties, impacting its durability and mechanical characteristics. Hence, the careful selection of appropriate photocatalysts is essential to ensure the environmental friendliness and long-term sustainability of concrete.

2.2. Preparation technology of photocatalytic concrete

Currently, the primary methods for preparing TiO_2 photocatalytic concrete include immersion, internal mixing, spraying, thin-film coating, etc., each with advantages and disadvantages. In the immersion method, the sample is soaked in a suspension containing TiO_2 for a specific duration. This method can achieve higher photocatalytic efficiency with a smaller amount of TiO_2. Xu et al. [54,55] determined the optimal TiO_2 concentration for pervious concrete containing recycled aggregate, as shown in Fig. 5. When the immersion concentration is below 0.3 wt%, the hydration products generated during the hydration process will wrap around the cement particles together with TiO_2 particles. Then, as the concentration approaches 0.3%, the TiO_2 particles and hydration products located in the hydration layer will gradually form spherical hydrates driven by the TiO_2 core and release into the liquid phase. With further increase in the immersion concentration, ettringite and spherical hydrates will be present on the surface of the cement particles. However, it is worth noting that this process consumes TiO_2 particles, which will lead to a reduction in photocatalytic effectiveness. Consequently, the study determined that the optimal immersion concentration was 0.3%.

On the other hand, Rachel et al. [56] found immersion can effectively enhance the photocatalytic performance of mortar, mainly due to the presence of ionic materials that inhibit charge recombination. Simultaneously, the study pointed out that carriers such as glass, red brick or cement can affect photocatalytic activity. Although Guo et al. [57] highlighted that combining spraying and compaction methods can effectively reduce the TiO_2 dosage and lower costs, they noted that TiO_2 particles on the sample surface are vulnerable to environmental factors and wear, which significantly diminishes the photocatalytic performance. This is mainly attributed to the negligible binding force between the TiO_2 particles and the cement material.

The spraying method involves precisely mixing the photocatalyst (usually TiO_2 nanoparticles) in the concrete mixture, and then uniformly spraying it onto the surface using a specialized spraying technology. This method requires meticulous handling of material selection, mixing ratios, application consistency and curing to achieve optimal photocatalytic activity. When the sample is exposed to light, it triggers a photocatalytic reaction that degrades organic pollutants and enhances self-cleaning properties. Sirota et al. [58] heated and sprayed TiO_2 onto concrete substrates. This method ensures that the photocatalyst is evenly distributed and adheres to the surface, thereby enhancing its performance to a photocatalytic activity of 95%.

Kim et al. [59] confirmed that pretreating with a silicate solution before spraying can effectively enhance the durability and self-cleaning ability of concrete, mainly due to the synergistic effect produced after the silicate solution impregnation. However, there are some issues with the spraying method: i) Studies have shown that increasing the

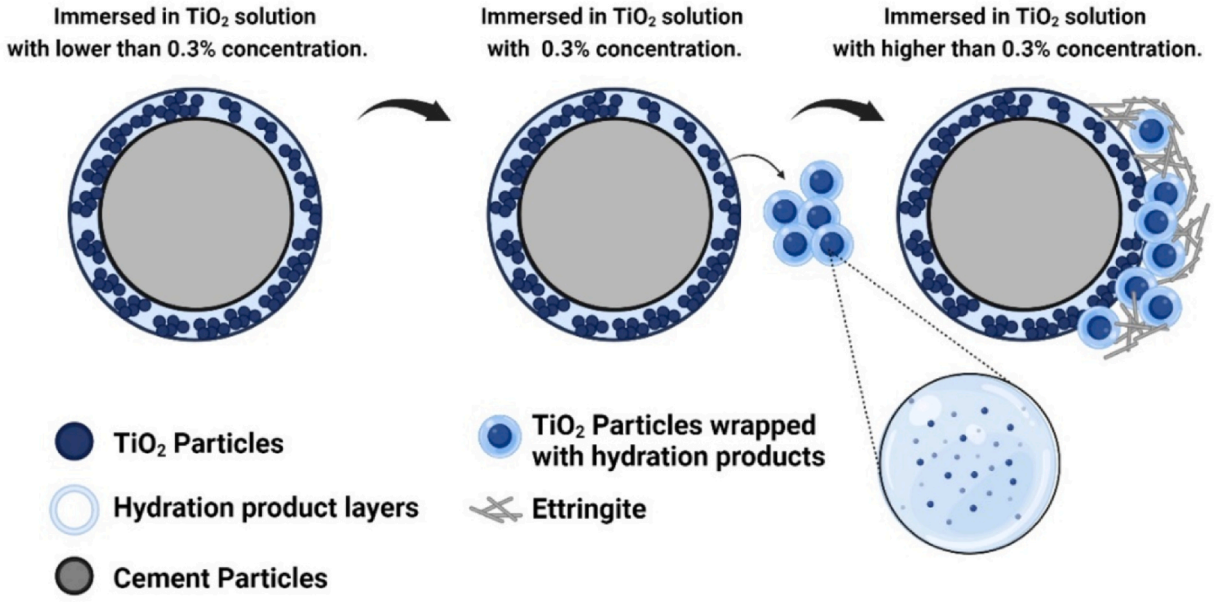


Fig. 5. Variation of hydration products under different TiO_2 concentration immersion [54,55].

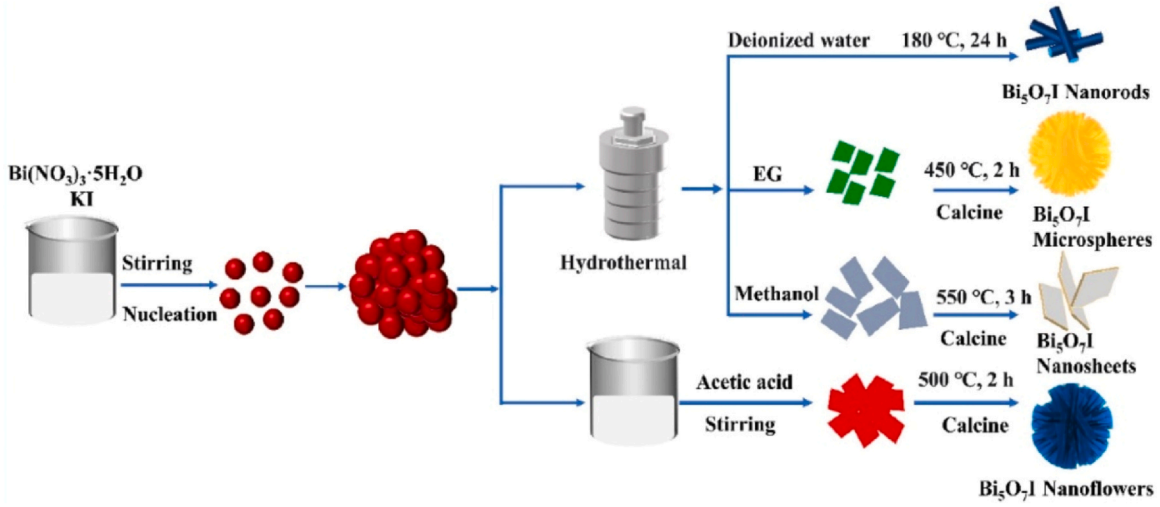


Fig. 6. Preparation of Bi_5O_7I of different morphologies [67].

concentration of TiO_2 nanoparticle slurry does not significantly improve photocatalytic efficiency, especially for TiO_2 nanocomposite concrete [60]; ii) Ordinary concrete coated with TiO_2, when exposed to natural environments, may have limited photocatalytic durability, as the TiO_2 can be easily washed away by rain and snowstorms; and iii) The spraying method poses toxicity and hazards to the environmental and worker health [61].

Compared to other methods such as spraying and soaking, the direct mixing method is more straightforward as it does not require any additional treatment of the samples. By directly mixing the photocatalyst into the sample, an integrated structure is formed, which helps to improve the bond strength between the photocatalytic material and the matrix. In fact, many studies have demonstrated that the synthesis of photocatalytic gel materials with good photocatalytic effects can be achieved by incorporating different types of TiO_2 nanoparticles into the gel materials.

A suitable amount of TiO_2 nanoparticles can significantly enhance the early hydration degree, mainly because these powders can provide nucleation sites for hydration products; nanoparticles help to refine the pore structure, thereby improving strength performance [62–64]; TiO_2 nanoparticles have a high surface area and can increase the water content of the mix to maintain the workability of the paste [65]. However, Calvo et al. [66] found that when expansive agents (ettringite-based (type-K) or CaO-based (type-G)) and photocatalysts (TiO_2 nanoparticles) are mixed to prepare photocatalytic expansive concrete, the workability of the paste is reduced but does not change the mechanical properties, while refining the microstructure. Therefore, photocatalysts might affect the workability of cementitious materials, such as fluidity and strength. Additionally, ensuring a uniform distribution of the photocatalyst in the cementitious materials during the preparation process is a challenge. An uneven distribution of the photocatalyst may lead to poor photocatalytic performance in some areas, affecting the overall effect.

In summary, each preparation method has its advantages and limitations. The mixing method provides long-lasting photocatalytic activity but is more costly and may impact the workability of cementitious materials when additional additives are present, an aspect that lacks research. The spraying method can achieve high photocatalytic efficiency with a smaller amount of TiO_2 but may not maintain long-term photocatalytic activity. The soaking method attempts to find a balance between cost-effectiveness and performance. Therefore, selecting an appropriate method requires a comprehensive consideration of specific application needs and resource constraints. In future research, extending the lifespan of photocatalytic activity and enhancing the bonding strength between the coating and the substrate will be key research directions to better exploit the potential of TiO_2 photocatalysts in cement-based materials.

2.3. Surface modification

Surface modification is instrumental in augmenting the photocatalytic effect and enhancing the durability of concrete against weathering. This enhancement can be realised through the integration of efficacious photocatalysts or by refining the surface architecture. Such modifications increase the number of active photocatalytic sites and accelerate reaction rates, thereby significantly boosting air purification and the self-cleaning capabilities of photocatalytic concrete. When exposed to outdoor environmental conditions, including UV radiation, rainfall and wind erosion, photocatalytic activity can diminish over time. However, surface treatment can substantially improve the material resistance to these elements, thereby decelerating its ageing process and sustaining its photocatalytic efficacy over an extended period.

The photocatalytic activity of a single photocatalyst is limited and has a relatively poor conversion efficiency with solar light. Therefore, the photocatalytic performance can be further enhanced through methods such as surface modification and elemental doping. Currently, new catalysts such as Bi_5O_7I, known for their stability and excellent photocatalytic activity, are considered ideal for use in marine construction structures. However, challenges such as low visible light absorption, inefficient electron-hole pair transmission and poor binding of photocatalyst particles in powder form to concrete structures impede their application in this field. Hence, modifications are necessary to successfully incorporate them into various construction structures.

Wang et al. [67] synthesized Bi_5O_7I photocatalysts with various morphologies, as shown in Fig. 6. Their results demonstrated that marine concrete prepared using the modified Bi_5O_7I photocatalysts of flower-like microsphere could degrade organic matter by up to 100% under visible light irradiation within 90 minutes, showing the highest photocatalytic activity during this period, along with high stability. Additionally, previous studies have always shown weaker binding strength between the cementitious carrier and TiO_2 [68–70]. Therefore, to broaden the methods of different cementitious material carriers, Liao et al. [71] first used recycled concrete as the primary carrier to synthesize recycled concrete/TiO_2 composites with high photocatalytic activity, with the microstructure of some samples shown in Fig. 7. According to the elemental analysis of Spot 1, a large amount of flocculent TiO_2 was found to cover the cement particles. Moreover, the hardened cement particles can provide numerous nucleation sites for anchoring these TiO_2 particles. Therefore, these results are

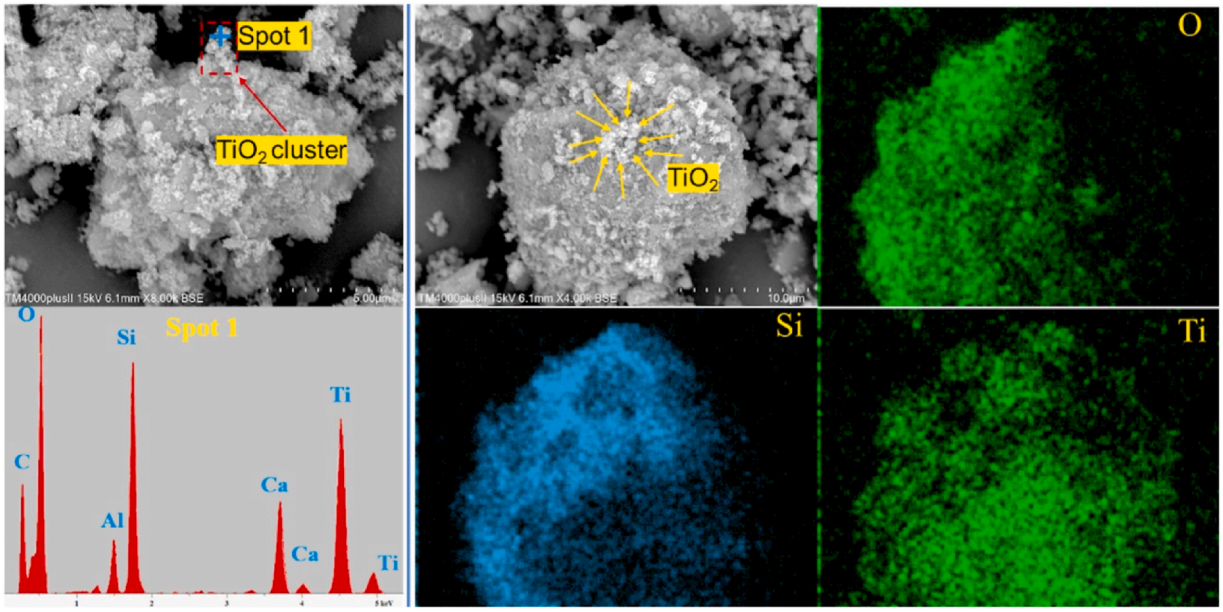


Fig. 7. Microscopic characterization of recycled concrete/TiO_2 composites by means of modification techniques [71].

encouraging, providing an experimental basis for the composite approach with different materials.

In recent years, researchers have developed various modification techniques to enhance the properties of photocatalysts. These include aerogel modification, which ensures stable photon efficiency and offers low dissolution rates alongside excellent self-cleaning properties [72]; mixed crystal hetero-enhancement, a method that synthesises photocatalysts with greater thermodynamic stability and increased activity by combining different crystal faces [73–75]; and chemical vapour deposition coupled with thermal annealing. The latter process not only improves photocatalytic activity but also facilitates the phase transformation from anatase to rutile [76].

However, these methods for improving cement-based photocatalytic materials face several obstacles, including their technical complexity, high cost and doubts about their long-term stability and effectiveness in practical applications. Studies [77,78] indicate that the photocatalytic activity of the modified TiO_2 might even be inferior to pure TiO_2 due to issues with carrier fixation; certain modification methods may encounter challenges in long-term stability in practical use, as the low adsorption capacity of cement materials can lead to weak binding between the photocatalyst and the cement matrix [79]. Therefore, these issues need to be addressed through further improvements and research to promote the wider application of photocatalytic cement-based materials.

2.4. Method for measuring photocatalytic properties

In the domains of sustainable development and energy transition, understanding the properties of photocatalytic cementitious materials is fundamental to both scientific research and technological advancements. This section outlines standard methods for assessing the properties of photocatalytic cementitious materials, covering their photocatalytic activity, self-cleaning properties and degradation of environmental pollutants. In Fig. 8(a), photocatalytic activity is characterized by the degradation rate of the material surface to a specific pollutant gas under UV light irradiation. Prior to the experiment, the samples must be pre-treated with UV light to remove organic matter, followed by dark state measurements of the target gases until equilibrium is reached. The photocatalytic efficiency of the samples was evaluated using an ISO flow-through reactor, based on the change in gas concentration before and after the reaction [80].

For the self-cleaning performance test, the pink organic dye Rhodamine B (RhB) was chosen as a test substance according to the Italian standard UNI 11259, and the photocatalytic self-cleaning ability of the material was assessed by its degradation level. During the test, RhB was uniformly coated on the sample surface and exposed to UV irradiation to observe how the photocatalyst triggered the generation of electron-hole pairs and reactive oxygen radicals $\cdot O_2^-$ and ·OH, leading to the degradation of RhB molecules pre-adsorbed on the cementitious material surface, as shown in Fig. 8(b). Further degradation of RhB under visible light irradiation occurs as the photocatalyst serves as an electron transfer medium, resulting in notable changes in the colour and reflectance of the sample surface [81–83]. Therefore, the self-cleaning performance of the sample can be characterized by the change in colour or reflectance of the sample surface.

Conversely, the test for the degradation capability of environmental pollutants is depicted in Fig. 8(c). It involved immersing the samples in an organic solution, such as methyl blue, and measuring the absorbance with a spectrophotometer to establish the relationship between the solution concentration and its absorbance. The degradation rate of the pollutant is calculated by comparing the difference in solution concentration before and after the experiment [84]. In conclusion, the above set of test methods provides a scientific basis for evaluating the functionality of photocatalytic cementitious materials in environmental applications.

3. Efficiency of photocatalytic concrete in purifying pollutants

3.1. NO_x removal rate and influencing factors

In photocatalytic concrete products, the degradation mechanism of NO_x is mediated by photocatalysts, which facilitate the interaction between NO_x and photo-induced electrons alongside reactive oxygen species. Fig. 9 illustrates the process of purifying NO_x in photocatalytic concrete. This process can be divided into three main steps: (i) the adsorption and transfer process of pollutants; (ii) the excitation of the photocatalyst and the degradation of pollutants; and (iii) the release of purified products. NO_x is absorbed due to the porous nature of the concrete structure. Then, the photocatalyst added to the concrete is excited by UV irradiation, leading to electron excitation. These excited electrons form high-energy electrons capable of reducing other substances; the photo-excited electrons react with oxygen molecules (O_2),

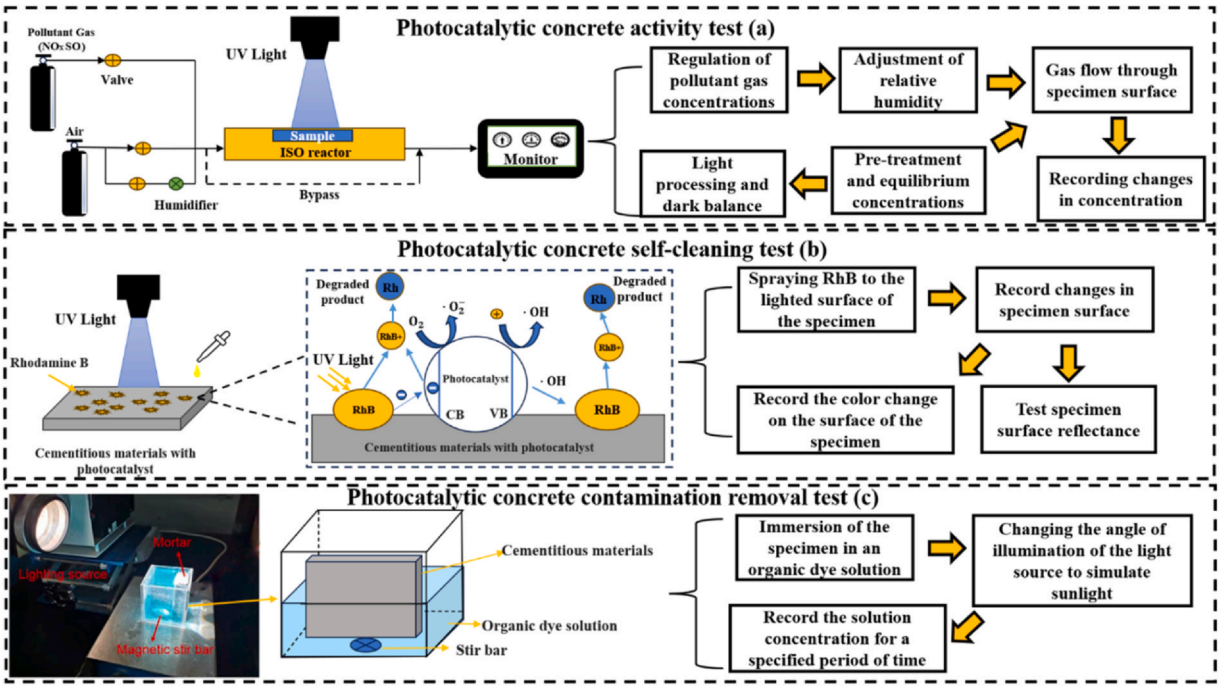


Fig. 8. Methods used to measure the properties of photocatalytic cementitious materials: (a) testing photocatalytic activity; (b) self-cleaning ability; (c) pollutant handling [80,82–84].

producing reactive oxygen species, superoxide radicals and hydroxyl radicals. These reactive oxygen species, with strong oxidizing properties, participate in the NO_x. Subsequently, the photocatalytic reaction produces compounds such as nitrates and nitrogen, which, while having environmental and human health impacts, are less harmful than nitrogen oxides [85,86]. The chemical equations of the above photocatalytic reactions are given by Eq. (15–18) [85].

$$NO + OH^- \rightarrow HNO_2 \quad (15)$$

$$HNO_2 + OH^- \rightarrow NO_2 + H_2O \quad (16)$$

$$NO_2 + OH^- \rightarrow NO_3^- + H^- \quad (17)$$

$$2NO_2 + H_2O \rightarrow HNO_2 + HNO_3 \quad (18)$$

The above reactions can effectively convert NO_x in the atmosphere into less harmful substances, and this technology has potential environmental and health benefits for improving urban air quality, reducing the concentration of harmful gases in the air, and mitigating adverse effects on human health and the environment. However, the efficiency of degradation has always been a focal point of research. Degradation efficiency is influenced by several factors: (i) The type of photocatalyst is a key factor, as it determines the active sites for the reaction and the light absorption capacity [87–89]; (ii) The color of the concrete can indirectly affect the degradation efficiency of NO_x in photocatalytic concrete; and (iii) Different mixtures in the concrete matrix affect efficiency, as they influence the contact and reaction between the photocatalyst and the matrix. At the same time, the presence of temperature and humidity can affect photocatalytic efficiency [90–92]. Chen et al. [93] used different types of recycled glass extracted from broken waste beverage bottles. This recycled glass was used to replace sand in the preparation of photocatalytic concrete. The photocatalytic activity of concrete using recycled glass as aggregate was significantly higher than that of traditional concrete prepared with natural sand, as shown in Fig. 10.

The photocatalytic activity of concrete prepared with various types of recycled glass demonstrated significant variability, primarily due to the translucency of the glass used. Furthermore, the study indicated that variations in the particle size range of the aggregates did not

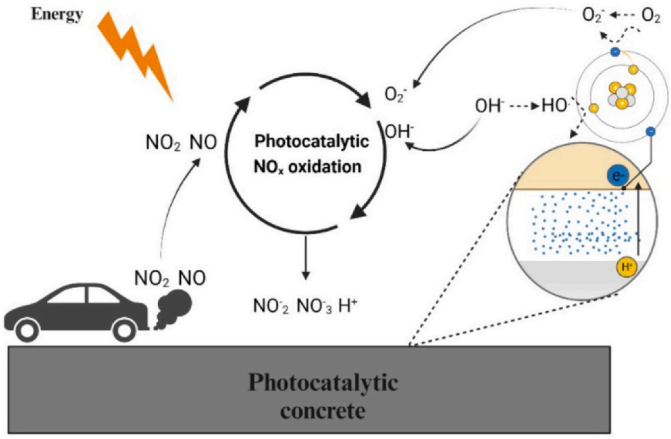


Fig. 9. Photocatalytic concrete degradation mechanism of NO_x [85,86].

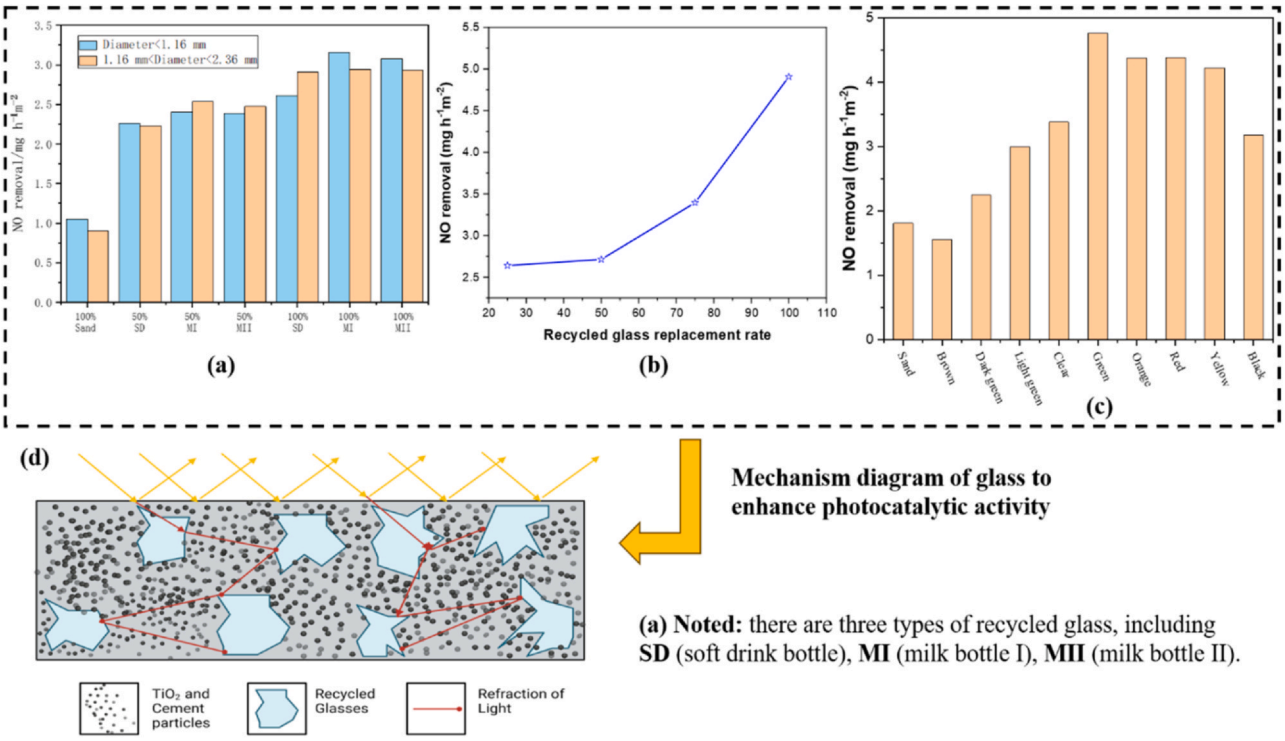


Fig. 10. Influence of recycled glass on NO removal rates from photocatalytic concrete: (a) effect of different types of recycled glass; (b) effect of substitution rate; (c) effect of different colored glass; (d) Mechanism diagram [93,94].

influence the photocatalytic activity. Guo et al. [94] studied the effect of the replacement rate of recycled glass for natural sand (25–100%) on photocatalytic activity, with results shown in Fig. 10 (b). Using 100% recycled glass instead of sand achieved an NO removal rate of 4.90 $mg{\cdot}m^{-2}$ h^{-1}. In comparison, a sample with a 25% replacement rate showed a lower NO removal rate of only 2.67 $mg{\cdot}m^{-2}h^{-1}$, indicating that the translucency of glass is beneficial for enhancing photocatalytic performance.

It is believed that in Fig. 10 (d) the dispersion of glass particles in the concrete matrix can promote the scattering and reflection of light. This can increase the propagation of light within the concrete, improve the utilization of light by the photocatalyst, enhance the photocatalytic reaction, and thereby help to increase the removal rate of NO. On the other hand, the color of the glass plays an important role in the removal of NO in photocatalytic concrete. In Fig. 10(c), the efficacy of different colored glasses in NO removal varies significantly. Darkly pigmented glasses (green, orange, red) exhibit lower scattering abilities and greater absorption capacities across the entire solar spectrum. In contrast, light-colored pigments, such as white, reflect more light and thus enhance the intensity of illumination which can increase photocatalytic activity. Multicolored pigments offer a compromise, balancing light absorption and reflection. However, the specific characteristics of the pigments and the catalysts must be jointly considered to optimize photocatalytic efficiency effectively. Therefore, the choice of color should be comprehensively considered in conjunction with the optical properties of the pigments and the needs of the catalytic reaction to achieve optimal results.

On the other hand, photocatalytic concrete in the process of NO_x removal is comprehensively influenced by key factors such as initial NO concentration, relative humidity, and gas flow rate. Si et al. [95] investigated how the rate of NO_x removal is influenced by the initial concentration of NO, the relative humidity, and the rate at which the gas flows. The results in Fig. 11 indicate that a higher initial NO concentration helps increase the reaction rate. Relative humidity plays an important role in the photocatalytic removal of gaseous pollutants [96,97], and an increase in relative humidity is not conducive to the NO_x removal reaction. Optimization of the gas flow rate helps improve the effective contact between NO_x and the catalyst. Following this, Poon et al. [98] conducted further studies on factors such as porosity, cement content, different particle sizes of aggregates and curing time of the blocks. This study demonstrated that porous materials composed of recycled aggregate can achieve enhanced performance. Reducing the cement content increases the NO removal rate due to a decrease in the available surface area, which occurs as cement particle voids are filled. Furthermore, an extended curing period results in a reduction in the NO removal rate; this is attributed to a decline in the number of active sites as ongoing hydration and carbonation lead to pore closure. Finally, although increasing the TiO_2 fraction within a 10% range appears to enhance the NO removal rate, this observation requires further verification through future research.

Currently, in the field of photocatalytic concrete, there are some experimental gaps regarding factors affecting the NO_x removal rate. Although previous studies have explored the effects of factors such as initial NO concentration, relative humidity and gas flow rate on the removal rate, more in-depth experiments are needed to fully understand the interactions and optimization conditions among these factors. Particularly, in-depth research is awaited in aspects such as the catalytic reaction mechanism under high initial NO concentrations, changes in catalytic performance under different relative humidities, and catalyst efficiency under different gas flow rates. Filling these experimental gaps will help further enhance the performance of photocatalytic concrete systems and provide more effective NO_x purification solutions for environmental management.

3.2. *CO_2 removal rate and influencing factors*

Photocatalytic cementitious materials actively contribute to the reduction of atmospheric CO_2 by utilizing sunlight. When exposed to natural light, these materials catalyze the conversion of carbon dioxide into useful by-products, including carbonates and various organic compounds. Richter et al. [99] summarized the process of CO_2 conversion in photocatalysis in Eq. 19. The study examined the

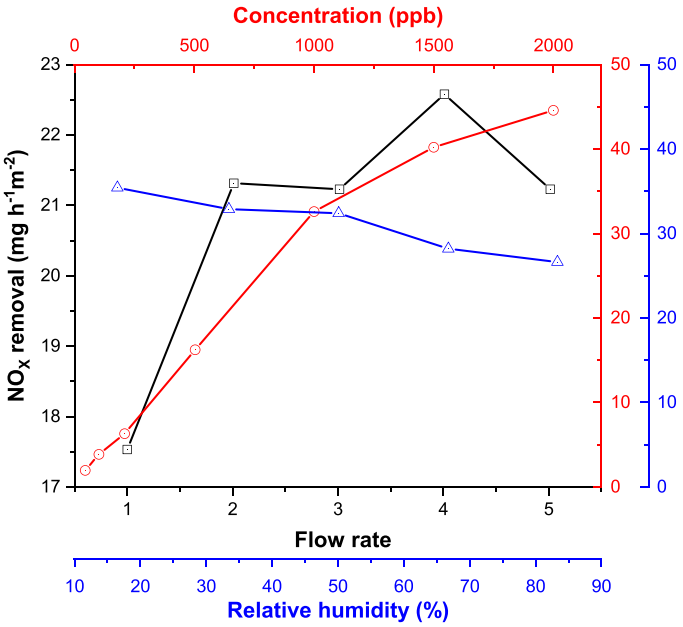


Fig. 11. Effect of different external factors on NO_x removal rate [95].

contribution of the photocatalytic process to the formation of various carbonates and organic compounds, including CH_4, H_2 and CO, alongside more complex hydrocarbons. Here, advanced hydrocarbons specifically denote molecules containing more than five carbon atoms. These products are crucial for offering carbon-neutral solutions and have the potential to serve as significant alternative energy sources to fossil fuels. In this way, the photocatalytic process not only demonstrates the ability to convert carbon dioxide in the environment into useful chemicals and fuels but provides an innovative way to reduce dependence on traditional fossil fuels and alleviate the problem of global warming.

$$CO_2 + H_2O \xrightarrow{h\upsilon} \text{Carbonaceous products} + O_2 \quad (19)$$

Awadalla et al. [100] investigated how the principle of photocatalysis can be used to reduce CO_2 pollution in buildings through incorporating TiO_2 into composite concrete panels. The research focused on understanding the factors that affect the performance of composite concrete panels, such as the porosity of the panels, the different types of waste and the amount of TiO_2 used in the mix design. The performance of these photocatalytically active concrete products under laboratory conditions was assessed by chemiluminescence analysis and it was found that the ability to photodegrade CO_2 was closely related to the porosity of the samples: the higher the porosity, the greater the ability to remove CO_2. Similar to the NO_x removal mechanism, porous materials such as recycled aggregate are beneficial for increasing the CO_2 removal rate. Therefore, we believe that incorporating porous materials such as zeolite and recycled aggregate in cement-based materials would enhance photocatalytic efficiency, primarily due to the adsorption effect and the provision of more nucleation sites. Furthermore, the study indicates that the porosity of the matrix is directly proportional to the CO_2 removal rate.

In fact, previous research has shown that forming a porous structure in cement-based materials is advantageous for the progress of photocatalytic reactions [101]. Wang et al. [102] confirmed that cement-based materials with a porous structure significantly enhanced photocatalytic performance. Additionally, Yang et al. [103] found that the density significantly impacted the agglomeration or dispersion of TiO_2 particles; moreover, pore distribution within 50 nm significantly affected photocatalytic performance. However, it is worth noting that current research on photocatalytic concrete for carbon dioxide removal is relatively scarce, with most studies focusing on CO_2 capture and adsorption. While it is important to cut down carbon dioxide emissions, recent studies seem to focus on tackling other environmental issues. For example, current research is increasingly exploring the self-cleaning properties of materials and their ability to remove nitrogen oxides through photocatalysis, which seems to be receiving more attention than the removal of carbon dioxide.

3.3. SO_2 removal rate and influencing factors

The reaction process of SO_2 degradation via photocatalytic principles is outlined in Eqs. (20) to (22). Upon exposure to light, SO_2 molecules are adsorbed by the photocatalyst and react with free radicals, initially forming HSO_3. This intermediate then reacts with O_2 to produce SO_3, which subsequently combines with atmospheric moisture to form sulphur-containing compounds such as H_2SO_4. These products are readily removable from the environment, thereby reducing SO_2 pollution [104].

$$SO_2 + OH\cdot \rightarrow HSO_3 \quad (20)$$

$$HSO_3 + O_2 \rightarrow SO_3 \quad (21)$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad (22)$$

Lee et al. [104] developed a nano-SiO_2-TiO_2 photocatalyst by coating TiO_2 with a support (SiO_2) using the sol-gel method. They evaluated the environmental performance of concrete blocks containing this photocatalyst and found that the prepared concrete specimens could achieve a 14.4% reduction in SO_2 concentration. The study confirmed that an increase in UV intensity and solar radiation had a positive effect on reducing SO_2 concentration. In addition to light intensity, several parameters can significantly affect the photocatalytic process of SO_2 removal. These parameters include relative humidity and the presence of different gases in the media environment, such as NO_x [105]. The degradation of SO_2 during the photocatalytic process involves both chemical reactions facilitated by the photocatalyst and physical adsorption processes that occur simultaneously with the catalysis.

In addition to chemical reactions, physisorption processes occur when SO_2 molecules meet the surface of the cementitious material. These molecules can physically adhere or adsorb to the surface or pores of the cementitious material without an immediate chemical transformation. According to Krishnan et al. [106], 40% of the concentration

can be removed by the photocatalytic reaction in a silicate-TiO_2 system. The article highlighted that ~30% of SO_2 is removed through physical adsorption. Fernandes et al. [107] investigated the role of airborne SO_2 in the adsorption and reaction process of photocatalytic cementitious materials. The study found that SO_2 molecules adhered to the surface of the cement mortar through adsorption. Sulphate is generated through a redox reaction with reactive ions, such as OH·and O_2^-, resulting in the formation of SO_3^{2-} and SO_4^{2-}. Additionally, the study showed that increasing the porosity of cementitious materials was effective in removing SO_2.

In the field of SO_2 removal or degradation using photocatalytic concrete, previous scholars have developed novel materials such as SiO_2-TiO_2 nanoparticles. These materials have demonstrated significant efficiencies in SO_2 reduction and are also sensitive to variations in UV intensity and different gaseous environments. Additionally, research has shown that parameters such as relative humidity significantly affect the photocatalytic process. Meanwhile, scholars have confirmed the symbiotic relationship between chemical reactions and physical adsorption in the process of removing SO_2. They have revealed that enhancing the porosity of cementitious materials can improve efficacy. However, research on the complex mechanisms, chemical kinetics and influencing factors of photocatalytic SO_2 removal in the context of cementitious materials is lacking.

4. Mechanical properties of photocatalytic concrete

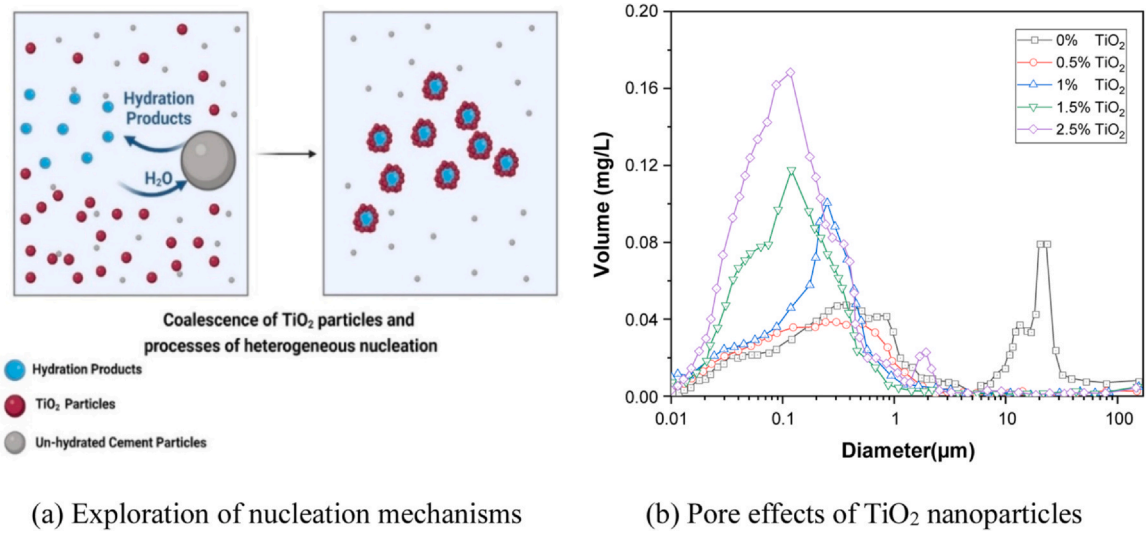
4.1. Effect of photocatalytic materials on the cement matrix

Cement plays a vital role in concrete by creating a strong cementitious matrix through a hydration reaction that produces a gel. This matrix is tightly bound to the aggregates, providing concrete with superior mechanical strength. Therefore, an in-depth understanding of the photocatalyst-cement interaction is crucial for the mechanical properties of photocatalytic concrete. The interaction directly affects the mechanical properties of photocatalytic concrete, determining its robustness and long-term reliability in environmental and engineering applications. This work takes the commonly used TiO_2 nanoparticle photocatalyst as an example to discuss its effects on the cement matrix. When mixed with cement in Fig. 12 (a), TiO_2 nanoparticles initially facilitate the hydration process, leading to the formation of hydration products. Subsequently, TiO_2 nanoparticles can act as high-quality heterogeneous nucleating agents. They can agglomerate and bind with hydration products and form a dense coating on the surface of the hydration products. Lastly, due to their high specific surface area, the nanoparticles can provide stable crystal nuclei, which is conducive to the rapid growth and accumulation of hydration products [108]. Therefore, TiO_2 nanoparticles accelerate the hydration process of cementitious materials, especially in the early stages [109].

Nazari et al. [111] demonstrated through a series of tests, such as isothermal calorimetry, thermogravimetric analysis and X-ray diffraction, TiO_2 nanoparticles help promote the hydration process of the cement matrix, namely accelerating the formation of C-S-H gel. On the other hand, apart from the role of TiO_2 nanoparticles in promoting cement hydration, previous studies [112–114] have proven that ultrafine particles, whether inert or reactive, have a good promoting effect. The mechanism of these particles is similar to that discussed for TiO_2 nanoparticles, involving heterogeneous nucleation and agglomeration effects; and these studies conclude that smaller particle size and larger specific surface area can provide more sites for the deposition of hydration products.

However, current research predominantly concentrates on the promotion of hydration in cement matrices by TiO_2 nanoparticles. Notably, TiO_2 not only provides nucleation sites but also exhibits pozzolanic activity, unlike other nanomaterials such as nanosilica. This pozzolanic activity significantly enhances the hydration process[115,116]; however, according to the results discussed previously, TiO_2 nanoparticles can only act as a stable crystal nucleus to provide nucleation sites for hydration products. Therefore, future research should focus more on the modification of TiO_2 nanoparticles. Although some scholars have successfully synthesized SiO_2-TiO_2 [117], this area of research is scarce.

Lucas et al. [110] investigated the effect of TiO_2 nanoparticles on the microscopic pore size of cementitious materials. Fig. 12 (b) shows that increasing the TiO_2 fraction in cementitious materials refines the pore size. This study found a combined effect of pore size reduction and porosity increase for higher TiO_2 content (5 wt%). It is important to note that high porosity does not necessarily result in higher photocatalytic activity. However, large pores can have a negative effect as pollutants may be adsorbed into the matrix, preventing further light transmission in the cement matrix. According to Şahin et al. [118], TiO_2 nanoparticles have a relatively small particle size which provides better particle size distribution, ensuring pore size refinement and reduced particle build-up. This makes TiO_2 nanoparticles useful for reducing porosity and improving microstructure. It highlights the efficacy of nanoscale materials with high specific surface area in providing heterogeneous nucleation sites. Furthermore, the study found that the addition of polycarboxylic ether-based high-efficiency water-reducing agents and polyacrylic acid, as well as ultrasonication of the slurry, aided in achieving a homogeneous distribution of TiO_2 nanoparticles in



(a) Exploration of nucleation mechanisms (b) Pore effects of TiO_2 nanoparticles

Fig. 12. Pore regulation and nucleation mechanism of cementitious materials with TiO_2 nanoparticles [108] [110].

the cement matrix. This homogeneous dispersion in the cement system prevented agglomeration and the formation of weak points.

These afore-mentioned studies have demonstrated that TiO_2 nanoparticles can contribute to the gelling of materials by forming dense coatings as heterogeneous nucleation carriers. Specifically, heterogeneous nucleation refers to the introduction of an external substance into a material to facilitate the formation and growth of a new phase. When TiO_2 nanoparticles interact with other components in the gelling material, they can form a dense coating on the surface. This dense coating has several functions. It increases the structural stability of the gelling material, making it more durable. Secondly, the coating provides an extra layer of protection against external factors such as water and chemicals. This accelerates the growth and accumulation of hydration products, particularly in the early stages of hydration. However, it is important to note that it cannot provide additional cementitious products through particle activity, unlike other volcanically active nanoparticles. Instead, it can only provide stable nuclei based on more nucleation sites for hydration products. It has been emphasized that the increase in cement strength due to TiO_2 nanoparticles is mainly caused by the decrease and change in the orientation index of the nucleation function, rather than the increase in the amount of hydration products [119]. Further attention is necessary for the modification of TiO_2 nanoparticles. Although successful cases of SiO_2-TiO_2 synthesis exist, research into other nanomaterials remains limited.

In addition, studies on microscopic pore size have shown that the use of TiO_2 nanoparticles can refine the pore size of the cement matrix. It is important to note that a high fraction of TiO_2 nanoparticles may lead to a decrease in pore size and an increase in porosity. A high porosity does not necessarily correlate with a high photocatalytic rate. Furthermore, the packing and defect bridging effect of TiO_2 nanoparticles results in a more desirable particle size distribution. This, in turn, leads to a reduction in porosity and an improvement in microstructure. Additionally, the uniform distribution of TiO_2 nanoparticles in the matrix can be achieved using efficient water reducers, polyacrylic acid and ultrasonication, thereby preventing agglomerates and weak points. In conclusion, these studies demonstrate the beneficial effect of TiO_2 nanoparticles on cement hydration and suggest further research on diverse photocatalytic nanomaterials, such as SiO_2-TiO_2 synthesis, to improve their efficiency and performance.

4.2. Effect of photocatalytic materials on mechanical properties of concrete

4.2.1. Compressive strength

Cement and concrete systems are closely related. The previous chapters have explored the role and effect of photocatalysts on the cement system, which acts as a binder for the concrete substrate. The composition of cement mainly consists of compounds such as tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A) and tetra-calcium ferro-aluminate (C_4AF). On the other hand, the concrete system is a composite material that comprises a mixture of cement, aggregates (such as sand and gravel), water and admixtures. This section expands on the previous discussion of the photocatalytic cement and delves into the mechanical properties of the concrete system when photocatalysts are present. The inclusion of photocatalytic materials, particularly TiO_2 nanoparticles, can impact the compressive strength of the concrete in several ways. The development of compressive strength can be significantly affected by different classes of photocatalysts, incorporation ratios and mixing of modified photocatalytic materials. Table 1 summarizes recent studies carried out on how photocatalysts affect concrete strength.

Sorathiya et al. [122] analyzed the effect of TiO_2 nanoparticles on conventional M20 concrete at different dosages (0.5–1.5 wt%) and found that TiO_2 significantly increased the compressive strength of the concrete, especially at 1.0 wt%. Above this percentage, the strength of the concrete decreased, mainly because too many TiO_2 particles reduced the formation of C-S-H gels and affected the compatibility of the

Table 1
The compressive strength of photocatalytic concrete and its influencing factors.

Refs.	Research content	Influencing factors	Compressive strength (MPa)		
			3 days	7 days	28 days
[120]	Impact of 3 different types of TiO_2 on the mechanical properties of concrete	The 3 types of TiO_2: anatase I, anatase II and rutile, with contents of 3%, 6% and 10% in mass	24–30	32.5–36.5	40–48
[121]	Effect of TiO_2 content on mechanical properties of concrete	The variation of TiO_2 content (0–3 wt%)	26–33; 33–37 (with plasticizer)	32–40; 42–47 (with plasticizer)	38–52; 55–62 (with plasticizer)
[122]	The impact of anatase TiO_2 nanoparticles with a low mix ratio on conventional concrete of M20 grade	The variation of TiO_2 content (0.5–1.5 wt%)	—	31.8–36.6	32–38
[123]	An assessment of mechanical properties on self-cleaning concrete with rutile TiO_2	TiO_2 with percentages of 0.50–3.50 wt% of the total weight of the cement	—	14–20	20–28
[124]	Performance of photocatalytic concrete blended with M-Sand, POFA and TiO_2	TiO_2 (2–10 wt%) replacing cement; Use of palm oil fuel ash (POFA) as a binder in place of cement (4–20 wt%);	9–12 (TiO_2); 10–11.5 (POFA)	17–21 (TiO_2); 19–24 (POFA)	26–33 (TiO_2); 29–34 (POFA)
[125]	Examining the impact of TiO_2 nanoparticles and ZnO_2 nanoparticles on the mechanical characteristics of concrete.	The dosages of TiO_2 and ZnO (0–2.5 wt% and 0 – 3 wt%) replacing of the weight of the cement	—	49–56 (TiO_2); 21–37 (ZnO)	50–58 (TiO_2); 45–53 (ZnO)
[126]	Effect of TiO_2 nanoparticles on mechanical and electrical properties and microstructure of reactive powder concrete	The content levels of TiO_2 are 0.78 wt%, 2.32 wt% and 3.88 wt% by volume of cement	65–70	—	100–120
[127]	Reactive powder concrete reinforced with nano SiO_2-coated TiO_2	The SiO_2-coated TiO_2 was added in the amounts of 0, 1.0 wt %, 3.0 wt% and 5.0 wt% by cement weight.	60–70	—	100–112

material. Sokolnikova et al. [121] increased the TiO_2 content to 3 wt% and observed a significant increase in 28-day compressive strength at 1–2 wt% content, but no significant increase at 3%, which was attributed to the effect of TiO_2 on C_3S and its photocatalytic properties accelerating the hydration process. However, concentrations above 3 wt% resulted in lower strength due to uneven TiO_2 distribution and excessive $Ca(OH)_2$ consumption. Similarly, this finding is supported by the study [123], which found that TiO_2 at above 3.0 wt% had a negative effect on the compressive strength of concrete, while TiO_2 below 1.0 wt % improved the workability of concrete. Li et al. [126] investigated the effect of TiO_2 nanoparticles on activated powder concrete and found that they promoted early hydration and increased 28-day compressive strength. On the other hand, Melo et al. [120] studied the effect of different types of TiO_2 (anatase I, II and rutile) on the concrete strength. Anatase II or rutile type TiO_2 nanoparticles showed improved photocatalytic properties, mechanical properties and durability. However, anatase type I negatively affected the strength properties of concrete due to its propensity to agglomerate during preparation. This aggregation hindered uniform distribution within the cement matrix, thereby diminishing its potential benefits.

However, in a study by M. Mostafa et al. [125], the mechanical properties of photocatalytic concrete were compared using two different classes of photocatalysts: TiO_2 nanoparticles and ZnO nanoparticles. It was found that TiO_2 nanoparticles promote the early hydration process and reduces the crystallization rate of calcium hydroxide (CH). The study indicated an optimum 2.5 wt% for the incorporation of TiO_2 nanoparticles, which agrees with previous results. However, the optimum percentage for ZnO incorporation is only 1.0 wt %, making it less than ideal for the development of concrete strength. This depletion of necessary factors for gel product formation is mainly attributed to its reaction with CH during the early stages of splashing.

Currently, the modification of TiO_2 catalysts to incorporate volcanic ash activity is a prevalent area of research, as illustrated in Fig. 13 (a). The Ti-O-Si bond can be formed at the interface [128,129]. Han et al. [127], the mechanical properties of reactive powder concrete were enhanced by the addition of SiO_2 coating on the surface of TiO_2 particles. The reinforcing effect of this SiO_2 coating on concrete is primarily due to its reaction with CH in cement, which leads to the formation of more hydration products. The newly formed hydration products act on the cement matrix at the microscopic level, effectively enhancing the overall densification of the concrete by promoting nucleation between cement particles and increasing fillers. As a result, voids in the cement structure were filled and the number of harmful pores was reduced, which in turn significantly increased the strength of the concrete to 111.75 MPa. This approach involved modifying the surface of TiO_2 particles, which not only enhanced the structural integrity of the cement but also improved its durability and load-bearing capacity in Fig. 13 (b).

TiO_2 nanoparticles significantly enhance the mechanical properties of concrete, with moderate additions shown to improve compressive strength. It is recommended that the admixture should be less than 3.0 wt% of the mass fraction of cement in the matrix. Some studies have highlighted the role of TiO_2 in promoting the early hydration process. However, excessive addition may adversely affect the workability and strength of concrete. Additionally, different types of TiO_2 particles have differential effects on concrete properties. TiO_2 nanoparticles in anatase II or rutile forms showed enhancement benefits, while the presence of agglomeration in the anatase I form reduced the uniform distribution. Finally, the modification of TiO_2 catalysts was introduced to improve the mechanical strength of concrete by forming Ti-O-Si bonds. In conclusion, the incorporation of TiO_2 in concrete necessitates meticulous control over the quantity used to fully capitalize on its beneficial effects and to account for its intricate impact on the microstructure and properties of concrete.

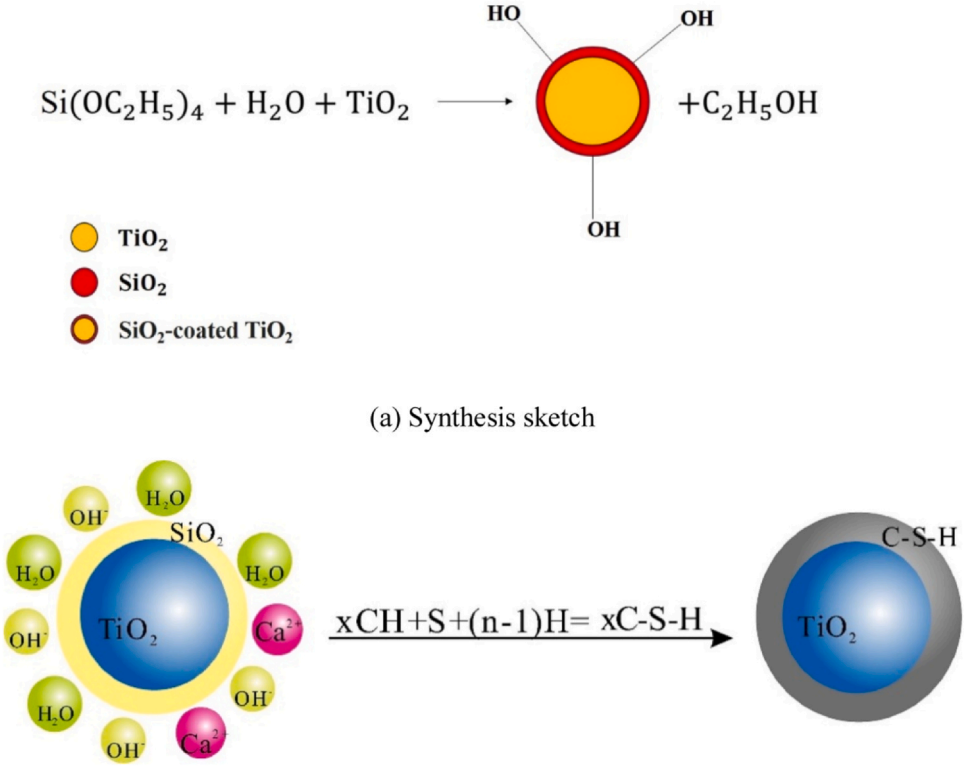


Fig. 13. Illustration of the SiO_2 coating process on TiO_2 particles and the resultant benefits for hydration enhancement [127,128].

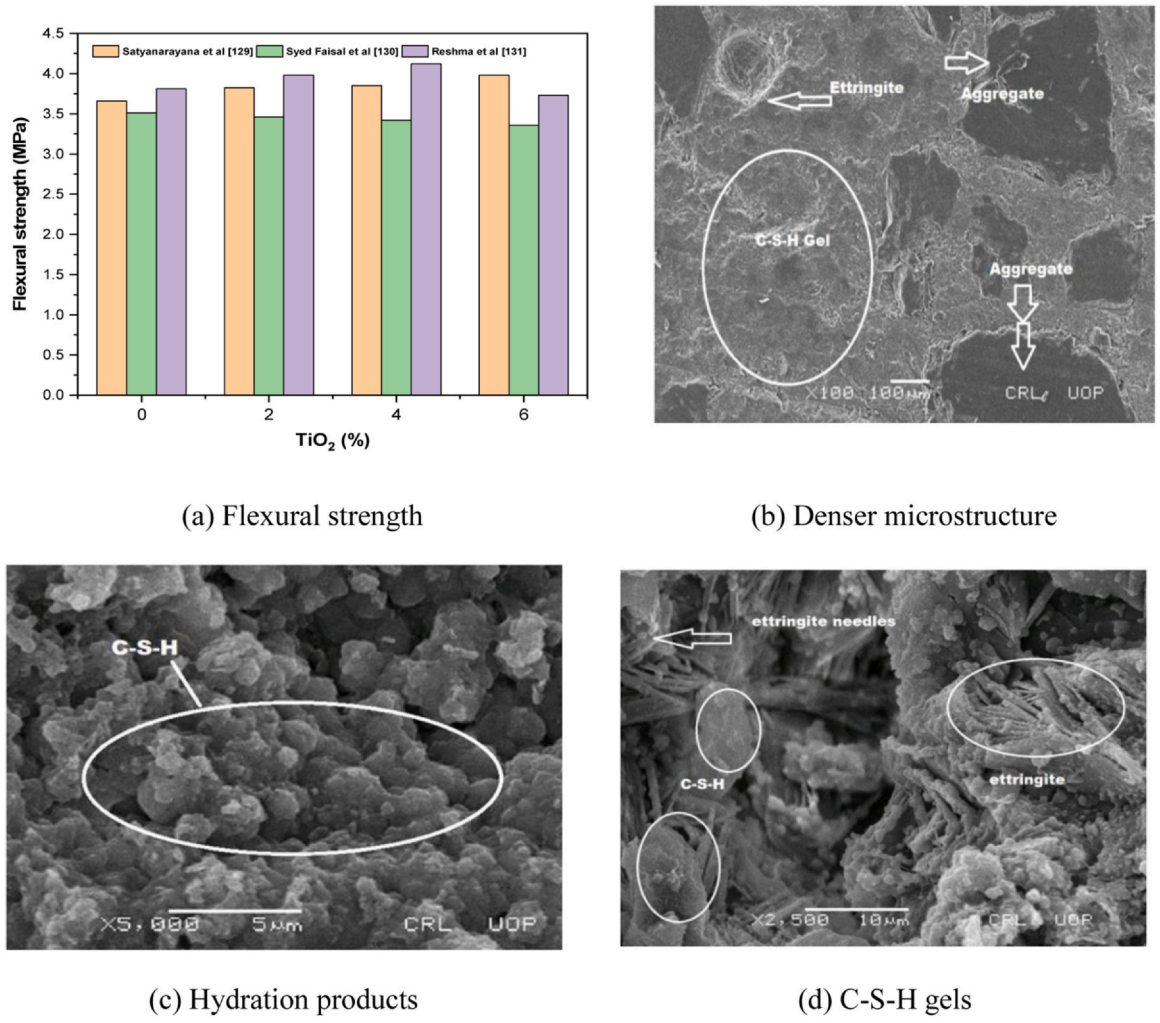


Fig. 14. Analysis of the effect of variation of TiO_2 content on microstructure and flexural strength of concrete [124,130,131] [134].

4.2.2. Flexural strength

Traditional concrete design and evaluation have predominantly focused on optimizing compressive strength, often at the expense of other mechanical properties such as flexural strength. Despite its critical importance, flexural strength has been relatively underexplored, particularly in the context of photocatalytic concrete. This section critically examines how photocatalysts, specifically TiO_2, influence the flexural strength of concrete, providing insights into recent performance outcomes.

In the study by Satyanarayana et al. [124], the effect of TiO_2 admixture on the flexural strength of concrete was investigated. The relationship between 28-day flexural strength and TiO_2 admixture is illustrated in Fig. 14 (a). The admixture ratios were all expressed as a percentage of the mass fraction of the cement. The study found that the optimum admixture was 10.0 wt%, resulting in a 28-day strength of 4 MPa. However, the mechanism of TiO_2 contribution to flexural strength was not discussed. According to Faisal et al. [130], TiO_2 did not have a positive effect on the flexural strength of M20 concrete. In fact, it caused a small negative effect. Reshma et al. [131] concluded that the optimal fraction of TiO_2 is 4 wt%. Exceeding this value negatively affects flexural strength. Previous research has demonstrated that crack initiation and extension have a significant impact on flexural properties [132,133]. Orakzai investigated the effect of TiO_2 on the microstructure of cementitious materials and found that the filling effect of nanomaterials and the promotion of the hydration process by TiO_2 resulted in good microstructures, as shown in Figa. 14 (b) to (d). The photographs showed a high degree of hydration, with numerous C-S-H gel products but no CH crystals [134]. Jabali et al. [135] demonstrated that an excess of TiO_2 can cause a deterioration of the cementitious pore structure.

Future studies should delve more deeply into the relationship between TiO_2 and flexural properties, elucidating the underlying mechanisms of contribution. Moreover, further investigations are needed into the impact of TiO_2 incorporation on the microstructure of concrete, including crack initiation and propagation, to comprehensively understand its role in enhancing concrete properties. Concurrently, exploring the use of alternative nanomaterials or different forms of TiO_2 could identify the optimal combination to improve the flexural strength of concrete.

5. Life cycle assessment of photocatalytic concrete

Life cycle assessment (LCA) analysis of photocatalytic concrete plays a crucial role in ensuring the environmental sustainability of building materials. It is capable of comprehensively assessing the energy consumption and carbon footprint of materials throughout their entire life cycle, as well as identifying key environmental hotspots, such as greenhouse gas emissions, water resource consumption and pollutant emissions. Through LCA, one can clearly understand the environmental performance of building materials during their preparation and the environmental impact of recycling building materials, which is conducive to promoting the development of more eco-friendly alternatives. Additionally, the results of LCA are beneficial in driving the green transformation of the construction industry, encouraging the adoption of cleaner and more efficient production methods.

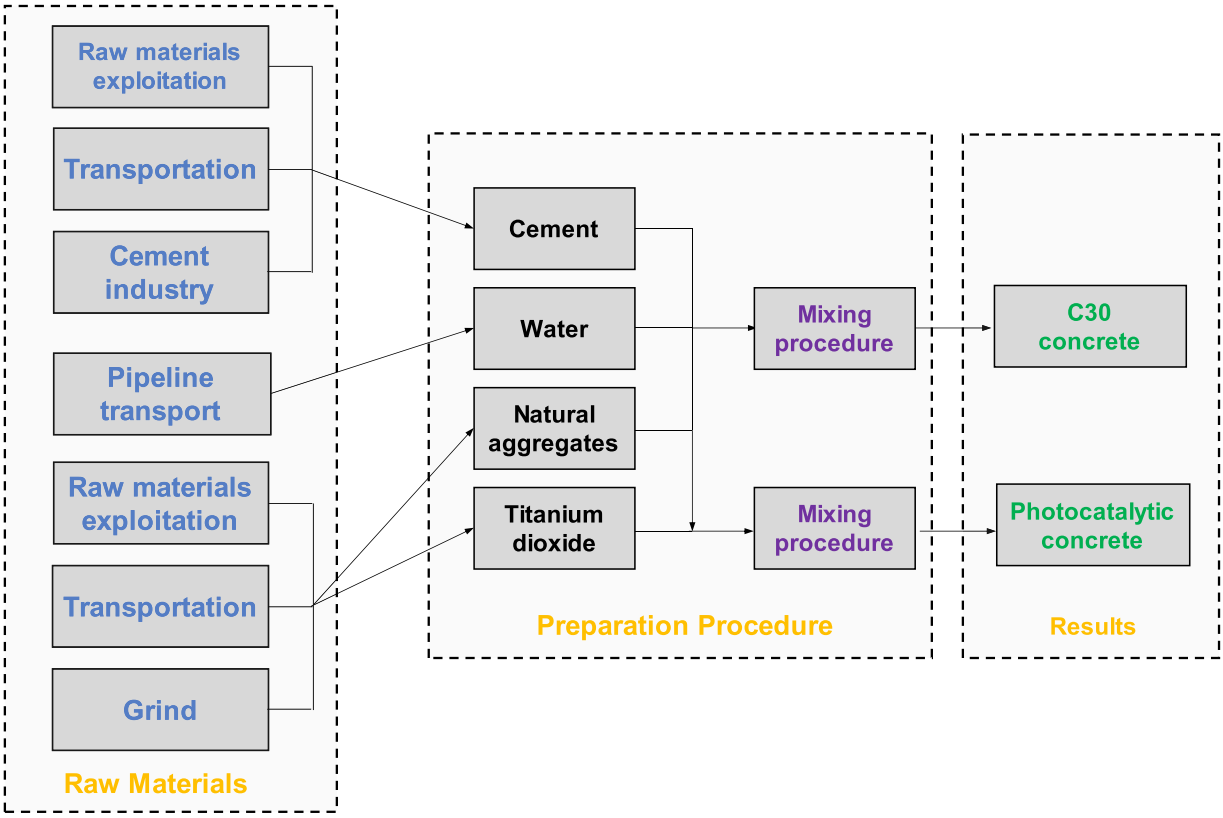


Fig. 15. Photocatalytic concrete as well as system boundaries for C30 concrete [136–138].

Table 2
LCI data for each stage of the life cycle.

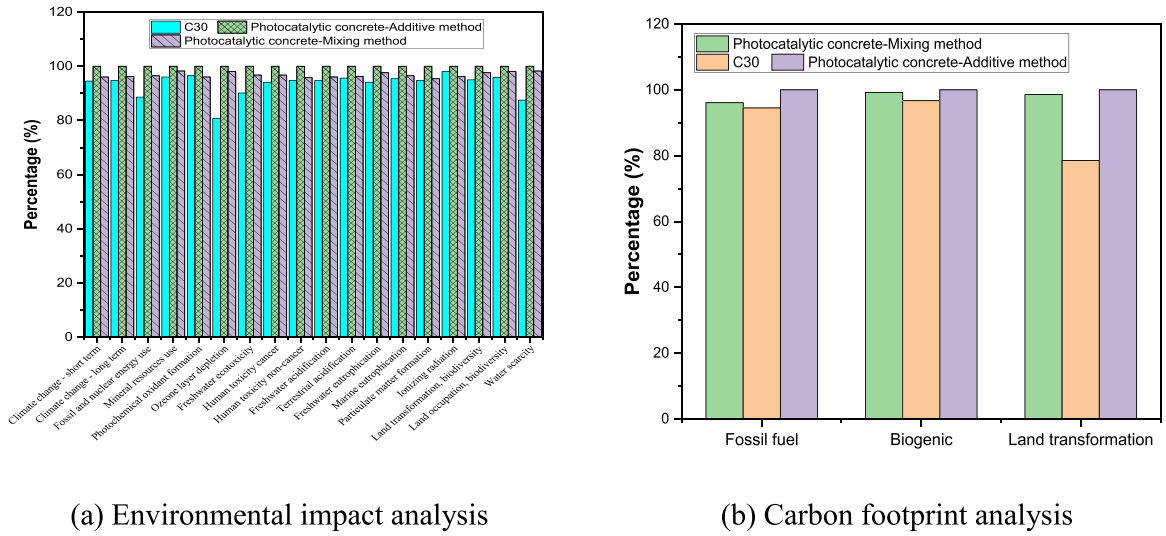
Production of cement (kg)		Production of natural aggregate (kg)	Production of concrete (1 m^3)	Production of water (1 m^3)	Production of TiO_2 (kg)	Transport (km)
Energy (MJ)						
Coal	3.370140	—	—	—	—	—
Natural gas	0.083178	—	—	—	8.9	—
Diesel	0.024369	0.014780	—	—	9.3	1.540900
Electricity	0.507672	—	20.06894	0.48 (kWh m^3)	1.83 (kWh m^3)	—
Emissions to air (g)						
CO	4.203224	0.003475	0.722680	—	159	0.318850
NO_x	2.279068	0.015579	13.22440	—	1.44	0.984380
SO_x	3.646948	0.005447	98.75360	—	1.28	0.430940
CH_4	1.002748	0.001296	0.433290	—	—	0.123860
CO_2	861.2028	1.377926	5698.210	—	923	110.7700
N_2O	0.000756	0.000055	0.029100	—	—	0.002950

The system boundaries are mapped in Fig. 15, which is supported by previous studies [136–138]. These key stages cover the process from the processing and transport of raw materials to the production of concrete. The system boundaries cover not only the main steps of concrete production but the processing and transport of materials and the handling of artificial aggregates. Additionally, we have included the energy required for concrete mixing to ensure a comprehensive system boundary. This article aims to improve overall environmental efficiency and reduce the ecological footprint of the life cycle through in-depth analysis and optimization of these key stages. During our analyses, we carefully examined the inputs and outputs at every stage. Life Cycle Inventory (LCI) data was collected for the elements necessary to produce concrete, including TiO_2, aggregates, cement, concrete production, and transport [139,140]. Table 2 provides further details. To ensure data completeness, any gaps were filled using Ecoinvent v.3. To ensure comparability, the same transport distances were assumed when comparing the environmental impacts of different mixtures. This approach ensured the accuracy and reliability of our findings, providing a solid foundation for deeper insights into the environmental impact of concrete production.

We selected conventional C30 concrete as the reference case for this analysis to conduct a comparative LCA analysis with the current scenario, which involves using the photocatalytic concrete mix preparation method [141–143] (substitution of cement mass - mixing method) or (extra admixture - additive method) with the fitment examples presented in Table 3. The potential environmental impacts of the photocatalytic concrete preparation method were assessed through this comparison, providing important insights into sustainability and eco-efficiency. This comparative analysis aims to identify differences between two concrete preparation methods and suggest directions for improvement and optimization to promote sustainable and environmentally friendly concrete production. The analysis will provide valuable information for sustainability decisions related to concrete production.

Table 3
C30 concrete and photocatalytic concrete ratios.

Types of concrete	Water/cement ratio	Mix proportions (by weight of cement)				
		Cement	Coarse	Fine	Water	TiO_2
C30	0.65	1	3.5	3	0.65	
Photocatalytic concrete-Additive method (PCA)	0.65	1	3.5	3	0.65	0.05
Photocatalytic concrete-Mixing method (PCM)	0.65	0.95	3.5	3	0.65	0.05



(a) Environmental impact analysis (b) Carbon footprint analysis

Fig. 16. Examining the environmental impact and carbon footprint of photocatalytic concrete and C30 concrete [144,145] [146–150].

The three mixtures (C30, PCA and PCM) underwent analysis using Simapro 9.5, a software designed for LCA – a method used to evaluate the environmental impact of products, processes, or services. The research objective was defined through systematic modelling, which encompassed the raw material production, manufacturing, use and disposal phases. Data were collected for each life cycle phase, including information on raw material production, transport, manufacturing, use and disposal. The life cycle was divided into phases and processes, and interrelationships and interactions among them were identified. Finally, the environmental impact of each life cycle phase was assessed using selected LCA indicators to evaluate environmental impacts, including various types of pollution and resource consumption.

Fig. 16 (a) shows a comparison of the environmental impacts of the concrete mixtures prepared in three ways, i.e. climate impacts (long and short term), resource use and impacts on human health. The analysis shows that C30 plain concrete has the lowest environmental impact compared to all other categories. However, photocatalytic concrete, whether prepared by additional admixtures or by mixing, has greater negative environmental impacts than C30 concrete. This is particularly evident in terms of ozone depletion, climate change impacts and energy consumption.

Cement is a key material in the production of concrete, binding various particles into a strong body. However, the production of cement not only consumes a lot of natural resources, such as limestone and iron ore, but emits a lot of CO_2. In addition, the production of cement requires a large amount of energy, such as coal and natural gas, and may produce wastewater containing hazardous substances, leading to water pollution [151–156]. On the other hand, although photocatalytic materials such as TiO_2 can decompose pollutants under the action of sunlight and improve air quality, there are environmental risks associated with their production and use. The production process of these materials is energy and carbon intensive and can damage the environment. TiO_2 nanoparticles may pose environmental and human health risks [144,145]. Therefore, despite the air-cleansing properties of photocatalytic concrete, its overall environmental impact must be thoroughly assessed to ensure its sustainability.

The production and use of construction materials is a significant contributor to global greenhouse gas emissions. Statistics show that the construction sector is responsible for ~30% of global CO_2 emissions. To address this issue, the development of photocatalytic concrete has emerged as a potential solution to reduce the environmental impact of construction [146–150]. Photocatalytic concrete can effectively degrade certain air pollutants and improve urban air quality. However, it is important to note that its production process still has certain carbon emission issues, which require further optimization and improvement to mitigate its negative impact on global climate change.

We conducted a comprehensive analysis of the carbon footprint of concrete as shown in Fig. 16 (b). Upon comparison, the carbon impact of photocatalytic concrete, whether prepared by substituting cement with TiO_2 or by adding TiO_2 to the mixture, is higher than that of conventional C30 concrete, particularly regarding soil impact. This is primarily due to the significant energy and resource inputs required to produce photocatalytic materials. It shows that the carbon footprints of these methods differ significantly at the production stage. It is important to consider the environmental cost of the preparation process when pursuing environmental purification. Therefore, future research and applications should focus on finding more sustainable and environmentally friendly methods for preparing photocatalytic concrete to reduce the environmental impact of construction materials [157–160].

6. Conclusions

Photocatalytic concrete technologies are emerging and revolutionizing sustainable building practices. This review explored the multifaceted aspects of this innovative technology, highlighting its potential to improve urban air quality and enhance the environmental sustainability of building materials. Photocatalytic concrete, which

utilizes photocatalysts such as TiO_2, can catalyze the decomposition of hazardous air pollutants, thereby contributing to improved air quality and self-cleaning capabilities. The review delved into pollutant removal rates, influencing factors, preparation methods, mechanical properties, and Life cycle assessment, providing a comprehensive understanding of photocatalytic concrete. Based on the review, the following conclusions and comments can be drawn:

(1) Although cementitious materials are regarded as ideal supports for photocatalysts, the performance of various photocatalysts can vary significantly within these matrices. For example, the two morphologies of TiO_2, anatase and rutile, have a huge impact on performance, with the former being more active but less stable. Therefore, the choice of a suitable photocatalyst is critical to the environmental benefits and durability of concrete.
(2) When using photocatalytic concrete, it is extremely important to balance the advantages and disadvantages of each synthesis method. The mixing method maintains long-term activity but is costly, the spraying method is efficient but has poor durability, and the impregnation method attempts to balance cost and performance. The choice of a method needs to consider application requirements and resource conditions. Future research should aim to improve the efficiency of photocatalysis and its bonding to cement substrates to maximize its potential.
(3) Improving the efficiency of photocatalysts through surface modification and elemental doping is promising, with challenges including technical complexity, cost, and stability. Modified TiO_2 may be less active and may bind poorly to cement, and further research is needed to promote its use in cement.
(4) Photocatalytic concrete with the addition of recycled glass is superior to the use of sand for NO_x removal due to the translucency of the glass and the scattering and reflection of light by the color. Although studies have been carried out to investigate the factors affecting NO_x removal, such as initial NO_x concentration and relative humidity, a comprehensive study is needed to understand the detailed mechanisms under conditions of high NO_x concentration, varying humidity and gas flow rate.
(5) The incorporation of porous materials, such as zeolites or recycled aggregates, can improve the photocatalytic efficiency in cement for the removal of SO_2 and CO_2 through adsorption and increased nucleation sites. However, the mechanism and kinetics are not well understood and further research is needed to optimize the system.
(6) TiO_2 nanoparticles accelerate cement hydration to form dense coatings that act as nucleation sites rather than increasing cementation products. TiO_2 nanoparticles enhance the pore structure of cementitious materials; however, excessive concentrations may adversely affect their porosity. Future research should focus on various photocatalytic nanomaterials, such as fusion modification between SiO_2 and TiO_2, to improve efficiency and performance.
(7) Controlling the TiO_2 nanoparticle content below 3.0 wt% improves the compressive strength of concrete, while an excessive amount is detrimental. Different forms of TiO_2 have different effects on concrete properties. Modification of TiO_2 catalysts, such as the formation of Ti-O-Si bonds, can help to improve concrete strength.
(8) TiO_2 content has a significant effect on the flexural strength of concrete, with optimum levels in the 4–10 wt% range. Excessive amounts can affect performance and the content needs to be optimized to improve flexural strength.
(9) Although cement is an important binder, its production poses significant environmental challenges. On the other hand, although TiO_2 improves air quality, there are environmental risks associated with its production and disposal. Therefore, to promote the use of photocatalytic concrete, its environmental impact needs to be thoroughly assessed to ensure that it is truly environmentally friendly.
(10) The production of construction materials is a major source of greenhouse gas emissions. Photocatalytic concrete has the potential to improve urban air quality, but the high carbon emission production process needs to be improved and future research should develop more environmentally friendly production methods.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Declaration of Generative AI and AI-assisted technologies in the writing process

AI can be used in the writing process before submission, but only to improve the language and readability of the paper. Human oversight is needed. This requires appropriate disclosure at the bottom of the paper in a section before the references. Authors are responsible and accountable for the contents of work.

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