Self-Cleaning Concretes: An Overview

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Abstract

Many studies have been carried out on the problems of civil engineering with the change of human problems today and in the past. These studies contributed to the development of concrete technology. Concrete is an important building material consisting of mixing aggregate, cement and water with or without chemical and mineral additives since the first day of use. Concrete technology has made great progress and continues. With developing concrete technology, self-cleaning concretes have emerged. Many studies have been conducted on self-cleaning concretes by researchers. This article reviews the research published on self-cleaning concretes and presents its role in reducing environmental pollution and its place in future engineering studies. When we look at the studies on self-cleaning concretes that emerged as a result of the developments in concrete technology, it is seen that the developments have progressed considerably. Contemporary civil engineering has provided a highly effective solution for the solution of modern problems. Environmentally friendly building materials will fulfil their duty in reducing air pollution, one of the biggest problems of our time. Self-cleaning buildings and roads that reduce pollution may sound like futuristic ideas, but it is not far away to encounter these structures more widely in our country and our world.

1. Introduction

It is accepted by everyone that one of the most basic needs of humanity is shelter. Until people reached the technology of building housing for shelter, they met their needs in places ready for nature such as caves, rock shelters. As people continue their cultural evolution, biological, physical, chemical and technological developments have enabled humanity to reach a level to build housing. With the emergence of the need for binding materials in housing construction, no material improvement was observed in the binding materials until the 18th century, although materials such as pozzolanic ash were used by the Roman times, and possibly even by older civilizations. Although there are important works on cement and reinforced concrete in the 19th century, the widespread use of concrete reached the beginning of the 1900s[1].

Today, concrete is the most used building material. Many studies have been carried out in the field of concrete technology since the first use of concrete and concrete technology is still developing. Although concrete is mostly used as a carrier material by the civil engineers due to the sufficiency of compressive strength, it is also used for aesthetic purposes in the form it comes out of the mould. The first example of this is the 'Jahrhunderthalle' building, designed and built by M. Berg in Germany-Bresslau in 1916. In this building, the first example of the application called "Architectural Concrete, Bare Concrete or Exposed Concrete" is given by using the concrete surface as it comes out of the mould without being covered with any material. Today, there are thousands of examples of aesthetic exposed concrete application[2-4].

Today, one of the most important negative effects of urbanization is the increase in environmental pollution. With the increase of environmental pollution, the awareness of the people and society has been created to protect the environment. The public's awareness of the need to protect the environment has deepened and improved the work on new technologies used in civil engineering. Researchers have carried out studies to prevent the appearance of bare concrete on the concrete surface from being affected by environmental pollution, changing the colour of the concrete and getting dirty. As a result of the studies, it has been observed that by joining the photocatalytic

TiO₂ concrete mixture, the concrete does not lose its aesthetic properties, it is separated from the pollutants on its lifetime and remains in the same colour and texture. In this concrete technology, called self-cleaning concretes, photocatalytic reactions are caused by nitrogen oxide, etc. on the surface. It prevents permanent adhesion of contaminants containing to the surface and separates them [5].

2. Photocatalysis Process

Photocatalysis is a word derived by combining the word photo, which means light, and the catalyst words, which are defined as the substance that accelerates a chemical reaction [6]. Photocatalytic reaction is one of the Advanced Oxidation Technologies applied to clean water and air. TiO₂ is often used as the catalyst for this process (TiO₂ is activated by UV light of certain wavelengths) [7].

The self-cleaning principle was discovered in 1973 by Wilhelm Barthlott and his team at the University of Bonn. The products from the self-cleaning reaction are shown in Figure 1. These products are easily removed from the surface with the help of water thanks to the photocatalytic effect with the addition of TiO₂ (titanium dioxide) [8].

Figure 1. Photocatalytic Reactions on a Self-Cleaning Substance Surface [8].

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The photocatalytic effect found by accident in Japan offers us many application opportunities in different areas of our daily life. Plants get their energy from daylight. Plants produce oxygen by oxidation of water and reduction of CO_2 . In other words, photosynthesis takes place with energy from sunlight. Similar to the photosynthesis reaction, photoelectrolysis of water has been started to be investigated using solar energy. Due to their similarity with the photosynthesis reaction in plants, photocatalysis can also be called artificial photosynthesis [9]. The similarity of the photocatalysis reaction with photosynthesis is shown in Figure 2.

Figure 2. The relationship between photocatalysis and photosynthesis [10].

TiO_2 is a metal that exists in nature (Figure 3a). TiO_2 oxygen has three different molecular structures, rutile, anatase and brocite [12]. Rutile is known as a pigment in white paints and has low photocatalytic reactivity (Figure 3b). Anatase is preferred as a photocatalytic cell (Figure 3c).

Figure 3. a) TiO_2 b) Crystal structure of rutile [7]. c) Crystal structure of anatase [7]

To use anatase in heterogeneous photocatalysis, the UV beam with a wavelength of less than 387 nm must be present. Besides, the intensity of the light is important to optimize photocatalytic activity. Normal daylight can be used for photocatalytic reaction. Photocatalytic reaction applications are many materials such as water treatment, air conditioning (air cleaning), self-cleaning glass, ceramic tiles (self-cleaning, anti-bacterial, etc.) textile (anti-odor), mirrors (anti-condensation), tunnel lighting and used in the industry.

Figure 4. Conversion of nitrogen oxides (NO , NO_2) to raw material in photocatalytic oxidation [7]

The use of TiO_2 with cement and other building materials has a positive effect on the removal of nitrogen oxides (Figure 4) [12-14].

3. Photocatalytic Cement

Products containing photocatalytic cement reduce harmful organic and inorganic materials in the air and maintain the aesthetic quality of the products that have deteriorated over time. It is used in building materials such as photocatalytic cement, concrete mortars, paints, precast elements and interlocking stones. They are environmentally friendly [14]. Concretes produced with photocatalytic cement may show different properties depending on the formulation used.

Photocatalytic cement with self-cleaning properties - Concrete counteracts most of the organic and inorganic contaminants that accumulate on the surface and causes discoloration. Organic and inorganic polluting substances:

- Soot, grime and organic particulates
- Mould, mildew, fungus and their spores
- Algae, bacteria and allergens
- Tobacco smoke and stains [15]

Self-cleaning photocatalytic cement are cement used in high-level architectural structures where the quality of the construction material used and the final appearance are equally important [14].

Photocatalytic cement capable of self-cleaning and reducing environmental pollution - In addition to the self-cleaning effect, these photocatalytic cements remove a significant amount of environmental pollutants that are thought to be harmful to health caused by human activities (industry, transportation, residential and heating systems of other structures).

Environmental pollutants [11, 13]:

- Nitrogen Oxides (NO_x) – major component in the formation of acid rain, ground-level ozone (smog), certain toxic chemicals and water quality deterioration
- Sulfur Oxides (SO_x) – component to acid rain and the formation of many harmful sulfates and other products.
- Volatile Organic Compounds (VOC's) – such as benzene and toluene
- Ammonia – NH_3
- Carbon monoxide
- Organic chlorides, aldehydes, polycondensated aromatics, among others [15]

Photocatalytic cements: short history shows in Table 1[13].

4. Photocatalytic Concrete

In concretes prepared with TiO_2 admixture, the quality of the concrete should not be put at risk in the plastic state and hardened state. Collapse test, air content ratio, unit weight, setting time, workability of concrete, permeability, flexural strength and compressive strength should give similar results. Concretes containing TiO_2 must be prepared by current standards for ordinary Portland cement concrete and other accepted industry applications [16]. From this point of view, it is seen that concretes containing TiO_2 generally have the same physical and mechanical properties as conventionally prepared concrete. On top of that, it cleans itself and delivers exceptional shine so that its original beauty can be preserved for years to come. It can be used in horizontal and vertical structures as well as tunnels where air quality and safety conditions are improved. Photocatalytic concretes are one of the most important factors in the fight against dirty air accumulation [15]. Areas where photocatalytic concrete can be applied in horizontal construction; concrete pavements, interlocking paving stones, paving and road coverings, cement-based tiles and concrete roof tiles. The areas where it can be applied in vertical construction; architectural precast facade panels, plaster, exterior plaster, cladding elements, noise barriers and concrete refuges for roads and highways [14].

Table 1. Photocatalytic cement: short history [13]

1995-1998	R&D program on photocatalysis in cement-based systems
	The first patent application (1996, Italy)
	First publications First concrete trials
1999-2001	First project: 'Dives in Misericordia' Church, Rome
	'Cité de la Musique et des Beaux-Arts', Chambéry Rhodamine B (discoloration) test, (CSTB, France)
2002-2005	Start of PICADA Project (2002–2005)
	Street canyon experience
	Pilot depollution tests in Segrate, Calusco, Bergamo
	Launch of the first version of photocatalytic cement (TX Millenium®) Development of laboratory test methods
2006-2017	Launch of TX Active® Cements (Italy, France, Spain, USA).
	Publication of first UNI standards (Italy)
	'Umberto I' Tunnel (Rome, 2007)
	The licence agreement with Heidelberg Cement Group (2008)
	Projects: i.lab, Bergamo Vodafone Village, Milano

In the experiments, it was found that the nitrogen oxide density measured in the area occupied by the photocatalytic concrete blocks was lower than the other areas compared. An average reduction of 45% was recorded. In the case of sunlight or artificial light, building materials containing TiO_2 can also be used indoors [15].

According to the test results of Italcementi, which produces building materials containing TiO_2 in Italy, it has been observed that a road paved with photocatalytic cement can reduce NO_2 levels by 20 to 80% depending on the atmospheric conditions. A building built with photocatalytic precast siding can do the same [16]. In 2002, a photocatalytic mortar was tested for the first time to cover the asphalt surface of a road in which 1000 vehicles per hour, 230 m long, 10 m wide, on Via Morandi in Segrate (Milano). In this way, it has been observed that there is a 60% reduction in nitrogen oxides. In 2003, interlocking paving stones containing over 8,000 m² of titanium dioxide were applied to an industrial area in Bergamo, Italy. Compared to another area with the same properties as nitrogen oxides in this area, it has been observed that there is much lower density nitrogen oxide. An average reduction of 45% was measured [17].

5. Literature Review

It was described by Frank and Brad in 1977 as the decomposition of cyanide in a liquid titanium dioxide (TiO_2) solution; this is one of the first reports to reveal the usability of semiconductor powders in photocatalytic production [18]. The first examples used in photocatalysis building materials are cement mortars, paving stones, tile materials, glasses and PVC fabrics. In 1997, concrete paving stone production containing TiO_2 was started for the first time in Japan. It was used for the first time in 1998 in a self-cleaning concrete structure. The Jubilee Church (officially known as the Misericordia Church) in Rome is the first building containing self-cleaning concrete [19,20]. Folli et al. They investigated how TiO_2 dimensions used as photocatalysts of self-cleaning concretes affected self-cleaning performance in 2009. They reported that samples containing micro sizes of TiO_2 showed better photocatalytic performance than samples containing nano sizes of TiO_2 [21]. Rout et al. In 2009 he compared the self-cleaning performance of cement paste and mortars. They produced samples containing 0%, 1%, 3% and 5% TiO_2 for both groups. In both groups, they performed better than the mortars in the samples containing 5% TiO_2 and cement paste [22].

Hüsken et al. In 2009, they produced photocatalytic concrete with TiO_2 additives. As a result of the experiments, they reported that they separated NO and NO_2 compounds under UV light [23].

Chen et al. In 2011, he investigated the self-cleaning performance of cement mortars. Rhodamine-B was chosen as the pollutant. The color change was observed in products containing TiO_2 . No colour change was observed in non- TiO_2 products [24].

In a thesis study on self-cleaning concrete in 2010, four different types of concrete were produced: white concrete, white concrete with TiO_2 additive, self-setting concrete and self-settling concrete with TiO_2 additive. To examine the effect of photocatalytic properties, physical properties and durability properties on surface properties; To examine the effect of photocatalytic properties, physical properties and durability properties on surface properties; Three different formwork types have been produced for each type of concrete, metal, wood and rubber. To investigate photocatalytic properties, Rhodamine-B experiment and experiments for Phenantroquinone cleaning properties were investigated. The results obtained in the research are presented comprehensively. At the end of 28 days, the series that remained under UV in Rhodamine-B experiment was found to be in white concrete samples containing TiO_2 additives with the highest colour change 43.5% colour change was observed in self-compacting concrete. While natural concrete and self-settling concrete with TiO_2 additive and without TiO_2 additive are left to natural environmental conditions, the most colour change was observed in the samples left open to the rain. In the samples left horizontally and vertically closed and exposed to sunlight, colour change was observed at the end of 28 days. Here, it has been observed that substances that break down by photocatalytic reaction and cause contamination with TiO_2 must be removed from the concrete surface by rainwater [2,4].

Khatee et al. In 2013, the self-cleaning properties and mechanical properties of white cement were investigated with TiO_2 . X-ray diffraction, electron microscopy transmission, and BET were used to characterize TiO_2 nanoparticles. Mechanical properties such as hydraulic cement socket time, compressive strength and bending strength of cement samples with TiO_2 additive and non- TiO_2 were investigated. The increase in the amount of TiO_2 nanoparticles in the mixed cement increased the self-cleaning properties of the samples. It has been reported that flexural strength increases and hardening times decrease [25].

Biolzi et al. In 2013, they investigated the photocatalytic properties of self-cleaning concretes under high temperatures. While maintaining the self-cleaning feature at 500 °C, it has been reported to rise to 750 °C while a decrease in this feature has been reported [26].

Mendoza et al. In 2015, they investigated the photocatalytic performance of mortars containing silicon TiO_2 . They reported that the discoloration reached 90% at the same time, the samples were reported to have good early performance [27].

Shen et al. in 2015, they produced an ultra-smooth surface photocatalytic concrete using cement calcium silicate hydrates (C-S-H) main hydration product, nano granular nature and photocatalytic properties of TiO_2 nanoparticles. As a result, they found that the surface of the produced ultra-smooth surface concrete was coated with nanoparticles, C-S-H and TiO_2 . They reported that the concrete surface containing TiO_2 had an obvious effect on the degradation of MB blue. They observed that due to its ultra-smooth surface and photocatalytic properties, dirt on the surface can be washed with rain [28].

Azevedo et al. in 2015, they researched the cleaning ability of photocatalytic mortars. In this study, they investigated the mechanical properties and self-cleaning properties of photocatalytic mortars in case of various factors. Four different components were produced for each sample, each with a different TiO_2 content. TiO_2 content is 0%, 2%, 4% and 6%. As a result, they reported that as TiO_2 increased, they reached lower strength. The highest self-cleaning properties and highest strength were obtained in samples containing 2% TiO_2 [29].

Andalora et al. In 2016, they researched on self-cleaning concretes with photocatalytic properties for the maintenance of the facades of buildings. In their work, they investigated the self-cleaning performance of titanium dioxide-based coatings applied to different facade materials. Before outdoor testing, they conducted preliminary laboratory tests with water contact angle measurements to verify hydrophobic and hydrophilic behaviour. They then observed for 36 months to test their stamina. In line with the results, they reported that applying the nanotechnological coating to a facade can significantly facilitate cleaning operations. They came to the idea that there was no need for extra cleaning on the facades where the nanotechnological coating was used [30].

Duan et al. The effects of the addition of TiO_2 particles on fly ash-based geopolymer properties were investigated. They conducted studies on compressive strength, drying shrinkage, carbonation and microstructure of fly ash-based geopolymer containing TiO_2 particles. Geopolymer samples were prepared by alkaline activation of fly ash in sodium silicate and sodium hydroxide solutions and hardened in periods of radiation with additional heat. The properties of samples with and without TiO_2 particles were evaluated and compared. Experimental results have shown that the addition of TiO_2 particles increases the early and improved wet compressive strength of samples with increased TiO_2 particles. They found that TiO_2 increased the strength in the samples more clearly by adding 5% TiO_2 in 28 days [31].

Liang et al. In 2019, they developed some experiments (zebrafish water toxicity, alkaline dissolution and photocatalytic oxidation reaction, etc.) to measure the processing methods of porous concrete treated with TiO_2 material for the purification of running water pollution. They observed that TiO_2 's surface treatment method had a minor effect on the physical properties and pH of porous concrete. The results of the zebrafish water toxicity experiment found that porous concrete's survival rate of zebrafish embryos reached 91.7%, and as a result of this technology, the use of TiO_2 in porous concrete is not an environmentally friendly material and has no significant acute biological toxicity. As a result of other experiments, methylene blue (organic pollutant) achieved a good purification effect on total phosphorus and ammonia nitrogen, and its efficiency reached 60-90%. The durability test results showed that the samples had a reduction in the water treatment effect after decomposition. Finally, porous cement concrete has been reported to have a promising purification function in flowing water pollution [32].

Feng et al. In 2019, he made studies to measure the photocatalytic performances and durability of cement composites containing TiO_2 prepared for the separation of organic wastewater. The composition, microstructure, hydrophobicity, photocatalytic properties and durability of the cement composites containing TiO_2 were investigated gradually. The TiO_2 particles are evenly distributed over the surfaces of the cement mixtures. They observed better performance than soaking in samples immersed in acid or alkaline solutions. Good processing performance was achieved for photocatalytic cement mortars and a decomposition efficiency of more than 93% was achieved for 20 processes. For durability, photocatalytic cement mortars have been found to have good water permeability, chloride penetration resistance and carbonation resistance. It has been found that carbonation of cement mortars containing water absorption, chloride electric flux, chloride penetration and photocatalytic TiO_2 is better than pure cement mortars. Cement-based composites provided a new direction for photocatalytic application in water pollutant treatment [33].

Koli et al. in 2019, they prepared a nitrogen-added TiO_2 - SiO_2 photocatalysts with a sol-gel method and coated it on hardened concrete for a photocatalytic concrete application. The effect of heat treatment has been investigated by low and high-temperature annealing. Textural properties of coated and uncoated samples were examined. The sample calcined at a low temperature of 180 °C was confirmed to have more functional parts than a high temperature of 450 °C. In the morphological study, it was shown that the high-temperature calcined sample contains more cracks than one at the low temperature. Photodegradation of toluene gas and methylene blue (MB) dye under UV light confirmed that the sample temperature processed at 450 °C showed high photocatalytic activity. The recyclability experiment proved that the coated samples had very good stability [34].

Yang et al. In 2019, they compared the photocatalytic efficiencies of TiO_2 supported on mortar surfaces with the more traditional TiO_2 dispersed in the mortar. In this study, the effects of environmental conditions such as NO concentration and flow rate, UV light intensity and relative humidity on photocatalytic performance were also investigated by using photonic efficiency as an indicator. As a result of the researches, a significantly higher usage efficiency (about 150 times higher) than TiO_2 was obtained in photocatalytic mortars. Also, its efficiency and low cost further confirmed TiO_2 's advantages in photocatalytic concrete technology [35].

Xu et al. in 2020, they did some experiments on porous concrete containing TiO_2 and recycled aggregate. First, the optimum mixture

design of permeable concrete containing TiO_2 recycled coarse aggregates was investigated to obtain higher compressive strength. Then, the photocatalytic performance of permeable concrete coated with TiO_2 photocatalysts and containing recycled coarse aggregate was investigated. The photocatalytic performance of the permeable concrete was tested with heavy rainwater for 10 minutes. Experimental test results have shown that the internal cavities of the adhering mortar absorb more TiO_2 particles with recycled coarse aggregates. As a result of the experiments, they reported that the permeable concrete using the recycled aggregates coated with TiO_2 can greatly preserve their photocatalytic capacities. This study addresses two main social and environmental issues in developing countries. These are large amounts of construction, demolition waste and air pollution. Cleaner production will be achieved in concrete coating construction by optimizing the reuse of wastes, air cleaning and engineering properties of porous concrete. It also demonstrates that the application of recycled aggregates has many potential advantages, including reuse of wastes and improved photocatalytic performance with improved durability by addressing the durability of photocatalysts concrete from a rain wash. They reported that in the future, self-cleaning concrete can be removed from the air using recycled aggregates with a photocatalysts [36].

In 2020, Chen et al. They prepared composite photocatalysts by loading nano- TiO_2 on recycled clay, brick sands and recycled glass. And they did a study to observe the rheological behaviour, mechanical performance and NO_x removal of the photocatalytic mortar prepared with these photocatalysts. The combined use of Nano- TiO_2 with recycled clay, brick sands and recycled glass is beneficial for rheological behaviour. The inclusion of composite photocatalysts has been found to support photocatalysis. Besides, NO_x removal was reported to be increased by 18.8% while cost was reduced by 80% due to mixing [37].

6. Advantages of Self-Cleaning Concretes

As the temperature increases in areas where urbanization is intense, air pollution and chemical reactions that produce pollution also increase. The purpose of using self-cleaning concretes is to use materials that stay cool in sunlight and have at least 29 solar reflective indexes (SRI). SRI value of ordinary Portland cement is about 35, and the SRI value of new concrete produced with white cement is 86. It is seen that the concrete produced with TiO_2 added cement will maintain a higher SRI value for a much longer time [38].

All building energy simulations prepared by computer modelling will show us the benefits of concrete prepared with TiO_2 cement by solving the complex problems between the sunlight reflection rate and emission rate between concrete prepared with TiO_2 added to cement and concrete prepared with traditional methods [38].

Exceptional performance in reducing air pollution - The use of concrete prepared with cement containing TiO_2 will reduce organic and inorganic substances causing air pollution [38]. Durability - Concrete prepared with cement containing TiO_2 preserves its durability for a longer period without using protective coating materials [38].

7. Disadvantages of Self-Cleaning Concretes

White spots are formed on the surface of self-cleaning concrete due to titanium dioxide. Light is needed to react chemically and is therefore not suitable for indoor applications. The cost of self-cleaning concrete is much higher than conventional concrete and no standard has been published in our country [38].

8. Examples of Photocatalytic Concrete Applications in the World

- Self-cleaning locked cobblestone pavement on 'Borgo Palazzo' Street (Bergamo, Italy). In the measurements made on the street, a reduction in air pollution between 30% and 40% was observed.
- Painting of the tunnel of 'Umberto I' (Rome, Italy) with photocatalytic material
- 'Dives in Misericordia' Church (Rome, Italy) is one of the architectural works that preserves its beauty with its self-cleaning feature (Figure 5).
- 'Cit de la Musique et des Beaux-Arts' (Chambery, France) (Figure 6)

- 'The Hôtel de Police' (Bordeaux, France) (Figure 7)
- MSV Arena Football Stadium (Germany) (Figure 8)
- 'Bienvenue a Ciments du Maroc'(Morocco)(Figure 9)
- 'Charles de Gaulle' Airport (Paris, France)
- Saint John's Court Montecarlo Bay residence (Monaco)
- Manuel de Gonzalez Hospital, Mexico (Figure 10)
- Tüpras Refinery, Kocaeli - Turkey (Figure 11)

Figure 5. "Dives in Misericordia" Church-Rome, 1996[39]

Figure 6. 'Cité de la Musique et des Beaux-Arts'-France, 2001[40]

Figure 7.'Hotel de Police'-France, 2003[41]

Figure 8.MSV Arena Football Stadium-Germany, 2004 [42]

Figure 9.'Bienvenue a Ciments du Maroc'-Morocco, 2005[43]

Figure 10. Manuel de Gonzalez Hospital-Mexico, 2013[44]

Figure 11. Tüpras Refinery -Kocaeli-Turkey, 2014[45]

9. Conclusions

When the literature studies are reviewed on self-cleaning concretes that emerged as a result of the developments in concrete technology, it is seen that the developments have progressed considerably. The current knowledge shows that the influence of adding of photocatalysts (as nano-TiO₂) in the cementitious material to be as filler or replacing part of cement improve the performances of self-cleaning behavior. Photocatalysts will also remove odours and indoor air quality, so that, the city will become clean and beautiful. Nowadays, atmospheric pollution also causes the external decay of buildings due to the deposition of organic matter and contaminants. Environmentally friendly building materials will fulfil their duty in reducing air pollution, one of the biggest problems of our time. Self-cleaning concrete has a potential to keep the city clean by reducing the air pollutants. It is obvious that self-cleaning concrete will reduce air pollution by 30-40%.

Declaration of Conflict of Interests

The authors declare that there is no conflict of interest.

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