



Self Cleaning and Reduction of Air Nitrogen Oxide by Cement Composites Facade Containing TiO_2 and Nano-Silica

**Foroozan Mostofi¹⁾, Fatemeh Nasehi²⁾, Babak Pordel Maragheh^{*3)},
Ebrahim Fataei⁴⁾ and Mehdi Nezhadnaderi⁴⁾**

¹⁾ Department of Environmental Science and Engineering, Ardabil Branch, Islamic Azad University, Ardabil, Iran.

^{*2)} Department of Environmental Science and Engineering, Ardabil Branch, Islamic Azad University, Ardabil, Iran.

³⁾ Department of Civil Engineering, Ardabil Branch, Islamic Azad University, Ardabil, Iran.

⁴⁾ Department of Environmental Science and Engineering, Ardabil Branch, Islamic Azad University, Ardabil, Iran.

⁵⁾ Department of Civil Engineering, Tonekabon Branch, Islamic Azad University, Tonekabon, Iran.

³⁾ civil_babak2005@yahoo.com

ABSTRACT

At present, self-cleaning and anti-pollution concrete products are produced by various companies for use in the facades of buildings and road floors, and have been widely used in Europe and Japan. The facade of the building, as one of the most important items of the building, is always exposed to sunlight and harsh weather conditions, and its repair and repair will not only have high costs for the owners. Production of catalytic cement and new concrete compounds with titanium dioxide coating was designed and implemented for roofing and facades of buildings. In this paper, the effect of using nanosilica and titanium dioxide on the self-cleaning properties of concrete is investigated. Four designs were tested with zero, 2.5, 3.5 and 5% replacement of cement with TiO_2 , each with 2% nanosilica percent adsorption percentage. Reducing the color change of soot between the designs of the studied mixtures shows the positive effect of TiO_2 in reducing the color of soot pollution on the concrete surface. An increase of titanium dioxide up to 5% leads to a decrease in soot color in the samples, and this is due to the photocatalytic properties of titanium dioxide for higher percentages. The intense chemical activity of titanium dioxide in the presence of ultraviolet rays can prevent bacteria and dirt from sticking to the facade of concrete walls and buildings.

6 samples each with 2 water absorption percentage tests and 3 nitrogen oxide absorption tests make the number of the statistical population to 30. To perform these tests, UV tests were taken from the samples. This test will be done according to ASTM C642 method. The addition of the highest percentage of titanium dioxide compared to other percentages (5%) and 2% nanosilica causes greater density and the highest percentage of nitrogen oxide absorption up to 13.2% of

¹⁾ Ph.D. Student

^{2,3 and 5)} Assistant Professor

⁴⁾ Associate professor



the original sample. As a result, adding a higher percentage of titanium dioxide up to 5% and nanosilica up to 2% will be effective in reducing and absorbing about 30% of air nitrogen dioxide, which can be used in environmental concrete for sustainable development.

Keywords: Titanium dioxide, Prefabricated facade, Nanotechnology, Self-cleaning property, Nanosilica, Reduction of nitrogen oxide in air.

INTRODUCTION

Photocatalytic self-cleaning is one of the most important applications of nanotechnology in the construction industry. Natural and industrial pollutants such as NO_x, carbon monoxide, VOCs, chlorophenols and aldehydes from automobiles and industrial effluents are degraded by photocatalysts with the help of a highly active catalyst of titanium dioxide nanoparticles [1]. To activate the self-cleaning effect of titanium oxide, natural daylight, air humidity and oxygen are required. The activity of titanium dioxide-based photocatalytics can be comprehensively studied in the article [2]. For example, we can name the Jubilee Church in Rome, Italy [3] as shown in Figure (1).



Figure 1. Jubilee Church, Rome, Italy. In the facade of this structure, concrete with a combination of titanium dioxide nanoparticles has been used [3].

Nanosilica can reduce cement consumption, improve concrete quality and increase its efficiency [4, 5].

The incorporation of TiO₂ into cementitious composites has been investigated by some researchers. Sanaf et al. (2012) added fractions of different volumes of Nanosilica and Nano



TiO₂ to the mortar mixture and evaluated the hardened properties. It has been reported that when the content of nanosilica and TiO₂ is low, mechanical strength is not significantly affected, but with increasing weight percentage of nanosilica and TiO₂, mechanical strength increases by 3% and 12%, respectively [6].

In a study, the effect of using nanosilica (NS) and microsilica mixtures on the mechanical properties of ultra-high performance cementitious composites (UHPCs) is presented.

To do this, two concrete groups with and without silica fume with a ratio of water to fixed cement and fixed cement were designed. Commercial NS used in partial replacement of cement with weight of zero percent, 0.5 percent, one percent, two percent and 3 percent. The results show that among the different contents of NS, UHPC containing 2% NS showed the best results in compressive strength, tensile strength, modulus of elasticity, flexural strength, load handling behavior and rupture energy in 90 days. UHPC samples containing cementitious materials (NS and SF) gave better results than concretes containing only NS. In addition, the effect of 1% NS is approximately equal to 10% SF [7].

In one study, the photocatalytic activity of TiO₂-coated self-compacting glass-containing mortars (SCGMs) was investigated in terms of removing air pollutants and inactivating bacteria. TiO₂-impregnated glasses were used as controls to compare performance [8].

Nitric oxide (NO) and Escherichia coli K12 were used as air pollutants and bacterial pollutants, respectively. In addition, the weathering resistance of TiO₂ coated samples was investigated. Regarding NO removal, it is clear that no significant difference was observed between TiO₂ and SCGM impregnated glass, and both showed high NO removal efficiency when using conditions 1 (C1, without weathering) (for EtOH mortar, Up to 14.33 mg per square meter H1). However, after a period of abrasive weathering, condition 3 (C3, abrasive process), the ability to remove NO from TiO₂-impregnated glass samples almost disappeared. In contrast, the ability to remove NO from SCOM with TiO₂ coating remains high performance (for EtOH mortar, 8.75 mg / m² H1). The porosity of the SCGM surface with the coating appears to help maintain the desired TiO₂ particles after abrasion. For antibacterial activity, a general inactive action of E. coli removal was observed in TiO₂-impregnated glass and SCGM samples within 60 min of UV irradiation.

After the wear process (C3), the inactivation of E. coli in TiO₂-impregnated glass is almost negligible, whereas, the concentration of E. coli remaining on the surface of TiO₂-coated SCGM is only about 10⁵ to 10³ CFU / ml. Dropped. The results indicate that TiO₂ is retained in the porosity of the coated SCGM and can still play a role in inactivating E. coli. Considering all the results, it can be concluded that inactivation of photocatalytic bacteria is a more complex process and the results of photocatalytic activity of NO removal can not always be transferred to photocatalytic antibacterial activity. The results of this research are presented in Figure (2) [8].

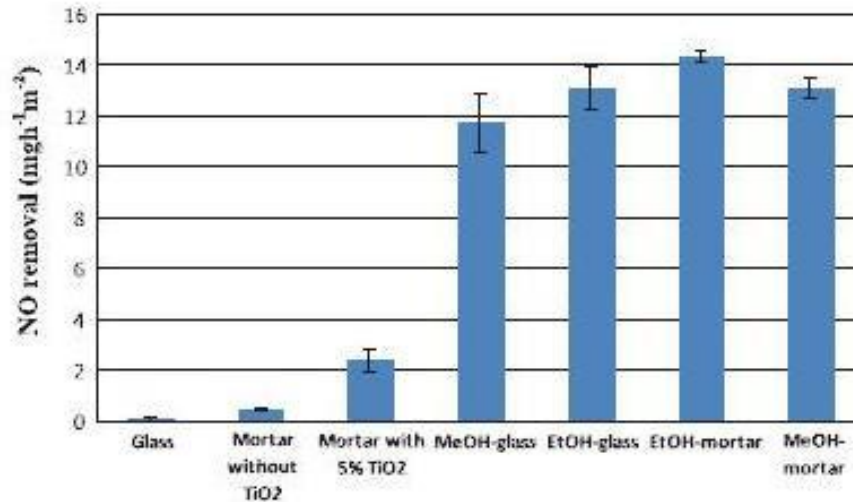


Figure (2) Comparison of NO removal by different samples subjected to UV testing in 60 minutes [8].

In all experiments, a commercial nano TiO₂ powder (P25, Degussa) was available and used as a photocatalyst. The size of TiO₂ was 20-50 nm, BET with a specific surface area of 15 50 50 m² / g and ordinary white Portland cement (WC, TAIHEIYO) product of Japanese cement and metakaolin (MK) were used as samples. Recycled glass (RG) from post-beverage glasses was used as a fine material in SCGM. The light green glass bottles are washed and then crushed by a mechanical crusher, followed by a sieve with a particle size of less than 5 mm. Self-compacting glass mortar (SCGM) was prepared by compression of 50 MPa resistance (28 days) using a mixture ratio of 0.8: 0.2: 0.2: 0.4 (WC, MK, RG, water). In order to evaluate the effectiveness of TiO₂ on the photocatalytic activity of mortar, a amount of 5% (cement mass) of nano TiO₂ in the complete mixture was used. The mixing schemes used are given in Table (1) [8].

Table (1) Mixing schemes used to test water absorption percentage [8].

Sampels	Status of projects
Mortar without TiO ₂	Self-compacting glass mortar (SCGM) without TiO ₂
Mortar with 5% TiO ₂	Self-compacting glass mortar (SCGM) with 5% TiO ₂
Glass	Reference glass without TiO ₂ coating
EtOH-glass	TiO ₂ -impregnated glass suspended in ethanol and glycerol solution at 450 ° C for 120 min
MeOH-glass	TiO ₂ impregnated glass suspended in methanol solution at 60 ° C for 120 min
EtOH-mortar	Mortar impregnated with TiO ₂ suspended in ethanol and glycerol solution at 120 ° C for 120 min
MeOH-mortar	Mortar impregnated with TiO ₂ suspended in methanol solution at 60 ° C for 120 minutes



In an article to determine the effect of cement containing titanium dioxide, nanosilica and the presence of waste glass in cement mortar for its potential application in self-cleaning facades. Studies have shown that waste glass can be a successful alternative to sand, especially when part of it is mixed with sand. In addition, a positive effect of nanomaterials was observed due to self-cleaning and mechanical properties. Visual observation of rhodamine B discoloration on cement mortar surfaces showed that the presence of waste glass did not affect the cleaning properties of titanium-containing commercial cement. The use of waste glass in general can neutralize the negative effect of high nanosilica water demand. Hence, nanosilica can be successfully incorporated and embedded in cementitious composites without the aid of any additional material. The use of nanosilica improves the cohesion between the glass aggregates and the cement due to its paste property and consequent compaction of the cement mortar structure. The use of 3% by weight of nanosilica significantly increases the flexural and compressive strength of specimens containing waste glass materials [9].

The importance of corrosion protection of reinforced concrete in aggressive and corrosive environmental conditions and protection against water absorption has led to the allocation of 150 publications of the Program and Budget Organization entitled Environmental Concrete Structures (translated by ACI 350-89) [10].

1. Experimental Study

In this research, using nanosilica replacement percentages and different percentages of using 0, 2.5, 3.5 and 5 percent titanium dioxide. The water absorption percentage of the samples has been investigated. The reason for using these two materials together to compensate for the increase in the percentage of water absorption by titanium dioxide in concrete using nanosilica, which has a role of reducing the percentage of water absorption and are used in precast concrete elements.

2- Experimental section or laboratory activities

In this paper, with the help of laboratory methods, the effect of using titanium dioxide and nanosilica on reducing the contamination of prefabricated concrete parts in the architectural facade of concrete structures has been investigated.

Type I-42.5 cement was used in accordance with the American Association Standard for Testing and Materials (ASTM) -C150 [11]. The chemical composition of cement is presented in Table 2.

Table 2: Chemical composition of cement

Compound	Cement(%)
CaO	64.38
SiO ₂	21.08
Al ₂ O ₃	5.36
Fe ₂ O ₃	3.64
MgO	2
K ₂ O	0.82



Na ₂ O	0.5
L.O.I(Loss On Ignition)	0.9

TiO₂ with a diameter of about 800 nm and a specific gravity of 3.91 g / cm³ was used in the mixture ratio.

Mixing was performed by the method performed in ASTM C109 [12]. The dry materials were first mixed gently to obtain a homogeneous dry mixture. A mixture of water and superplasticizer was added slowly, and mixing was continued at high speed for 5 minutes to obtain a homogeneous mortar. In all mixed designs, water was kept constant relative to the cementitious material to prevent loss of mechanical strength. But due to the use of TiO₂ instead of cement in the ratio mixture, to obtain the mixture with optimal performance, a superplasticizer was added in different volume percentages. Samples were emptied after 24 hours and immersed until test. Table 3 shows the mixed designs of this study.

Tab. 3: Mix designs

Mix designation	Cement content(kg/m ³)	Replacement percentage		Water/cement
		UFTiO ₂	NS	
N2T0	800	0	2	0.38
N2T2.5	800	2.5	2	0.38
N2T3.5	800	3.5	2	0.38
N2T5	800	5	2	0.38

Figure 3 - Comparison of the use of 5% titanium dioxide with 2% nanosilica for self-cleaning properties in concrete before and after 24 hours of UV exposure and the figure on the right before the effects of UV and the left figure after 24 hours of UV exposure. Be.

According to Figure 3, after 24 hours of using UV radiation, it was found that the color of soot impregnated on the concrete surface decreases and this is due to the photocatalytic reaction of titanium dioxide exposed to ultraviolet radiation.

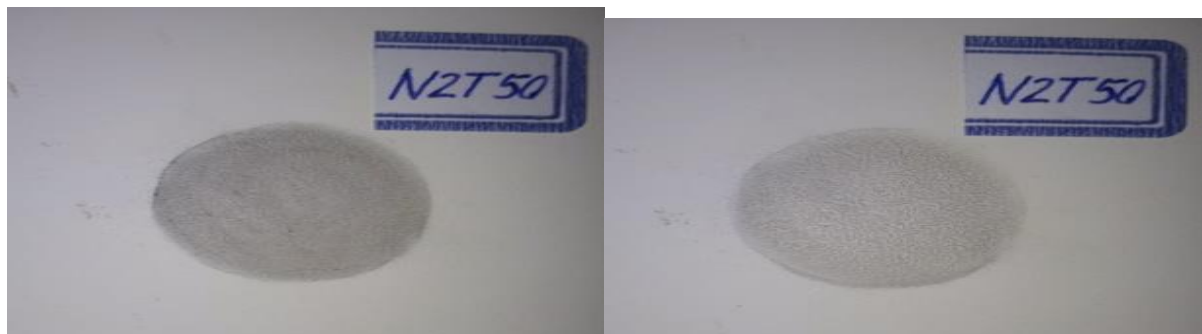


Figure 3 - Comparison of the use of 5% titanium dioxide with 2% nanosilica for self-cleaning properties in concrete before and after 24 hours of UV exposure and the figure on the right before the effects of UV and the left figure after 24 hours of UV exposure.



According to Figure 3, after 24 hours of using UV radiation, it was found that the color of soot impregnated on the concrete surface decreases and this is due to the photocatalytic reaction of titanium dioxide exposed to ultraviolet radiation.

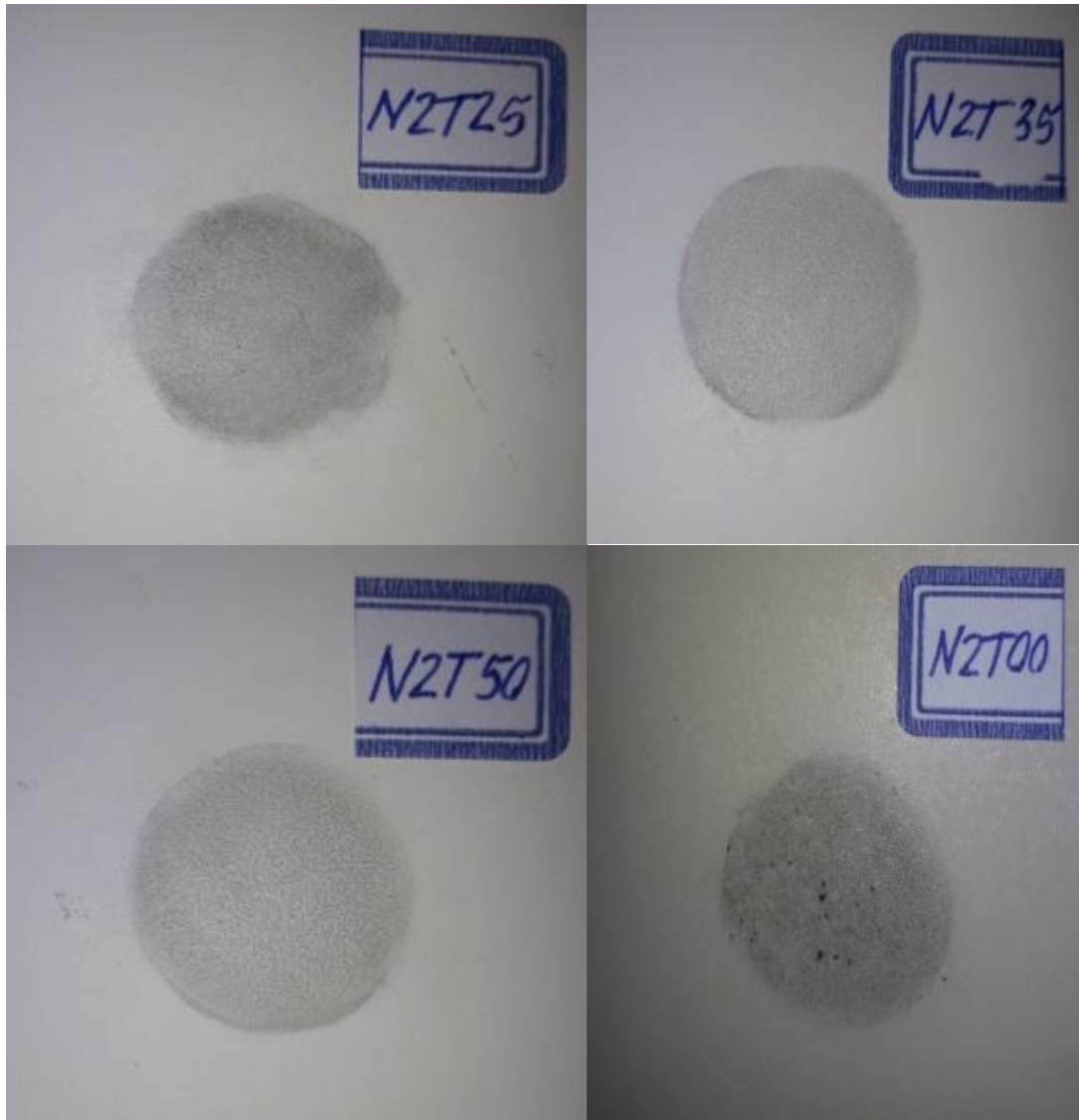


Figure 4 - Comparison of the use of zero, 2.5, 3.5 and 5% titanium dioxide with 2% nanosilica for self-cleaning properties in concrete after 24 hours of UV exposure.

According to Figure 4, increasing the percentage of titanium dioxide in samples containing 2% nanosilica reduced the color of soot impregnated on the concrete surface.



Figure 5. Device for determining the amount of reduction of nitrogen oxide adjacent to the samples in the tested designs



Figure 6. Device for determining the amount of reduction of nitrogen oxide adjacent to the samples in the tested designs



Figure 7. The input of nitrogen oxide gas from the tank to the surface of the sample in the test device to determine the reduction of nitrogen oxide adjacent to the samples in the tested designs.

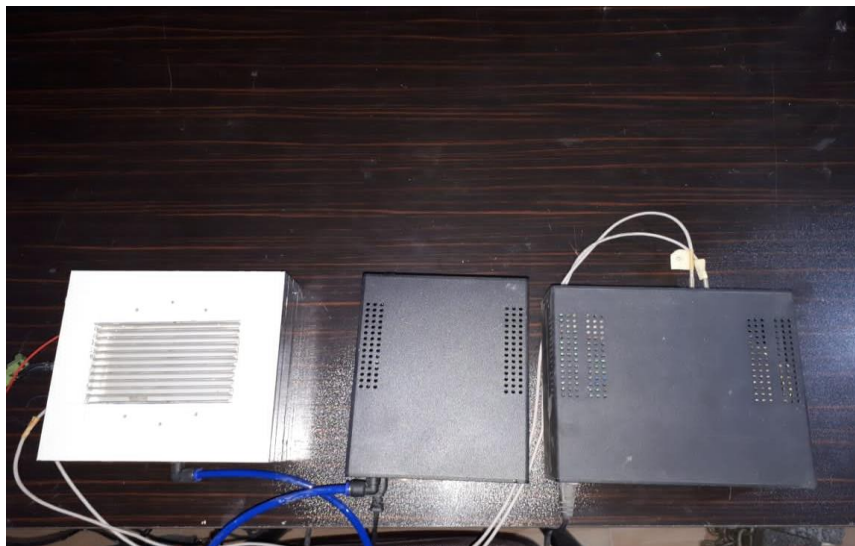


Figure 8. The output and input of nitrogen oxide gas from the tank to the surface of the sample in the test device to determine the amount of reduction of nitrogen oxide adjacent to the samples in the tested designs.



Figure 9. Placing the concrete sample in the device to carry out the photocatalytic activity of concrete to absorb nitrogen oxide gas around the surface of the sample in the test device.

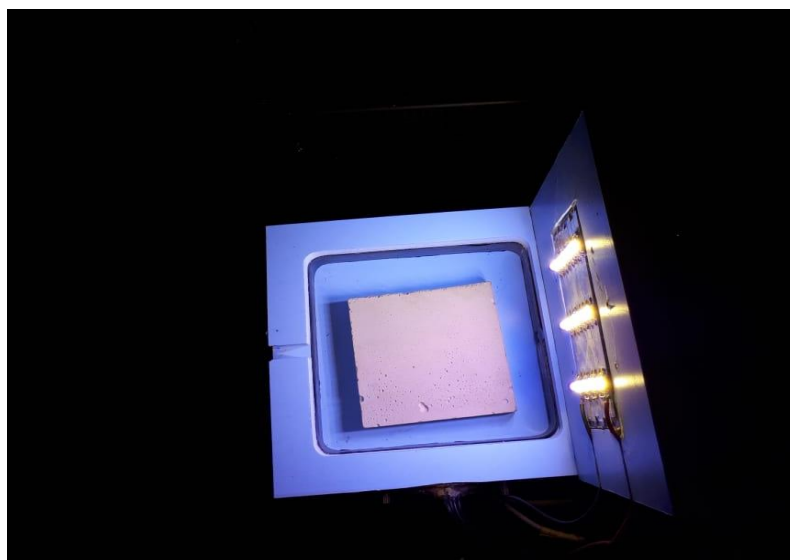


Figure 10. Radiant light spectrum on the surface of concrete to carry out the photocatalytic activity of concrete to absorb nitrogen oxide gas around the surface of the sample in the test device.



Figure 11. Starting the test for the radiant light spectrum on the surface of concrete to perform the photocatalytic activity of concrete to absorb nitrogen oxide gas around the surface of the sample in the test device.

Table 4. The density and amount of nitrogen oxide adjacent to the samples in the tested designs

Mix No.	Density(kg/m ³)	Density (kg/m ³)	NO Base Conc. (PPb)
1	1901	1896	2005
2	1908	1905	2001
3	1911	1917	2003
4	1925	1933	2004
5	1936	1941	2000
6	1945	1940	1997



Table 5- Nitrogen oxide rate of the samples in the tested plans

Mix No.	NO Base Conc. (PPb)	Output NO Conc. (PPb)		
		Specimen 1	Specimen 2	Specimen 3
1	2005	1896	1888	1889
2	2001	1893	1895	1889
3	2003	1884	1887	1881
4	2004	1759	1762	1765
5	2000	1760	1752	1753
6	1997	1746	1750	1741

Table 6- Nitrogen oxide absorption percentage of the samples in the tested designs

Mix No.	NO Removal (%)	NO Removal (%)	NO Removal (%)
1	5.4	5.8	5.8
2	5.6	5.5	5.8
3	6.0	5.9	6.2
4	12.3	12.1	12.0
5	12.2	12.6	12.6
6	12.9	12.7	13.2

2. Results

The use of titanium dioxide nanoparticles up to 5% of cement replacement, increases the speed of self-cleaning properties of concrete surfaces. After 24 hours of using UV radiation, it was found that the color of the soot impregnated on the concrete surface decreases and this is due to the photocatalytic reaction of titanium dioxide exposed to ultraviolet radiation. By adding a higher percentage of titanium dioxide, the reduction of color caused by soot on the concrete surface was determined.

6 samples each with 2 water absorption percentage tests and 3 nitrogen oxide absorption tests make the number of the statistical population to 30. To perform these tests, UV tests were taken from the samples. This test will be done according to ASTM C642 method. The addition of the highest percentage of titanium dioxide compared to other percentages (5%) and 2% nanosilica causes greater density and the highest percentage of nitrogen oxide absorption up to 13.2% of the original sample. As a result, adding a higher percentage of titanium dioxide up to 5% and nanosilica up to 2% will be effective in reducing and absorbing about 30% of air nitrogen dioxide, which can be used in environmental concrete for sustainable development.



3. Acknowledgement

The authors of the article would like to thank Ardabil Branch, Islamic Azad University for supporting this research.

4. Additional Information and Declaration

Funding:

There was no funder for this study. Grant Disclosures There was no grant funder for this study. Competing Interests The authors declare there is no competing Interests, regarding the publication of this manuscript.

Author Contributions:

Foroozan Mostofi: Proposed the plan, conceived the experiments, analyzed the data, prepared figures, and tables, authored or revised drafts of the paper, Approved the final draft.

Fatemeh Nasehi: Conceived and designed the experiments, analyzed the data, contributed reagents /materials/analysis tools, prepared figures, and tables. Analyzed the data, prepared figures, and tables, authored or revised drafts of the paper, Approved the final draft.

Babak Pordel Maragheh: Analyzed the data, prepared figures, and tables, authored or revised drafts of the paper, Approved the final draft.

Ebrahim Fatae: Analyzed the data, prepared figures, and tables, authored or revised drafts of the paper, Approved the final draft.

Mehdi Nezhadnaderi: Analyzed the data, prepared figures, and tables, authored or revised drafts of the paper, Approved the final draft.

5. Ethics Statement

The study was conducted by Ardabil Branch Islamic Azad University for the thesis of Sciences and Environmental engineering.

6. Supplemental Information

There is no supplementary information on this paper. Any questions and request for more information should be addressed on correspondence author.

References

1. Singh, G., Saini, B. Nanomaterial in cement industry: a brief review. *Innov. Infrastruct. Solut.* 7, 45 (2022). <https://doi.org/10.1007/s41062-021-00649-z>.
2. Wei, Yuanchen, et al. "Recent advances in photocatalytic self-cleaning performances of TiO₂-based building materials." *RSC advances* 13.30 (2023): 20584-20597.
3. https://en.wikipedia.org/wiki/Jubilee_Church.
4. Ghosh, Sudipta, Amiya K. Samanta, and Ashok K. Sahani. "Effect of Elevated Temperature on Diverse Properties of Concrete Containing Waste Materials:



DIVERSE PROPERTIES OF CONCRETE CONTAINING WASTE

MATERIALS." *Indian Journal of Engineering and Materials Sciences (IJEMS)* 30.2 (2023): 195-211.

5. Revathi, S., D. Brindha, and R. Harshani. "Effect of incorporating fibers in reactive powder concrete—A review." *Materials Today: Proceedings* (2023).
6. Ren, Zunchao, et al. "Optimizing the content of nano-SiO₂, nano-TiO₂ and nano-CaCO₃ in Portland cement paste by response surface methodology." *Journal of Building Engineering* 35 (2021): 102073.
7. Hakeem, Ibrahim Y., Fadi Althoey, and Akter Hosen. "Mechanical and durability performance of ultra-high-performance concrete incorporating SCMs." *Construction and Building Materials* 359 (2022): 129430.
8. Casagrande, César Augusto, Wellington Longuini Repette, and Dachamir Hotza. "Effect of environmental conditions on degradation of NO_x gases by photocatalytic nanotitania-based cement mortars after long-term hydration." *Journal of Cleaner Production* 274 (2020): 123067.
9. Ahmad, Soran Abdrahman, Serwan Khwrshed Rafiq, and Rabar H. Faraj. "Evaluating the effect of waste glass granules on the fresh, mechanical properties and shear bond strength of sustainable cement mortar." *Clean Technologies and Environmental Policy* (2023): 1-20.
10. Maslesa, Esmir, Per Anker Jensen, and Morten Birkved. "Indicators for quantifying environmental building performance: A systematic literature review." *Journal of building engineering* 19 (2018): 552-560.
11. Nayak, Dheeresh Kumar, et al. "Fly ash for sustainable construction: A review of fly ash concrete and its beneficial use case studies." *Cleaner Materials* (2022): 100143.
12. Gavela, S., et al. "Multifactorial experimental analysis of concrete compressive strength as a function of time and water-to-cement ratio." *Procedia Structural Integrity* 10 (2018): 135-140.