Photocatalytic mortar with tiO₂ for the reduction of air pollutants produced by vehicular emissions Mortero fotocatalítico con tiO₂ para la reducción de contaminantes del aire producidas por emanaciones vehiculares

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Abstract

Photocatalytic mortar with TiO2 leads to a reduction in air pollution due to vehicle emissions. For this purpose, the experimental method was used, which consisted of the preparation of mortars with the same proportion of 1: 4, a strength of 145 kg / cm2 and with different percentages of itanium dioxide (0% and 10%), which were evaluated under the same conditions through the following tests: Quantity of polluting gases, compressive strength and photocatalytic capacity. The results obtained—reduction of carbon dioxide by 97.9%, hydrogen sulfide by 72.9%, sulfur dioxide by 67.2%, nitrogen monoxide by 63.4%, carbon monoxide by 40.5% and oxygen recovery by 7.7%—confirmed the performance of the photocatalytic process through titanium dioxide (TiO₂) in terms of an improvement in air quality, and the reduction of colorants, rhodamine by 89.10% and methylene blue 53.06% confirmed its self-cleaning capacity, thus improving the reduction of air pollution.

Keywords: Photocatalytic mortar, air quality, TiO2, decontaminant

Resumen

El mortero fotocatalítico con TiO₂ conlleva a la reducción de la contaminación del aire por emisiones procedentes de vehículos en Lima, Perú. Para este fin se utilizó el método experimental que consistió en la elaboración de morteros con una misma proporción de 1:4, una resistencia de 145 kg/cm2 y con diferentes porcentajes de dióxido de titanio (0% y 10%), los cuales fueron evaluados en las mismas condiciones por los siguientes ensayos: Cantidad de gases contaminantes, resistencia a la compresión y capacidad fotocatalítica. Los resultados obtenidos, reducción del dióxido de carbono en un 97.9%, el sulfuro de hidrógeno en un 72.9%, el dióxido de autre en un 67.2%, el monóxido de nitrógeno en un 63.4%, el monóxido de carbono en un 40.5% y recuperación del oxígeno en un 7.7%, confirmaron el rendimiento del proceso fotocatalítico a través del dióxido de titanio (TiO₂) en términos de una mejora en la calidad del aire, y la reducción de los colorantes, rodamina en un 89.10% y el azul metileno en un 53.06%, confirmaron la capacidad autolimpiante del mismo mejorando de esta manera la reducción de la contaminación del aire.

Palabras clave: Mortero fotocatalítico, calidad del aire, dióxido de titanio, descontaminante

1. Introduction

According to World Health Organization data showing that nine out of ten people breathe air containing high pollutant levels, air pollution levels are still dangerously high in many parts of the world. These updated estimates alarmingly show that seven million people die each year from exposure to fine particles in the air that penetrate deep into the lungs and cardiovascular system, causing diseases such as stroke, heart disease, lung cancer, chronic obstructive pulmonary disease and respiratory infections, including pneumonia. (Osserian and Lindmeier, 2018) report that more than 90% of air pollution-related deaths occur in low- and middle-income countries, mainly in Asia and Africa, followed by low- and middle-income countries in the Eastern Mediterranean, Europe and the Americas. (Peláez and Ullauri, 2021) claim that green cleaning is a worldwide concern. For this reason, the U.S. Green Building Council (USGBC), following the guidance of other international councils, developed LEED "Leadership in Energy and Environmental Design" as a rating system for buildings that require a "green cleaning policy." Therefore, it is important to give continuity to these environmental contributions.

According to the WHO (Zanatta, 2019), Peru is one of the most polluted countries in Latin America; around five thousand deaths are registered annually due to air pollution, with Lima, the Peruvian capital, being one of the most polluted cities in Latin America and having one of the worst air quality in the region. The current problem is described by (Prada, 2021), who points out that atmospheric pollution has the greatest negative consequences. This is mainly due to the emission of polluting gases into the atmosphere by anthropogenic activity. Atmospheric pollutants can cause many harmful chemical reactions; therefore, their removal should be a priority. The purpose is to contribute to environmental aspects as this factor affects the concrete by 34.3% in relation to its quality, according to (Orozco and Avila, 2018).

 TiO_2 nanoparticles can accelerate cement hydration and improve the strength development of cementitious materials at room temperature. However, the performance of cementitious materials containing TiO_2 nanoparticles at low temperatures is still unknown (Chavarry et al., 2021). The benefit of the photocatalytic properties of TiO_2 (titanium dioxide) in coating mortar occurs when it is exposed to UV light. Air-purifying, self-cleaning and antimicrobial properties can be generated spontaneously and simultaneously on the surface of the material that contains it. This is because titanium dioxide is a photocatalytic material with an electronic structure composed of two bands, the valence band (full of electrons) and the conduction band (no electrons).

Regarding the energetic aspect, it is important to mention that the conduction band and the valence band form the so-called forbidden band. When a photon with higher energy than this band comes into direct contact with this photocatalytic material, an electron (e-) from the valence band moves towards the conduction band, thus leaving an electronic band gap (h+). A portion of the photo-excited band gap-electron pair diffuses to the surface of the photocatalytic material, where it is retained to participate in chemical reactions with oxygen and water molecules present in the environment. The electronic band gaps (h+) can react with adsorbed donor molecules such as water to produce hydroxyl radicals (highly reactive). Acting as an electron acceptor, oxygen in the air can react with electrons to form superoxide radical anions (O_2 -). The hydroxyl radicals (oxidizing) and superoxide radical anions (reducing) generated on the surface of TiO₂ have demonstrated a great ability to degrade different types of microorganisms, almost all types of organic pollutants and other inorganic compounds such as NOx and SO₂ (Maury and De Belie, 2010).

The performance of photocatalysis in urban applications is determined by several environmental factors such as the intensity of incident radiation, relative humidity, temperature and wind. This also depends on other variables such as the photocatalyst when associated with cement-based support, as well as the porosity, type and size of aggregates, application method, the quantity applied and aging (Shen et al., 2012). Also, the adsorption of pollutants on the active centers of TiO₂-cement composites has been identified as the determining factor of photocatalytic efficiency (Chen et al., 2011). Therefore, the purpose of the study was to demonstrate the effects of TiO₂ photocatalytic mortar on the reduction of air pollution from vehicle emissions.

2. Discussion and Analysis

The analysis comprises three study categories oriented to environmental factors of "air," photocatalytic capacity and compressive strength. For this purpose, dosages were projected in simple (0%) and photocatalytic (10%) mortars taking into account the characteristics of fine sand, cement and titanium dioxide, designing an f'c of 145kg/cm² and a cement/sand ratio of 1:4. Then, specimens were prepared for each sample (M1, M3, M5 without titanium dioxide and M2, M4, M6 with titanium dioxide) according to the specifications of each test, which complements the research design of (Moreno, 2018) as shown in the (Table 1):

Materials	М1	М2	М3	M4	M5	M6				
Materials	Quantity (kg)									
Cement	4.671	4.624	1.196	1.184	0.49	0.485				
Sand	16.857	16.687	4.315	4.272	1.768	1.75				
Water	3.698	3.661	0.947	0.937	0.388	0.384				
Ti02		0.462		0.118		0.048				

Table 1. Mixture dosing in evaluation specimens

The referred analysis of pollutant gases (based on the UNE-ISO 22197-1 standard) as shown in (Figure 1), samples M1 and M2 were placed in a hermetic glass urn injecting pollutant gases into it and placing the cannulas of the measurement devices to proceed with the nine readings (in real time) of the decrease of pollutants by the action of photocatalysis.



Figure 1. Test of quantity of pollutant gases

The (Table 2) shows the results for sample M2. On average, the polluting gases were reduced except for oxygen, which showed negative values. This phenomenon is explained by its increased concentration due to the degradation of the other gases by the action of photocatalysis. Sample M1 showed slight variations on average due to the erratic movement of the gases inside the glass urn. In sample M2, carbon dioxide (CO₂) decreased by 97.9%, hydrogen sulfide (H2S) decreased by 72.9%, sulfur dioxide (SO₂) decreased by 67.2%, nitrogen monoxide (NO) decreased by 63.4%, carbon monoxide (CO) decreased by 40.5%, and oxygen increased by 8.5%. The authors (Ramirez and Wesso, 2019) conclude that there is a degradation of CO₂ ions caused by light factors and suitable climatic conditions. In addition, as a complement to these authors, the present research proposes the analysis of the degradation of gases such as SO₂, CO, NO, O₂, as shown in (Table 2).

Percentage variation in gases													
	Time	co		H2S		O2		SO2		NO		CO2	
Med.	fed. (h) (ppm)		(p	(ppm) (%)		(ppm)		(ppm)		(ppm)			
		M1	M2	M1	M2	Ml	M2	Ml	M2	Ml	M2	Ml	M2
1	01:00	2.00%	40.00%	2.70%	57.00%	1.30%	-6.20%	1.00%	100.00%	0.00%	100.00%	0.50%	88.50%
2	01:00	0.00%	44.40%	-	-	0.00%	-15.60%	8.70%	-	0.30%	57.90%	0.00%	100.00%
3	01:00	0.00%	26.10%	2.70%	68.40%	0.60%	-10.80%	0.00%	36.50%	2.50%	55.60%	0.50%	100.00%
4	01:00	0.00%	19.30%	0.00%	68.80%	1.30%	-10.10%	4.00%	64.50%	0.60%	63.50%	0.00%	97.00%
5	01:00	0.00%	37.30%	0.00%	43.50%	0.00%	-8.60%	-1.00%	70.80%	8.70%	56.10%	7.40%	100.00%
б	01:00	0.00%	44.60%	0.00%	-	0.70%	-5.40%	1.00%	65.70%	0.00%	46.70%	0.00%	-
7	01:00	0.00%	35.80%	0.00%	100.00%	0.70%	-4.50%	0.00%	63.60%	0.00%	52.20%	0.30%	100.00%
8	01:00	0.00%	17.70%	1.50%	-	0.00%	-2.10%	0.00%	60.00%	0.00%	50.00%	11.20%	-
9	01:00	0.00%	99.00%	-5.60%	100.00%	0.60%	-12.80%	1.00%	76.10%	0.00%	88.60%	4.00%	100.00%
Aver	01:00	0.20%	40.50%	0.20%	72.90%	0.60%	-8.50%	1.60%	67.20%	1.30%	63.40%	2.70%	97.90%

able 2. Alteratior	n of gases -	Samples	M1	and	М2
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In the analysis of photocatalytic capacity (UNI-11259), when Rhodamine B and methylene blue dyes were applied to samples M3 and M4 with the Colorimeter, readings were obtained at 0 hours ($a^{*}(0h)$). Then the samples were placed under UVA light according to standards. Finally, readings were taken after 4 hours ($a^{*}(4h)$) and 26 hours ($a^{*}(26h)$) of exposure to UVA light to calculate R4 and R26 factors for the rhodamine dye, where the color variation is measured on the a* axis in the CIE L * a* b* color system, with the (Equation 1)(Equation 2):

$$R_{4} = \frac{a^{*}(0h) - a^{*}(4h)}{a^{*}(0h)} \times 100$$
(1)
$$R_{26} = \frac{a^{*}(0h) - a^{*}(26h)}{a^{*}(0h)} \times 100$$
(2)

R4 and R26 factors were determined for the dye Methylene Blue and Rhodamine, both by performing analyses with the proposed mixture designs with and without TiO_2 , considering that the color variation is measured on the b* axis in the CIE L* a* b* color system. A mortar is considered to be photocatalytic and self-cleaning if the R4 factor value is greater than 20% and the R26 factor is greater than 50%. See (Figure 2).



Figure 2. Photocatalytic capacity test

(Figure 3) shows the comparison of the average R4 for both samples, M3 and M4. In the case of rhodamine, sample M3 shows 3.14%, which is below the minimum level with a difference of 16.86%, while sample M4 obtained a result of 35.88%, exceeding the minimum level with 15.88%. In the case of methylene blue, sample M3 obtained a result of 3.13%, which is below the minimum level with a difference of 16.87%. In comparison, sample M4 obtained a result of 21.60% exceeding the minimum level with 1.60%. These indicators compared to (Zhang and Hou, 2015) show the use of 5% of TiO₂ to obtain results. However, in the current research with regard to TiO₂/Cement ratios of 9.99%, 9.97% and 9.90% were obtained for the analyzed sample. The assumption is that these ratios were obtained due to climatic reasons and the time of analysis for these first samples since they took four days to develop.



Figure 3. R4 factor of rhodamine in samples M3 and M4

(Figure 4) shows the variations of the R26 factor (reduction at 26 hours of exposure to UV A light) for methylene blue. In sample M3, the highest value for the R26 factor was found in specimen 2 with 11.11%, and the lowest value was found in specimen 1 with 1.29%; while in sample M4, the highest value was found in specimen 1 with 56.70% and the lowest value was found in specimen 6 with 50.32%. Also, the percentages of the specimens of sample M3 are below the minimum percentage (50%) which determines whether the mortar is self-cleaning. In the case of rhodamine, sample M3 obtained a result of 7.66%, which is below the minimum level with a difference of 42.34%. In comparison, sample M4 obtained a result of 77.79%, exceeding the minimum level with 27.79%. These results contrast with the research of (Ullauri and Solano, 2021) according to the premise that photocatalytic mortars possess self-cleaning attributes capable of degrading different atmospheric pollutants.



Figure 4. Average R26 factor for rhodamine and methylene blue in samples M3 and M4

Therefore, the sample M4 for rhodamine and methylene blue exceeded the minimum level in both R4 and R26 factors. According to UNI 11259 standard, it corresponds to a photocatalytic and self-cleaning mortar, which indicates that the residues of the gas degradations that remain in the mortar are easy to clean by rain or by spraying it with water, thus helping to improve air quality. Regarding the results obtained, the deduction of (Janus and Zając, 2016) is analyzed, which indicates that the probability of the self-cleaning effect occurs due to the shielding of the TiO₂ nanoparticles by carbonate precipitates and the clogging of the surface pores. In addition to this thesis, it is mentioned that the self-cleaning effect is simplified with the use of rhodamine and methylene blue, thus demonstrating this effect in humid conditions and at a temperature of approximately 25°C. This finding adds support to the authors' analysis since the analysis was carried out in frigid weather conditions.

The compressive strength of samples M5 and M6 was determined through axial studies of cores (MTC E 609 / NTP 334.051). Regarding the standard, breaks were made at 14 days, taking into account that the specimens reach

a significant percentage (85%) of the designed strength at that age. For comparison purposes, the strength that the specimens of each sample would have at 28 days (100% of the designed strength) was projected.



Figure 5. Compression strength test

(Figure 5) shows that in sample M5 the highest value for the strength at 14 days was 200 kg/cm² (138% of the F'c), and the lowest value was 142 kg/cm² (98% of the F'c), while in sample M6, the highest value was 159 kg/cm² (110% of F'c) and the lowest values were specimen 1 with 113 kg/cm² (78% of F'c) and specimen 2 with 105 kg/cm² (72% of F'c). The results taken at 20° C, also researched by (Hongliang and Gao, 2018), support the statement that hydration is fundamental for the addition of TiO₂ to the binder. It is important to mention that the behavior in hot climates benefits the absorption and performance of the analyzed samples, according to the proposed design (145 kg/cm²). The hypothesis was based on the strength reaching or exceeding 123.3 kg/cm² (85% of the F'c) at 14 days. In the case of sample M5, all the specimens exceeded this percentage, while two of the specimens of sample M6 were below this percentage and the compressive strength at 28 days, where the specimens must reach at least 100% of the designed strength—in this case, 145 kg/cm²—to determine if adding TiO₂ compromises the performance of the mortar.



Figure 6. Projected compressive strength at 28 days of samples M5 and M6

(Figure 6) shows that sample M5 has the highest value for the strength at 28 days. It was the first specimen with 235 kg/cm² (162% of the F'c), and the lowest value was 167 kg/cm² (115% of the F'c), while in sample M6, the highest value was 187 kg/cm² (129% of the F'c). The lowest values were specimen 1 with 133 kg/cm² (92% of F'c) and specimen 2 with 123 kg/cm² (85% of F'c). Specimens 1 and 2 of sample M6 show that despite being projected at 28 days, they do not reach the designed strength value. When comparing the averages of both samples (M5 and M6), it can be observed that they comply with the design value at 14 days since they exceed the expected strength of 123 kg/cm², and even when projecting the compressive strength at 28 days, both samples exceed the design strength of 145 kg/cm². The results complement the statement of (Saleem and Abdullah, 2021) as the authors provide the following statement: "Adding TiO₂ to cement could contribute to improving mechanical properties, such as compressive strength and durability of concrete." For this statement, the performance of the concrete will depend closely on the proposed mix design and the expected strength. Consequently, compared to the expected strength, the concrete with TiO₂ addition increases the compressive strength by 28.9%. See (Figure 7).



Figure 7. Average compressive strength of samples M5 and M6 projected at 28 days

Finally, based on the tests carried out, a correlation was established between the percentage of TiO_2 and the reduction of contamination. As additional factors, the compressive strength of the mortar decreases with a 10% increase in TiO_2 .

(Table 3) shows that the increase in the percentage of titanium dioxide (TiO_2) increases the reduction percentage of gases of CO (from 0.2% to 40.5%), H2S (from 0.2% to 72.9%), SO₂ (from 1.6% to 67.2%), NO (from 1.3% to 63.4%) and CO₂ (from 2.7% to 97.9%). In the case of O₂, it increases its concentration (from 0.6% to -8.5%). It is also observed that increasing the percentage of titanium dioxide (TiO₂) decreases the compressive strength from 214 to 158 kg/cm². Therefore, the correlation between the percentage of TiO₂ and the reduction of air pollution is direct, while the correlation between the percentage of TiO₂ and the compressive strength is inverse.

TiO ₂ % ↑	Reduction of contamination %							
	СО	H2S	O_2	SO_2	NO	CO_2	(kg/cm ²) ↓	
0.0%	0.2%	0.2%	0.6%	1.6%	1.3%	2.7%	214	
10.0%	40.5%	72.9%	-8.5%	67.2%	63.4%	97.9%	158	

Table 3. TiO2 % Ratio - Reduction of contamination - Compressive strength

3. Conclusions

Firstly, we conclude that the photocatalytic mortar with TiO2 has a positive effect since, on average, the pollutant gases were reduced; carbon dioxide (CO₂) decreased by 97.9%, hydrogen sulfide (H2S) decreased by 72.9%, sulfur dioxide (SO₂) decreased by 67.2%, nitrogen monoxide (NO) decreased by 63.4% and carbon monoxide (CO) decreased by 40.5%. In the case of oxygen (O₂), there was an increase of 8.5%. These results under

a mixture design with the addition of TiO_2 in weight 0.001037 m³ contrast with the conclusions of (Escobar and Solis, 2021), which indicate that catalyzation is obtained from thin layers. This comparison seeks to contribute to the development of new material technologies since TiO_2 can be applied by disaggregating with Zn modifications that reduce the pollution produced by vehicles and improve air quality.

Secondly, we conclude that the self-cleaning property of the photocatalytic mortar with TiO₂ has a positive effect, given that the sample M4 for rhodamine (R4: 35.88% and R26: 77.79%) and methylene blue (R4: 21.60% and R26: 52.63%) exceeded the minimum levels in R4 (20%) and R26 (50%), which are the main indicators that define the mortar as photocatalytic and self-cleaning. These results showed a significant change at the time of the observation. This attribute facilitates cleaning of the gas degradation residues that remain in the mortar by the effect of rain or by spraying water, thus helping to improve air quality. In addition, there is evidence that these photoactive changes can occur up to 65 hours of control, as reported by (J. I. Tobón et al., 2020) in his research. These results exceed expectations since these changes may depend on the exposure of the mortar to light, the curing and its dosage. Thirdly, we conclude that the increase in the percentage of titanium dioxide (TiO_2) increases the reduction percentages of gases of CO (from 0.2% to 40.5%), H2S (from 0.2% to 72.9%), SO₂ (from 1.6% to 67.2%), NO (from 1.3% to 63.4%) and CO₂ (from 2.7% to 97.9%), as well as the percentage increase of O_2 (from 0.6% to 8.5%). Also, when the percentage of titanium dioxide (TiO₂) increases, the compressive strength decreases from 214 to 158 kg/cm2. It is important to mention that even though there is a decrease in strength, these data are positive as the expected strength was 145 kg/cm2 as well as in contrast to other authors who obtained lower strengths close to the above mentioned as 10.96 Mpa or 111.82 kg/cm2 as concluded by (Oviedo and Mejia, 2019). Therefore, the correlation between TiO_2 percentage and air pollution reduction is direct, while the correlation between TiO_2 percentage and compressive strength is indirect.

Finally, we conclude that the photocatalytic mortar with TiO_2 has a positive effect on the reduction of air pollution since it reduces the polluting gases of CO (from 0.2% to 40.5%), H2S (from 0.2% to 72.9%), SO₂ (from 1.6% to 67.2%), NO (from 1.3% to 63.4%) and CO₂ (from 2.7% to 97.9%) from vehicle emissions and it improves air quality by self-cleaning, reducing the concentration of rhodamine dyes by 77.79% and methylene blue by 52.63%.

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