Pervious concrete (PC) with adition of TiO2 used in sanitary sewage treatment

Concreto permeable (CP) con adición de TiO2 utilizado en el tratamiento de aguas residuales sanitarias

W. Melo Prudêncio de Araújo* http://orcid.org/0000-0003-2517-6996
G. Barreto Sandoval^{1**} http://orcid.org/0000-0002-8702-8844
M. de Souza Picanço* http://orcid.org/0000-0001-7313-1229
C. Zoe Correa***
A. Negrão Macêdo* https://orcid.org/0000-0002-6097-8620

* Universidade Federal do Pará, Pará - BRAZIL

** Universidad Católica del Norte, Antofagasta, CHILE

*** Universidade Estadual de Londrina, Estado de Paraná, BRAZIL

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Abstract

Pervious concrete (PC) is considered a solution to reduce the effects of heat islands, runoff problems, and the concentrations of pollutants present in the water; however, its potential as a filter bed is used in the treatment of sanitary sewage has never been evaluated. This work aimed to assess the influence of photocatalytic TiO2 addition in PC on the concentration of total phosphorus, total ammonia, nitrate, total solids, and turbidity for application in the treatment of sanitary sewage. Sunlight was used as a radiation source. A flat photocatalytic reactor was built, in which 100 liters of raw sewage were pumped, with a flow of 100 L/min over the PC slabs, covering a total area of 1 m^2 . The TiO2 concentrations added to the PC were 3, 6, and 10%. The results indicate efficiency in the adsorption of total phosphorus, total ammonia, nitrate, total solids, and turbidity. Thus, using PC associated with 10% TiO2 can add efficiency to the sanitary sewage treatment process by maintaining good mechanical and hydraulic behavior.

Keywords: Pervious concrete; heterogeneous photocatalysis; titanium dioxide; sanitary sewage.

Resumen

El concreto permeable (CP) se considera una solución para reducir los efectos de las islas de calor, los problemas de escorrentía y las concentraciones de contaminantes presentes en el agua; sin embargo, nunca se ha evaluado su potencial como lecho filtrante en el tratamiento de aguas residuales sanitarias. Este trabajo tuvo como objetivo evaluar la influencia de la adición fotocatalítica de TiO2 en CP sobre la concentración de fósforo total, amoníaco total, nitrato, sólidos totales y turbidez para su aplicación en el tratamiento de aguas residuales sanitarias. La luz del sol se utilizó como fuente de radiación. Se construyó un reactor fotocatalítico plano, en el cual se bombearon 100 litros de aguas residuales sin tratar, con un caudal de 100 L/min sobre las placas de PC, cubriendo un área total de 1 m². Las concentraciones de TiO2 añadidas al PC fueron 3, 6 y 10%. Los resultados indican eficiencia en la adsorción de fósforo total, amoníaco total, nitrato, sólidos totales y turbidez. Por lo tanto, el uso de CP asociado con 10% de TiO2 puede agregar eficiencia al proceso de tratamiento de aguas residuales sanitarias al mantener un buen comportamiento mecánico e hidráulico.

Palabras clave: Concreto permeable; fotocatálisis heterogénea; dióxido de titanio; aguas residuales sanitarias.

1. Introduction

Pervious concrete (PC) is a non-fine concrete, characterized by its interconnected pores, which have the possibility of allowing the passage of water through its structure efficiently and safely. In view of this hydraulic characteristic, the material has been widely used for covering sidewalks and paving, responding to the problem of managing runoff (floods). Thus, the PC makes a great contribution to sustainability in many areas of civil construction; however, its use in the area of environmental sanitation (water and sewage treatment) is still being studied. Due to its high porosity (15-30%), the material can be used as a layer (filter) in which the effluent can be treated (percolate) with a chemical biological and physical treatment.

¹ Corresponding author:

Departamento de Gestión de la Construcción, Universidad Católica del Norte

E-mail: gersson.barreto@ucn.cl

The advanced oxidative process (AOP) can be a technique to be used when seeking the application of PC in the chemical treatment of sewage. These processes are defined as those capable of generating hydroxyl radicals (•OH), which have a high oxidation potential, and other oxidants, such as fluorine, ozone, and hydrogen peroxide. The hydroxyl radical is non-selective, allowing the degradation of several classes of compounds, reducing them to less severe compounds or even CO2, water, and mineral ions. In addition, they can be used to destroy organic compounds in both aqueous and gas phases. The high efficiency of these processes can be attributed to their high reduction potential ($E^\circ = 2.8 V$), lower potential only if compared to fluorine ($E^\circ = 3.0 V$) (Antonopoulou et al., 2021).

Among the POA, heterogeneous photocatalysis stands out, a process that involves redox reactions induced by radiation, on the surface, of mineral semiconductors (catalysts) such as TiO2, CdS, ZnO, and Fe2O3 (Ferreira, 2014). Among the semiconductors mentioned, due to its physicochemical properties, such as water insolubility, chemical stability, durability, low toxicity, and low cost, TiO2 is the most used (Nakata, 2012), (Antonopoulou et al., 2021).

Concrete with TiO2 is used in sidewalks, paints, concrete panels, and tiles. When added to concrete, TiO2 keeps the surface selfcleaning, eliminates biological organisms such as algae, bacteria, fungi and degrades airborne pollutants such as nitrogen oxides (NOx) (Augugliaro et al., 2010), (Liu et al., 2021). According to (Noeiaghaei et al., 2017), the incorporation of nanomaterials such as TiO2 in the concrete matrix or in coating formulations, even in small amounts, results in greater strength to biodeterioration and greater durability of concrete structures used in sewage systems.

Semiconductor photocatalysis based on TiO2 is a widely investigated advanced oxidative process for the degradation of a wide range of organic contaminants and microorganisms, and it is an effective photocatalyst when exposed to ultraviolet light (~ 300-400 nm) (Nakata, 2012). However, there is still a gap regarding the incorporation of this chemical in new semipermeable materials applied in the treatment of sanitary sewage, such as PC.

In developing countries, such as Brazil, the lack of sewage treatment is a high problem. About 54.1% of Brazilians have access to the sewage collection network and, of this collected percentage, only 49.1% is treated. Among the regions of the country, the most affected is the north and northeast, where only 12.3% and 28,5% of the population, respectively, have access to the collection network (as collected by the national information system on sanitation of Brazil in 2019). The data presented demonstrate the fragility of the existing treatment system in Brazil, as well as the need to develop alternative and efficient technologies.

Thus, this work aimed to produce a PC with TiO2 addition, for application in the treatment of sanitary sewage. For this, it was necessary to develop a flat plate reactor to analyze and quantify the efficiency of the photocatalytic activity of PC with the addition of TiO2.

2. Materials and Methods

2.1 Experimental program

The experimental program of this research was divides in two phases, as shown in (Figure 1). Phase 1 to verify the mechanical and hydraulic properties of the PC produced, and the second phase to obtain the sewage analysis of the PC with TiO2 and realize the microstructural analysis of the PC with TiO2 before and after the sewage treatment.

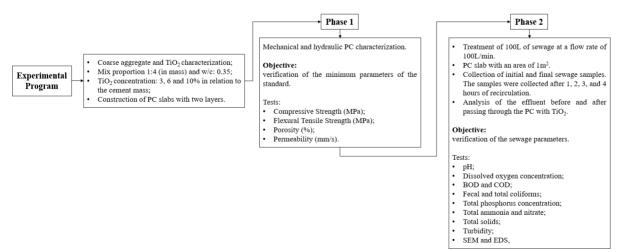


Figure 1. Experimental program

2.2 Materials

Portland cement with high initial strength (CP-V ARI RS Brazilian denomination) was used, in accordance with the Brazilian standard NBR 16697/18. This cement was chosen due to its property of resisting to aggressive sulfated media, such as those found in sewage, wastewater, or industrial networks, in seawater and in some types of soils.

As coarse aggregate, crushed stone of basaltic origin was used, with a single particle size of 2.36 mm (uniform granulometry distribution). Bulk specific gravity is 2.8 and unit weight 1,400 kg/m³, verified from Brazilian standard NBR NM 53/09 and NBR NM 45/06, respectively.

For the cast of photocatalytic PCs, TiO2 P25 was used as a catalyst. Its chemical composition was determined by X-ray fluorescence by dispersive energy (XRF). The mineralogical characterization to identify the crystalline phases present was carried out using the X-ray diffraction technique (XRD). The microstructural and morphological analysis of the TiO2 surface was performed by scanning electron microscope (SEM) and the identification of the chemical elements present by the X-ray dispersive energy spectrometry system (EDS).

2.2.1 TiO2 characterization

The presence of 99.95% of Titanium was verified by the XRF of titanium dioxide, a result that corroborates with the XRD (Figure 2) of the sample that presents the characteristic phases of Titanium.

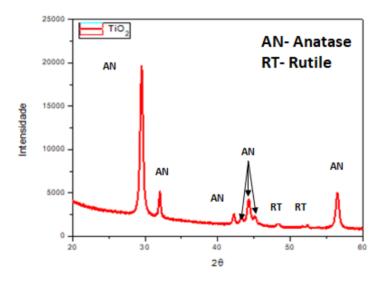


Figure 2. Diffractogram of Titanium Dioxide (TiO2) Adapted From (Araújo, 2020)

According to the micrographs obtained by scanning electron microscopy (SEM), at point A, the samples of TiO2 (Figure 3a) present particles with uniform distribution, spherical morphology, and slight agglomeration. There were no significant variations in grain morphology. According to (Casagrande, 2012), TiO2 powder tends to agglomerate and has a spherical shape; however, due to agglomeration, it does not present a well-defined shape.

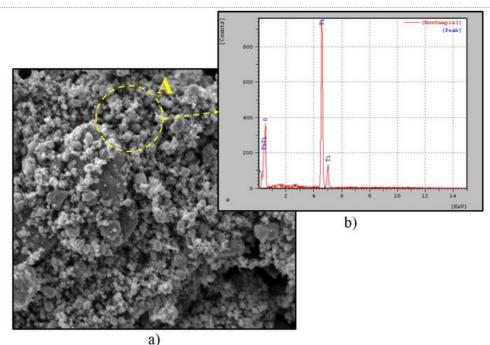


Figure 3. SEM results: (a) Image of TiO2 powder (400x); (b) EDS analysis of TiO2 powder Adapted from Araújo, 2020.

The graph of the TiO2 powder obtained by the dispersive energy spectroscopy (EDS) technique at point A (Figure 3b), shows high peaks of titanium (Ti) and peak of oxygen (O), which are the constituent elements of the TiO2 nanoparticles (Anandgaonker et al., 2019). The presence of these peaks in the sample confirms the purity found in the X-ray fluorescence analysis. A carbon peak (C) is observed in the EDS, which corresponds to the metallization tape used.

2.3 Mix proportion

Four PC mixtures were produced, and the respective mix proportions are provided in (Table 1). The specimens were prepared with 0% (reference), 3%, 6%, and 10% of TiO2 to replace the cement by mass. TiO2 concentrations in PC were based on the research of De (Melo et al., 2012).), when the efficiency of the incorporation of TiO2 in paving blocks was evaluated (Melo et al., 2012).

Mix	Water/cement ratio (w/c)	Cement (kg/m ³)	Aggregate (kg/m³)	TiO2 (kg/m ³)	Water (kg/m³)
P1	0.35	380.00	1520.11	0.00	133.00
P1.3.TiO2	0.35	368.60	1520.11	11.40	130.00
P1.6.TiO2	0.35	357.20	1520.11	22.80	126.00
P1.10.TiO2	0.35	342.00	1520.11	38.00	121.00

Table 1. Mix	proportion to	produce th	e PC mixes
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2.4 PC specimens and slabs casting and curing

Cylindrical (100mmx200mm) and prismatic specimens (100mmx100mmx400mm) were molded to evaluate the mechanical and the hydraulic properties of PC with the TiO2 produced. The production of the PC followed the procedure used by NBR 16416/15, and wet cure by immersion in water was used to test age according with the Brazilian standard NBR 5738/09.

The PC slabs were molded with the aid of a metallic mold with dimensions of 100 cm long by 25 cm wide and 6 cm high (Figure 4a). They were made using the mixture of TiO2 with the cement to produce PC slabs, which had two layers: the first one, a PC base without TiO2 incorporation, and the second one, a surface layer, with the addition of TiO2 (Figure 4b). To ensure greater homogeneity between TiO2 and cement, the mixture of the two materials was placed in a ball mill for two hours.

Before this time, the PC were produced according to the next procedure: first, 100% of the coarse aggregate was added to the concrete mixer with an additional 5% of the total cement weight and then the materials were mixed for one minute, then the remaining 95% cement and 100% water were added and mixed for three minutes, then the mixture was left to stand for three minutes and finally mixed one last time for two minutes. After the mix process the PC slabs were molded.

The photocatalytic PC slabs were molded with the aid of a metallic form (Figure 4a). According to Figure 4b, the slabs have two layers: a base layer without incorporating TiO2, and a surface layer, with the addition of TiO2. Each layer was compacted with a roller weighing 10 kg, passing 10 times to obtain the unit weight according to the Brazilian standard NBR 16416/15.

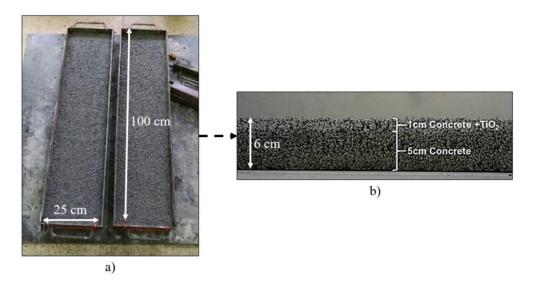


Figure 4. Profile of photocatalytic PC slabs: a) Geometric characteristics and b) Thickness distribution

2.5 Experimental phases

2.5.1 Phase 1

To evaluate the mechanical and hydraulic properties, the cylindrical and the prismatic specimens were used. The tests performed, the corresponding standard, the number of specimens, and the dimensions are shown in (Table 2).

Test	Standard/Method	Age	Specimens		
Test	Standard/ Method		Quantity*	Dimensions (mm)	Shape
Unit weight	ABNT NBR 9833:2009	6	6	100x200	Cylindrical
Porosity	Sandoval et al., 2019	28	6	100x200	Cylindrical
Permeability (constant head)	Sandoval et al., 2017	28	6	100x200	Cylindrical
Compressive strength	ABNT NBR 5739:2018	28	6	100x200	Cylindrical
Flexural tensile strength	ABNT NBR 12142:2010	28	6	100x100x400	Prismatic

Table 2. Mechanical and hydraulic properties evaluated in PC

*Number of specimens used per mix proportion PC used

2.5.2 Phase 2

To analyses the sewage filtered with the PD with TiO2, the tests were performed using a flat plate reactor (Figure 5a), with dimensions of 1.05 m long by 1.05 m wide and 1.10 m high. Four PC slabs were introduced, covering a total area of $1m^2$.

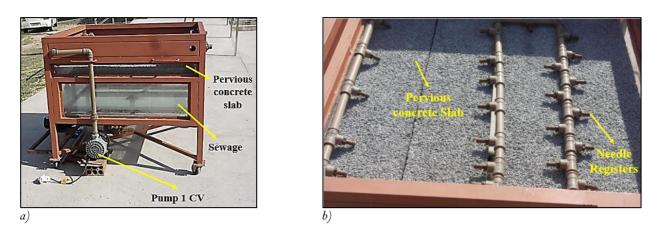


Figure 5. a) Flat plate reactor, b) Detail of the liquid dispensing device at the top of the slab

The tests consisted of pumping 100 liters of raw sewage, with a flow rate of 100 L/min over the PC slabs surface, with uniform distribution, so that the entire surface area was covered. The raw sewage percolated through the PC and, soon after, it was deposited in a reservoir located below the PC and, later, recirculated. For the release of the raw sewage to be treated on the PC slabs, a PVC pipe was made in which 23 needle registers with 17 mm in diameter were adapted to facilitate the rheology of the liquid, enabling the distribution of raw sewage over the entire length of the reactor (Figure 5b). For the pumping of raw sewage, a 1 CV centrifugal pump was used.

The operational variables of disinfection with photocatalysis were based on what was exposed in the research by Nogueira \mathcal{E} Jardim (1996) and are presented in (Table 3).

Table 3. Operational	variables of	^c disinfection	with photocatal	vsis.

Flow rate	Reactor volume	Hydraulic Detention	Thickness of the water sheet (m)	Radiation time
(L.min-1)	(L)	Time (HDT)(s)		(min)
100	10	6	0,01	24

The raw sewage used in the experiments came from the sewage treatment plant - ETE-Leste, located in Ininga, a neighborhood of Teresina-PI, and it is collected after the preliminary treatment: one optional aerated, two conventional optional (parallel to each other) and two maturing (parallel to each other) in this sequence. There is also a preliminary treatment consisting of grating, which intercepts the passage of coarse solids, and a sand grinder, which decants part of the sand present in the sewer.

There was no shading caused by nearby buildings or trees, to ensure solar use as the only source of radiation and without interferences.

All experiments were performed using PC slabs with 0%, 3%, 6%, and 10% TiO2 in relation to the cement mass of the mixture. The experiments lasted 4 hours, starting at 10 am and ending at 2 pm, all on clear, cloudless days. In addition to the initial and final raw sewage samples, spot samples were collected after 1, 2, and 3 hours of recirculation (reactor). In each collection, a total volume of 1.0 liter of raw sewage was removed for analysis.

The physical, chemical, and biological parameters, and their respective methods that were performed to characterize raw sewage are described in (Table 4). All parameters were analyzed in triplicate (n=3).

Parameters	Methods
1. pH	Potentiometric, APHA, 2012.
2. Dissolved Oxygen concentration	Winkler titration Azida Modification, APHA, 2012.
3. BOD and COD	Standard Flasks, APHA, 2012
4. Fecal and Total coliforms	Kit Colilert, APHA, 2012.
5. Total ammonia and nitrate (mg/L)	Kit Hach, APHA, 2012.

Table 4. Parameters and methods used for analyzing raw sewage

6. Total Phosphorus (mg/L)	Spectrophotometric of the Acid with Pre-Digestion by Ammonium Persulfate, APHA, 2012.
7. Total solids (mg/L)	Gravimetric, APHA, 2012.
8. Turbidity (UNT)	Turbidimeter, APHA, 2012.

The concentrations found of the analyzed parameters were evaluated according to the discharge patterns in the final effluent described in resolutions 357/2005 and 430/2011 of CONAMA (National Council for the Environment). Finally, samples of TiO2 PC slabs were collected after testing with the flat plate reactor to characterize its surface by the Scanning Electron Microscopy (SEM) method and with Dispersive Energy Spectroscopy (EDS).

2.6 Statistical Analysis

The statistical analysis of the physicochemical results obtained over the four hours of testing was performed using the free Bioestat 5.0 software, using the ANOVA hypothesis test (one criterion), with a decision level $\alpha = 0.05$ (p-value<0.05) to verify whether there was a significant difference between the results.

3 Results and analysis

3.1.1 Phase 1

3.1.2 Compressive and Flexural tensile strength

The results of the mechanical properties are presented and discussed in this section. Firstly, Figure 6 presents compressive strength and flexural tensile strength. The results are presented considering the % of addition of TiO2 used in the PC mixture.

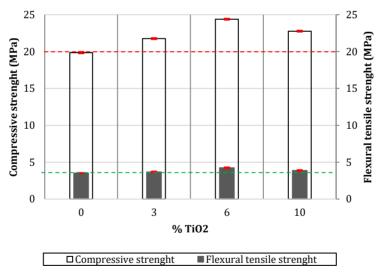


Figure 6. Mechanical properties results *all coefficient of variation were less than 5%

According to Figure 6, the results for compressive strength varied from 19.9 to 24.4 MPa. This range of results agrees with what is found in the literature (Kumaar, 2015), (De Andrade et al., 2015), (Zade, 2016), (Liang et al., 2019), (Xu et al., 2019). All PC mixtures with TiO2 showed higher (red line) Compressive Strength (3% TiO2-9.6%, 6% TiO2-22,8%, and 10% TiO2-14.7%) when compared to the reference PC (0%). Increasing the concentration of TiO2 in the PC mixtures increases the contact area between the cement paste and the coarse aggregate, making the PC more strength.

In the case of Flexural Tensile Strength (green line) the results showed variation from 3.5 to 4.5 MPa, showing the great mechanical potential of the PC with TiO2. Also, practically the same behavior was maintained. All PC mixtures with TiO2 presents higher results (3% TiO2-4.2%, 6% TiO2-20,4%, and 10% TiO2-9.9%) when compared with the PC reference (0%).

3.1.3 Porosity (%) and Permeability (k)

The results of the hydraulic properties: porosity (%) and permeability (k) are presented in (Figure 7). The results presented consider percentage addition of TiO2 to PC mixtures.

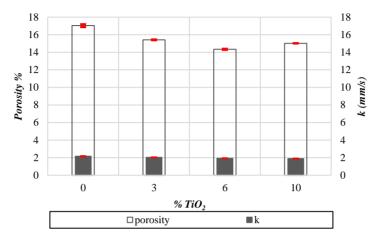


Figure 7. Hydraulic properties results *all coefficient of variation were less than 5%

(Figure 7) show that the porosity (%) varied from 14.3 to 17.0%. This range of porosity agrees with that found in literature (Sandoval et al., 2019), (Sandoval et al., 2019), (Xie, 2019). The PC with TiO2 presented lower porosity values (3% TiO2-9.5%, 6% TiO2-15.8%, and 10% TiO2-11.8%) in all cases when compared to the reference (red line). These results also validate the results of the mechanical properties (Figure 6), where generally PC with higher porosity present lower strength (Sonebi, 2013), (Joshaghani, 2015), (Chandrappa, 2016), whereas the permeability (k) varied from 1.9 to 2.3mm/s. Both hydraulic properties presented the same tendency proportionality between porosity and permeability agreeing with the literature review (Bolt, 2014), (Shen et al., 2020), (Shen et al., 2021).

3.1.4 Summary Phase 1

The mechanical and hydraulic properties were analyzed to study the feasibility of the addition of TiO2 to the PC. (Table 5) shows the average values obtained and were compared to the minimum requirements presented by the American guidelines ACI 522R-10, the Brazilian standard NBR 16416/2015 and VTT-R-080225-13 guidelines.

% of TiO2	Compressive Strength (MPa)	Flexural tensile strength (MPa)	Porosity (%)	k (mm/s)
0	19.86	3.53	17.03	2.16
3	21.76	3.68	15.42	2.04
6	24.39	4.25	14.34	1.93
10	22.77	3.88	15.02	1.89
ACI 522R-10	6.8-37.9	0.68-2.15	10-35	>1.0
NBR 16416/2015	20-35	>1.0	15-30	>1.0
VTT-R-080225-13	10-20	1.5-3.0	20-35	>1.5

Table 5. Summary results Phase	Table	lts Phase 1
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Regarding the PC TiO2 results, the presented results reach the intervals requirements (in mechanical and hydraulic properties) presented by the guidelines, and the standards are shown in (Table 5). Based on these results, the PC with TiO2 can be used for the construction of low traffic pavements and sidewalks, enabling the addition of TiO2 in the improvement of the mechanical properties without damaging the hydraulic performance of the PC.

3.1.5 Phase 2

3.1.6 Photocatalytic treatment

(Figure 8a) shows the means and standard deviation of pH for the influent and effluent sanitary sewage to the photocatalytic treatment. It is observed that after treatment there was a significant increase in this parameter, changing its values from the neutral range (7.44 \pm 0.01) to alkaline (pH >10.6), in all assays.

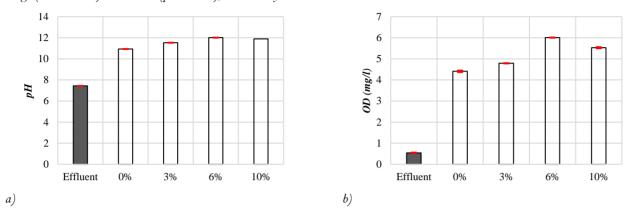


Figure 8. a) pH and b) Dissolved Oxygen concentration *all coefficient of variation were less than 5%

The results shown in Figure 8a can also be associated with the presence of calcium hydroxide, which gives alkaline characteristics to the concrete (Neville, 2013). Zola et al. (2014) reported similar results, where the pH increased from 7.5 to 13. The difference between the pH of the samples may be associated with the difference in porosity. According to (Bolt, 2014), as the time of contact of the effluent with the permeable concrete surface increases, its pH also increases.

As can be seen in (Figure 8b), DO concentration in the effluent also increased, going from 0.54 ± 0.04 mg L-1 in the influent to concentrations above 4 mg L-1 in the effluent (Figure 8b).

(Freudenhammer et al., 1997) found that when flat plate photocatalytic reactors are operated in the recirculation mode, the increased flow improves hydrodynamic parameters and, therefore, increases the concentration of dissolved oxygen and the rate of degradation.

(Figure 9) shows the means and standard deviation of the BOD (9a) and COD (9b) values obtained in the influent and effluent sanitary sewage to the treatment system.

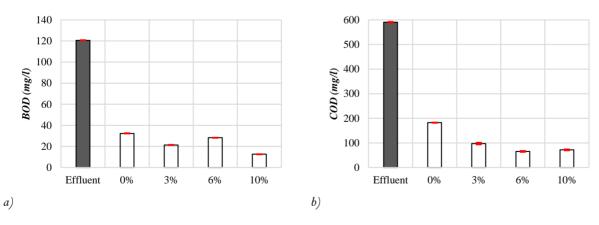


Figure 9. a) BOD and b) COD concentration *all coefficient of variation were less than 5%

The BOD removal efficiency was 73% for P1 and was above 90% in all CP with TiO2 addition (91% - P1.3.TiO2, 93% - P1.6.TiO2, and 96% - P1 .10.TiO2). From the statistical analysis applied to the data, a significant difference was obtained only between P1 and the others, which indicates that the addition of the semiconductor favored the removal of biodegradable organic contaminants.

In relation to COD, the removal percentage was 69% for P1 and 70, 71, and 89% for P1.3.TiO2, P1.6.TiO2, and P1.10, respectively. From ANOVA, applying the removal means, it was obtained that there was no significant difference between the results of

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COD removal between P1, P1.3.TiO2, and P1.6.TiO2; however, when the percentage of TiO2 was increased to 10%, the removal efficiency was significantly higher (p-value < 0.05). This data indicates that the increase in the availability of catalytic sites favored the removal of organic matter considered complex. From the results presented above, it appears that the photocatalytic degradation of organic pollutants presents in the sanitary sewage used in this research involved photolysis and photocatalysis reactions. It is also pointed out that the removal of organic matter, considered biodegradable (estimated by BOD), was not significantly influenced by the addition of TiO2; however, when observing the results obtained for COD, the result was different.

According to Cho & Cho (2002), the macromolecular complexity can reduce the yield of the oxidative step of heterogeneous photocatalysis for a certain time, as the macromolecules can be constituted by several redox centers, which can delay their rapid mineralization.

The use of heterogeneous photocatalysis favors the removal of organic contaminants in effluents, mainly helping in the decomposition of difficult-to-biodegrade organic matter, which would not be easily consumed by microorganisms present in the environment (Padovan, 2015).

When using photocatalysis processes as a treatment technique, some studies indicate that the non-selectivity characteristic of target compounds ends up becoming a problem when working with real effluents such as sanitary sewage (Zhang et al., 2012). (Miranda-García et al., 2011) studied the removal of more than 10 emerging contaminants using heterogeneous photocatalysis, with the use of TiO2, in three different media: electrolytic solution, synthetic sewage and real domestic sewage. Real sewage had the lowest removal efficiencies, which required an increase in the treatment time of about 46%, so that the same efficiency of the tests was obtained using only the electrolytic solution. Nasuhoglu et al. (2012), found similar results when they evaluated the removal of a drug in pure water, synthetic and real sewage. The authors find organic contaminant removal efficiencies of 60%, 41%, and 21%, respectively. According to what was presented in the studies by (Michael et al., 2012) and (Carbonaro et al., 2013) the decomposition of an organic compound is affected by the composition of the matrix and the concentration of organic matter and dissolved salts present in the medium.

According to (Zhang et al., 2015) the presence of ions HPO42-, NH4+, and HCO3- cause negative impacts in the removal of target compounds, due to the strong adsorption of these compounds on the surface of the used photocatalyst (TiO2).

From the discussion presented above, it appears that the use of the permeable concert as a means of support for the immobilization of the photocatalyst (TiO2) may have favored the removal of COD from the sanitary sewage, which allows us to indicate this technique as a feasible and promising model for treatment of this so complex matrix. (Figure 10) shows the removal of total phosphorus over four hours of recirculation of raw sewage.

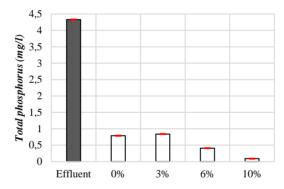


Figure 10. Total Phosphorus concentration results *all coefficient of variation were less than 5%

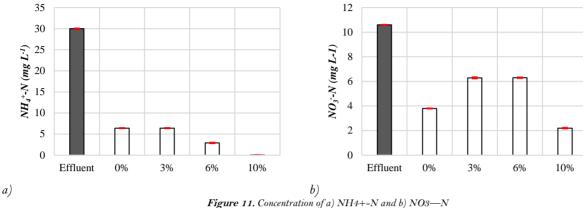
As shown in (Figure 10), only the PC with TiO2 in the concentration of 6% and 10% met the maximum values established by the CONAMA Resolution No. 357/2005 red line (Figure 11). The PC (0%) obtained a reduction of 81.69% of the total phosphorus when compared to the effluent, agreeing with the results found by (Park, 2004), who studied the water purification capacity of PC using coarse aggregate with granulometry ranging from 5 to 10 mm. There was a 96% reduction in total phosphorus. Moreover, (Vázquez-Rivera et al., 2015) investigated the removal of phosphorus in water from a PC mixture, and there was a 90% decrease in phosphorus concentrations. Both results indicate that the PC has the potential to totally remove phosphorus in an efficient way.

For the PC with TiO2, the reduction in concentrations of phosphorus varied from 96.4 to 99.9% compared to the effluent. Such reductions may have occurred due to the direct synergistic interaction between hydroxy radicals OH• in the surface of TiO2 with equilibrium species of PO_4^{3-} , as HPO_4^{2-} , H_2PO_4 , and H_3PO_4 (Arana et al., 2002). Even with the phosphorus showing removal values in the photocatalytic treatments, there may have been adsorption of phosphate ions on TiO2 surface, and not degradation. In a previous study by (Francisco et al., 2009) on the efficiency of a photocatalysis reactor using solar UV in a PET bottle in the final effluent treatment of the sewage treatment plant, they discussed that phosphorus affects the degradation of organic compounds in the wastewater, as the adsorption of phosphate ions can occur on the surface of the TiO2, not degradation.

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3.1.7 Total Ammonia and Nitrate

In the tests performed, a reduction of 78.68%, 78.68%, 82.30%, and 85.84% was observed for samples P1, P1.3. TiO2, P1.6.TiO2, and P1.10.TiO2, respectively, in the total ammonia concentration. (Figure 12) shows the results of total ammonia (Figure 11a) and total nitrate (Figure 11b). On the y-axis, the results of each parameter were plotted, as well as on the x-axis, the effluent, and PC with TiO2 in percentage (%).



*all coefficient of variation were less than 5%

In the tests performed, a reduction of 78.7%, 95.4%, 99.6%, and 99.9% was observed for samples of 0%, 3%, 6%, and 10%, respectively (Figure 11a). The oxidation of NH4+-N can occur through direct oxidation with the hydroxyl radical, which can lead to the formation of several compounds, including nitrogen oxides, nitrite, and nitrate and gaseous nitrogen (Kim et al., 2005), (Kim et al., 2006).

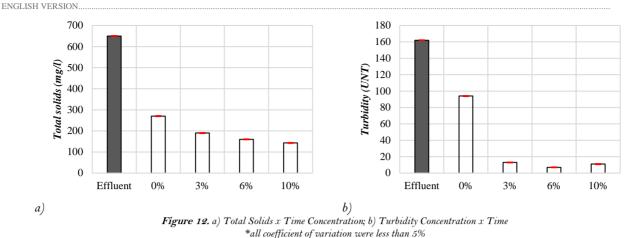
The removal of total ammonia in the test with the PC with TiO2 sample may have occurred due to the elevation of the effluent pH. According to (Ferreira, 2000), free ammonia (NH3) is susceptible to volatilization, while ionized ammonia cannot be removed by volatilization. As the pH rises, the reaction balance shifts to the left, favoring the greater presence of NH3. At pH close to neutrality, practically all the ammonia is in the form of NH4+-N. At a pH close to 9.5, approximately 50% of the ammonia is in the form of NH3 and 50% in the form of NH4 +-N. At pH higher than 11, practically all ammonia is in the form of NH3, contributing to nitrogen removal.

The removal of nitrate in the PC studied decreased in relation to the parameter verified in the effluent, with 10% (Pc with TiO2) showing the best removal results (Figure 11b). (Konstas et al., 2019) evaluating the photocatalytic degradation of organic compounds, found that after 180 minutes of treatment, there was an increase in the concentration of inorganic ions, especially for NO3--N. Phosphate ions (PO4-) were detected at relatively low concentrations, which according to the authors, indicates that they may have been adsorbed onto the surface of the photocatalysts.

3.1.8 Total solids and turbidity

The results for removal of total solids and turbidity are presented in (Figure 12a) and (Figure 12b). To total solids, all PC mixes evaluated, there was a trend in the reduction of total solids concentration compared to the analyzed effluent. The use of 10% TiO2 can improve the removal efficiency in 78%, followed by 6% TiO2 (75%), 3% TiO2 (70%), and 0% (58.5%), respectively. The statistical analysis applied to the data showed that there was significant difference only between the PC without and with addition of the catalyst (p-value < 0.05). Among the PCs with the addition of TiO2, there was no difference between the percentage of solids removal.

These results agree with (Cahill, 2012) who verified a removal of 60% of total solids suspended in a pervious pavement system, and 90% when this same system had a geotextile blanket. The removal of solids present in the medium is important not only for the removal of organic contaminants, such as organic matter, but also for the removal of heavy metals in the medium, as they can adsorb suspended particles in the effluent and be transported long distances in long periods when launched into receiving bodies (Jordão et al., 1999).

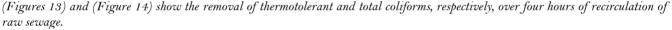


The reduction in the concentration of turbidity presents a trend similar to that of total solids. Then all the PCs produced show a decrease in turbidity compared to the original effluent. The largest reductions were 95% for 6% TiO2, followed by 93% for 10% TiO2, 92% for 3% TiO2, and finally 42% for 0% TiO2 (0%). Similar results were observed by (Pascoal et. al., 2007) and (Koupai et al., 2016), when analyzing the reduction in the concentration of total solids and turbidity in effluents from tanneries with immobilized TiO2 and solar radiation.

In order for heterogeneous catalysis to be efficient, it is necessary to remove suspended solids and, consequently, turbidity, as high turbidity values make it difficult for light to penetrate the medium, due to the scattering effect, reducing the amount of photons that reach TiO2 (Ferreira, 2014). According to (Chong et al., 2010), to ensure the optimization of photocatalytic reactions, it is necessary that the turbidity of the medium is kept below 5 NTU. In this research, the turbidity values were 12.8, 6.75, and 11.00 for 1.2 and 3, respectively, which indicates that the process could have been even more favorable to the degradation of organic matter if the concentration of suspended solids had been further reduced.

As the effluent flows through the permeable concrete, solid particles are restricted and captured on its surface or as it percolates through the voids present in the concrete. The larger the permeable concrete surface area, the more efficient is the adsorption process (Faisal, 2020). The reduction in total solids and turbidity in tests containing TiO2 may have occurred due to its ability to generate hydroxyl radicals (•OH), which are the main responsible for the mineralization of organic matter (Brito et al., 2010).

3.1.1 Thermotolerant coliforms and total coliforms



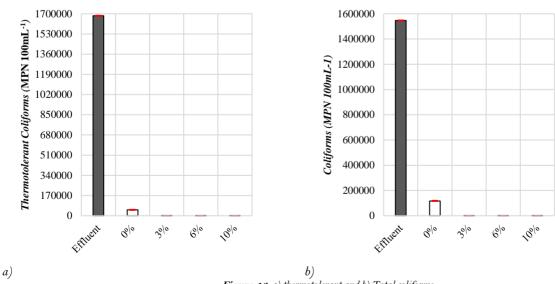


Figure 13. a) thermotolerant and b) Total coliforms *all coefficient of variation were less than 5%

Thermotolerant coliforms showed a reduction of 97.03, 99.99, 99.99, and 99.99% for samples PC (0%), 3% (CP.3.TiO2), 6% (CP.6.TiO2), and 10% (CP.10.TiO2), respectively, showing the great efficiency of both the PC and the addition of TiO2 to the mix. The reduction in total coliforms was 92.44, 99.99, 99.99, and 99.99% for samples CP, CP.3.TiO2, CP.6.TiO2, and CP.10.TiO2,

respectively. From the statistical analysis applied to the data, it was possible to verify that there was a significant difference only between the results without and with the addition of TiO2. Among PCs with different concentrations of TiO2, the results obtained were statistically equal (p-value > 0.05).

(Cho et al., 2004) evaluated the relationship between E. coli inactivation and TiO2 concentration. The authors found a significant dependence of the inactivation efficiency of E. coli with the photocatalyst concentration, showing that the increase in the TiO2 concentration generates more reactive species responsible for the inactivation of microorganisms, but not in a linear fashion. Although higher concentrations of TiO2 provide more surface area available for reaction, they also decrease the depth of light penetration into the suspension by increasing radiation scattering, which reduces the efficiency of photocatalytic inactivation of E. coli (Bekbölet, 1997).

The increased disinfection of domestic sewage in samples containing TiO2, may be associated with the presence in the medium of two main photochemical oxidants: •OH and the oxidative oxygen species (Cho et al., 2004), (Ganguly et al., 2018).

These oxidizing agents can exert oxidative stress on microbial organisms, as they can attack the peptidoglycan layer of the cell wall, leading to phospholipid peroxidation and protein membrane oxidation, which can cause cell wall disruption, leading to the release of the intracellular materials. In addition to this attack, when oxidative oxygen species enter the cell, they can damage its functions, altering or inhibiting the sequencing of proteins for DNA replication or even attacking the coenzymes that inhibit the respiratory functions of microorganisms (Cho et al., 2004), (Saito et al., 1992), (Maness et al., 1999), (John, 2021).

According to studies carried out by (Cho et al., 2004), the $\bullet OH$ is approximately one thousand to ten thousand times more effective for inactivating E. coli than other chemical disinfectants, such as chlorine, ozone, and chlorine dioxide, thus making the treatment an even more attractive study for sanitary sewage disinfection, and this explains the results obtained in this research.

The increases in the removal of fecal and total coliforms are related to increases in pH, where it is clear that in the PC TiO2 with very high pH values (Figure 13a), the greatest magnitude of reduction of this parameter was presented. These results agree with the literature review where the alkaline treatment of biosolids (solid by-products of sewage treatment, which has characteristics that allow its agricultural use) has been commonly used to reduce and inactivate pathogens and microorganisms. For example, Salmonella, fecal coliforms, Escherichia coli, and fecal streptococci were completely inactivated in the pig manure mixture, in an 8-day stabilization (Wong, 2009).

For Von (Sperling, 2005), the removal efficiency in stabilization ponds should be in the range of 90% to 99.9%, for fecal and total coliforms; therefore, all samples used in this research are in this range, thus being able to be used in the treatment of sanitary sewage.

3.1.2 SEM and EDS

Samples of the TiO2 PC slabs were collected before and shortly after the four hours of recirculation of the raw sewage and analyzed with the SEM and EDS. The formation of biofilm on the PC surface was detected, demonstrating that the organic matter was retained on the PC surface (Figures 14a) and (Figure 14b). The analysis of the sample surface by EDS (Figures 15a) and (Figure 15b) before and after the sewage recirculation in the PC indicates the formation of higher peaks of carbon and oxygen, providing an ideal environment for biofilm formation (Nuvolari, 2011).

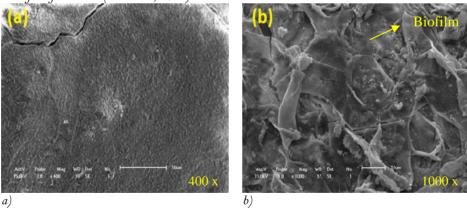


Figure 14. a) PC before recirculation of the wastewater; (b) PC after sewage recirculation.

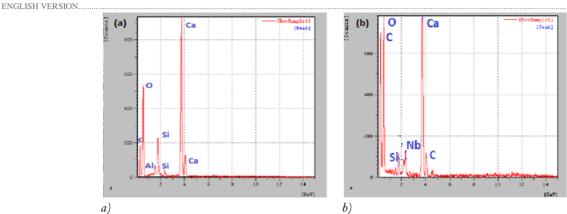


Figure 15. EDS analysis of the PC with 10% TiO2 after testing: (a) before the sewage recirculation; (b) after the sewage recirculation

Organic compounds, nutrients, and oxygen present in the wastewater penetrate the biofilm layers by diffusion. Depending on the thickness of the biofilm formed in the support medium, aerobic and anaerobic zones can occur. The support medium can be of the most varied materials, among them: gravel, blast furnace slag, wood chips, sand, and corrugated tubes (Sperling, 2013).

According to (Wolff et al., 2010), each support material has different shape, surface area, porosity, roughness, which influences the colonization of microorganisms and the formation of biofilm. With that, the organic matter adhered to the pervious concrete surface can be attributed to its physical characteristics, highlighting its great porosity and high surface area.

4 Conclusions

This research studied the addition of titanium dioxide (TiO_2) to PC to evaluate its efficiency in the sewage treatment. PC slabs were produced from a standard mixture with different concentrations of TiO_2 . Based on the results presented, it is concluded that:

- PC slabs containing TiO2 were more efficient as a method that can assist in the treatment of sanitary sewage when compared to slabs without the addition of TiO2;
- The flat plate reactor developed in this research was suitable for use in tests with PC slabs;
- It was found a greater efficiency using the samples with 10% of TiO2 in the removal of total phosphorus, total ammonia, total solids, and turbidity, reaching a removal of 95.56, 85.84, 77.29, and 95.98%, respectively;
- In relation to nitrate, PC reached a removal of 64.19%, whereas in samples containing 3% of TiO2, this percentage reached 65.91%. In the tests carried out for total ammonia and nitrate in the presence of TiO2 with a concentration greater than 3%, a tendency towards degradation of total ammonia and an increase in the concentration of nitrate was observed; however, this increase was below that allowed by current legislation;
- Using the ANOVA and Tukey statistical test, the addition of TiO2 significantly influenced, with a 95% confidence level, the parameters analyzed in the raw sewage. Only the total and fecal coliform assays did not show a significant difference between the means.
- It was found a greater efficiency using samples with 10% of TiO2 in the removal of total coliforms and fecal coliforms, reaching a removal of 99.99% and 99.99%, respectively;
- After four hours of recirculation of the raw sewage, the samples from all the tests performed showed alkaline pH values ranging from 10.63 to 12.02. There was no inhibition or deactivation of the photocatalytic process due to the high pH. Degradations occurred regardless of the pH values;
- The increase in the concentration of dissolved oxygen in all tests occurred due to the flat plate reactor used in the research of the open type, where contact with the atmosphere increases the concentration of dissolved oxygen in the effluent;
- The permeable concrete slabs removed the BOD and COD concentration rates by 73.15% and 69.07%, respectively. With the use of 10% of TiO2 in the plates, the mentioned removal rates increased by 96.43% for BOD and 89.48% for COD;
- Thus, PC with the addition of TiO2 was efficient for use in sewage treatment. Its use associated with 10% of TiO2 can add better efficiency to the sewage treatment process.
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