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Assessment of a Photoreactor with Immobilized Nanoparticle TiO_2 Films for the Purification of Rainwater

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ABSTRACT

The heterogeneous photocatalysis consists of the generation of reactive oxygen species ($^{\bullet}OH$, $^{\bullet-}O_2$) from a catalyst, UV light, and oxygen; these reactive species can degrade contaminants and eliminate microorganisms. The purpose of this research was to evaluate a heterogeneous photocatalysis system and an UV light disinfection system for the elimination of total coliforms and *Escherichia coli* bacteria present in rainwater stored in five cisterns in Mexico City. The elimination of total coliforms (MPN/100 mL) and *Escherichia coli* (CFU/100 mL) were evaluated both in the rainwater treated with TiO_2/UV and UV (in time periods of 30 and 60 minutes), according to the treatments established in the statistical model 2^2. The results show that although complete elimination of initial total coliforms (9.3×10^4 MPN/100 mL) and *E. coli* bacterium (1.5×10^3 CFU/mL) was achieved in one of the samples of rainwater using only UV light at 254 nm for 30 minutes, the use of 8 films coated with Degussa P-25 titanium dioxide, UV light at 254 nm and 1.5 vvm air in a reactor, achieves a total pathogen removal in a shorter time of 15 minutes. Thus, we anticipate that the combined treatment could be an alternative disinfection process for rainwater stored in cisterns, reducing costs and making the treatment viable for a larger-scale application.

1. INTRODUCTION

The increase in population density and urban growth has caused problems of availability and supply of water resources in developing countries [1]. In Mexico, the water availability per person has decreased due to the intensification of agricultural and industrial activities [2]. Thus, it is crucial to evaluate various treatment methods of providing improved drinking water [3]. In this context, interest in the collection and use of rainwater has grown [1] since it does not require a complex or expensive treatment for its purification.

Although rainwater is a good alternative for solving water scarcity, one of its main problems is that it has presented biological indicators such as *Escherichia coli* and total coliforms in storage cisterns in countries such as Korea, Nigeria, New Zealand, and Australia [4]. Studies carried out in Australia reported the presence of pathogenic microorganisms such as *Escherichia coli*, total and faecal coliforms, *Campylobacter, Salmonella, Legionella*, *Pseudomonas, Cryptosporidium, Enterococci, Giardia, Aeromonas*, and *Mycobacterium avium* in rainwater collected and stored in cisterns [5]. In the Adelaide region (Australia), Chubaka et al. [6] analyzed the microbiological quality of 53 samples of rainwater stored in tanks and found 10 tanks that contained *Escherichia coli* in concentrations that exceeded the limit of 150 CFU/100 mL for recreational water quality. The presence of these pathogens is often caused by the carry-over of solids, organic matter, and fecal deposits of animals in rainwater harvesting systems and is influenced by environmental conditions [7, 8]. To meet drinking water quality standards, various technologies have been developed to remove contaminants in rainwater. Among them, the technologies most used to treat rainwater are chlorination and ultrafiltration. However, ultrafiltration generally requires constant replacement of filters, and chlorination can generate trihalomethanes and by-products potentially harmful to human health [8, 9]. Currently, there are other efficient disinfection technologies such as ultraviolet light (UV) and heterogeneous photocatalysis with titanium dioxide (TiO_2). UV light water treatment is a chemical disinfection technology that is based on the removal of nucleic acids of the cell from bacteria and protozoan cysts through exposure to UV rays [10, 11]. Due to its good pathogen removal indicators, this technology has been widely installed in water treatment plants in North America and Europe [12]. On the other hand, heterogeneous photocatalysis consists of the excitation on the surface of a catalyst (semiconductor) with irradiation of light energy at least equal to that of the bandgap [13, 14]. This series of reactions leads to the formation of hydroxyl radicals (OH^-), which have an oxidizing effect on organic pollutants and microorganisms in the water. The elimination of pathogens happens by lipid peroxidation reactions on the surface of the particle that is used as a semiconductor, which destroys the double lipid layer of the outer wall of the microbial cell [15, 16]. Numerous studies have shown that these technologies are efficient when operating individually. Nevertheless, to guarantee the quality of the collection and storage of rainwater, a detailed analysis of the combination of these technologies must be carried out [17]. Therefore, it is important to compare the common components of these systems to seek better efficiency and profitability of rainwater treatment.

This research establishes a comparison between a heterogeneous photocatalysis system with TiO_2 and a UV light disinfection system for the elimination of pathogens, such as total coliforms and *E. coli* present in rainwater stored in 5 cisterns. This paper is organized as follows: the first section shows the physicochemical and microbiological characterization of the rainwater stored in cisterns in order to select the most contaminated water to carry out the treatment with the two proposed systems. Section 2 describes the operating conditions of the reactor with TiO_2 and UV as well as the establishment of the treatments. In section 3, the inactivation of total coliforms and *Escherichia coli* in the proposed treatments, as well as the kinetics of pathogen removal are evaluated. The purpose of this research is to evaluate the efficiency of these treatment systems individually and in combination in order to provide an efficient and economical alternative for the use of rainwater in Mexico City.

2. PHYSICOCHEMICAL AND MICROBIOLOGICAL CHARACTERIZATION

This study was carried out during the wet season in the Center for Research and Advanced Studies of the National Polytechnic Institute (CINVESTAV-IPN), located in Mexico City, Mexico (Figure 1). 1Lt samples of 5 rainwater storage tanks (cisterns) with 100 m^3 of capacity were collected (Figure 2). Then, the filtration of the rainwater samples was carried out, using a 0.5 mm and 7 cm mesh and diameter filter, to remove the coarse solids presents in the water stored in the cisterns and finally these were refrigerated at 4°C for 2 hours to perform the water characterization.

The physicochemical and microbiological characterization of the rainwater samples from the 5 storage cisterns was carried out, according to the Mexican official standard for the human consumption of water [18]. Metals in water were determined in ICP-MS NexION equipment, and the quantification of total coliforms was carried out with the Most Probable Number Method (MPN), established in the Official Mexican Standard Method [19]. Furthermore, the Colony Forming Unit method (CFU) was used to identify the amount of *Escherichia coli* bacterium in rainwater samples through Eosin Methylene Blue Agar plates.

According to the results obtained in the characterization of water (Table 1), it was demonstrated that most of the physicochemical parameters of water quality did not exceed the limits established in the official Mexican standard of water quality [12]. However, some parameters, such as the pH in the cistern 3, the total hardness as $CaCO_3$ in cisterns 1, 2, and 5, and the iron concentration (Fe) in cisterns 1, 2, 4, and 5, exceeded the limits established in the Mexican standard of water quality. On the other hand, total coliforms were present in all cisterns, among them cistern 3 presented fecal coliforms and *E. coli* in a higher concentration in comparison to the others. Therefore, the rainwater sample from cistern 3 was selected to carry out the treatments with TiO_2 and UV light in a photoreactor.

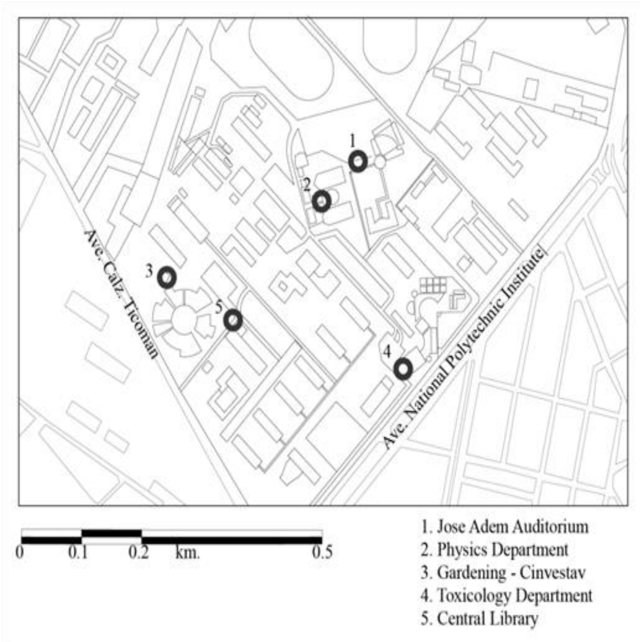

Figure 1. Location map of rainwater stored cisterns 19°30′33″N 99°07′46″O

Table 1. Physicochemical and microbiological characterization of the rainwater stored in the 5 cisterns

Physicochemical parameters (mg/L)	Cistern 1	Cistern 2	Cistern 3	Cistern 4	Cistern 5	MPL
Aluminum	0.004	0.017	0.102	0.015	0	0.2
Arsenic	0.015	0.004	0.001	0.002	0.008	0.025
Barium	0.044	0.108	0.127	0.107	0.162	0.7
Cadmium	0	0	0	0	0	0.005
Copper	0.003	0.001	0.005	0.004	0.002	2
Total chrome	0.005	0.004	0.003	0.003	0.008	0.05
Total hardness	**3124.8**	**3013.2**	334.8	409.2	**2641.2**	500
Iron	**0.604**	**0.747**	0.236	**0.449**	**0.749**	0.3
Manganese	0.001	**0.45**	0.036	0.015	0.001	0.15
Mercury	0	0	0	0	0	0.001
pH	7.21	6.57	6.42	7.09	7.65	6.5-8.5
Lead	0	0	0	0	0	0.01
Sodium	6.499	-	13.494	5.699	-	200
Zinc	0.047	0.035	0.06	0.098	0.05	5
Microbiological parameters	**Cistern 1**	**Cistern 2**	**Cistern 3**	**Cistern 4**	**Cistern 5**	**MPL**
Total coliforms (MPN/100 mL)	75000	93000	93000	46000	93000	absence
Fecal coliforms (MPN/100 mL)	ND	ND	40	1500	ND	absence
Escherichia coli (CFU/mL)	ND	ND	15000	2300	ND	absence

ND: Values are below the values expressed in the LDM cell, MPL: Maximum Permissible Limit, according to the source [19].

Figure 2. Sample taking of rainwater in the 5 cisterns

3. PHOTOREACTOR WITH TiO_2/UV AND OPERATING CONDITIONS

The treatments in the photocatalysis system were carried out using a pyrex glass photoreactor with a 1 L of total volume, operated at 80% of its capacity (0.8 L). Inside, this system had a porous silicone hose diffuser for 1.5 vvm constant air flow; a 60W UV lamp with a wavelength of 254 nm installed inside of the quartz tube. Throughout the internal contour of the reactor 8 glass plates coated with TiO_2 nanoparticles were placed, these were supported in a teflon rack (Figure 3). The area of the glass support for TiO_2 films was 45 cm^2 (63.2 mg/cm^2 of TiO_2) and disposed parallel to the walls of the vessel at a distance of 7 cm from the lamp [20].

The treatments with the UV disinfection system were carried out in the same photoreactor but without the 8 glass plates coated with TiO_2 nanoparticles (Figure 4).

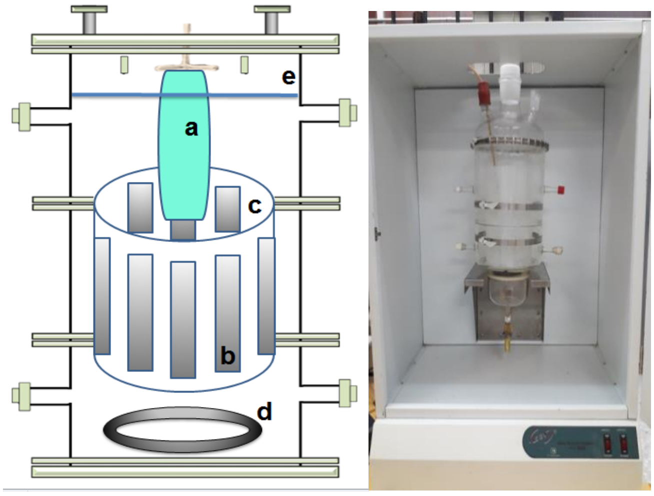

Figure 3. Photoreactor: (a) UV lamp, (b) Nanoparticle TiO_2 films, (c) Teflon rack, (d) Diffuser, (e) Water level

Figure 4. UV disinfection system

Statistical model 2^2 was used to evaluate the interaction between factors: TiO_2 (factor X_1) and residence time (factor X_2) into the photoreactor, which shows a response in the elimination of total coliforms (MPN/100 mL) and *Escherichia coli* (CFU/100 mL) in rainwater. This model gives us 4 treatments, which are presented in Table 2. In Addition, Table 3 shows the controls for the experiment.

Table 2. Experimental design 2^2 of the treatments

Treatments	Factor X_1 TiO_2 catalyst	Factor X_2 Residence time (minutes)
T1	+1 (TiO_2/UV)	-1 (30)
T2	-1 (UV)	-1 (30)
T3	-1 (UV)	+1 (60)
T4	+1 (TiO_2/UV)	+1 (60)

Table 3. Controls evaluated in the treatments

Controls	Factor X_1	Residence time (minutes)
C1	1.5 vvm air	-1 (30)
C2	1.5 vvm air	+1 (60)
C3	UV (254nm)	-1 (30)
C4	UV (254 nm)	+1 (60)
C5	X films of TiO_2	-1 (30)
C6	X films of TiO_2	+1 (60)

4. INACTIVATION OF TOTAL COLIFORMS AND BACTERIUM *ESCHERICHIA COLI*

In Figure 5, it is shown that the 4 treatments eliminated 100% of the fecal coliforms and the *Escherichia coli* bacteria in the rainwater samples, without presenting significant differences ($p < 0.05$) between them. Controls 1 and 2 did not show a significant reduction of total coliforms and *E. coli*, as they were only provided with air (without TiO_2 and UV light). The same happened with controls 5 and 6, in which only the TiO_2 films were used, which were not excited due to the lack of UV radiation; therefore, they did not present pathogen removal.

Figure 5 shows that T1 and T2 were the most efficient treatments since they managed to eliminate pathogens in 30 minutes. The results obtained are consistent with those reported by Pantoja-Espinoza et al. [21], who reached complete elimination of total coliforms and *E. coli* in a maximum of 20 to 35 minutes in a photocatalysis system with

UV-C lamp and photocatalyst UBE (TiO_2 / SiO_2).

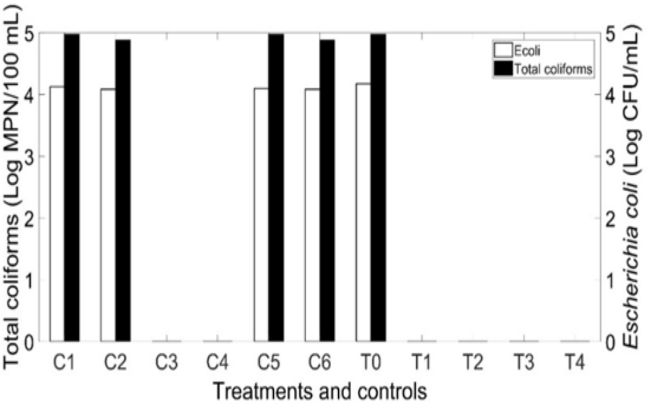

Figure 5. Reduction of total coliforms and *E. coli* in the 4 treatments and 6 controls

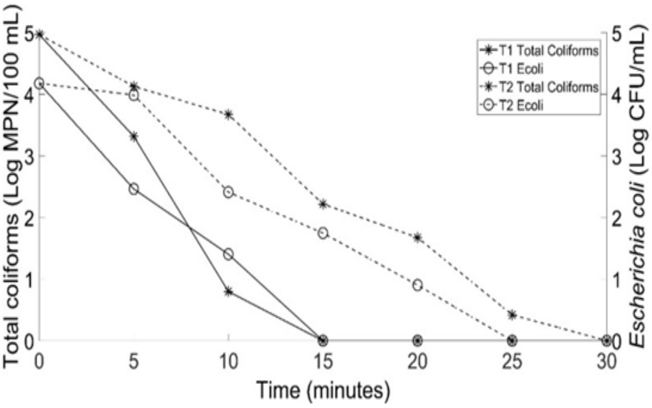

Figure 6. Reduction of total coliforms and *Escherichia coli* by T1 and T2 as a function of time

In addition, Rojas-Higuera et al. [22] achieved complete inactivation of *Escherichia coli* in samples of domestic wastewater after 30 minutes in a heterogeneous photocatalysis system with TiO_2 and UV light. Although, it is important to mention that T1 and T2 achieved the fastest pathogen inactivation, this experimental design did not determine the specific time in which they achieved this. For this reason, the kinetics evaluation of both treatments was carried out (Figure 6).

Figure 6 shows that the T1 (TiO_2/UV) achieves elimination of the pathogens in a shorter period of time than the T2 (UV). The T1 reaches the complete elimination of *Escherichia coli* and total coliforms in 15 minutes while the T2 reaches the elimination of the *E. Coli* bacteria in 25 minutes and the total coliforms in a time of 30 minutes, which indicates that photocatalysis treatment with TiO_2 and UV light could be a more efficient solution in terms of energy costs and time to disinfect rainwater. However, it is important to consider that both treatments benefited from the good physicochemical quality of the rainwater, which improved the efficiency of the photoreactor by not presenting inhibitors (organic matter and solids) of the photocatalytic reaction.

5. CONCLUSIONS

Rainwater collected in cisterns can play an important role in the supply of water resources in Mexico City if there is adequate and economic treatment. Our research demonstrated that considering the proposed experimental design, the best treatment for the removal of total coliforms and *E. coli* in rainwater was T1 (8 films coated with Degussa P-25 Titanium dioxide and UV light at 254 nm), reaching a completed removal of pathogens after 15 minutes. Although T2 (UV light at 254 nm) achieved complete elimination of pathogens in a longer treatment time (30 minutes), it could represent a high energy cost, mainly at pilot scale. However, the TiO_2 treatment worked efficiently due to the good physicochemical quality of the rainwater. For this reason, it is recommended to carry out a constant cleaning of the rainwater storage cisterns as well as to carry out periodic analysis of the water collected before treatment. Besides, to implement this system on a large scale, it is suggested that future studies evaluate the half-life time of TiO_2 in the reactor, the concentration of the TiO_2 nanoparticles in the treated water, as well as the investment and operating costs.

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