water

MDPI

Article

Towards a Novel Combined Treatment Approach Using Light-Emitting Diodes and Photocatalytic Ceramic Membranes

Jorge Bernardo [1,2], João Sério [1], Beatriz Oliveira [1,3], Ana Paula Marques [1], Rosa Huertas [1,2], João Goulão Crespo [2] and Vanessa Jorge Pereira [1,3,*]

[1] Instituto de Biologia Experimental e Tecnológica (iBET), Apartado 12, 2781-901 Oeiras, Portugal; j.andrebernardo@gmail.com (J.B.); joao.serio@ibet.pt (J.S.); beatrizreisoliveira@hotmail.com (B.O.); ana.marques@ibet.pt (A.P.M.); rosa.huertas@ibet.pt (R.H.)

[2] LAQV-REQUIMTE, Department of Chemistry, NOVA School of Science and Technology, FCT-Universidade NOVA de Lisboa, 2829-516 Almada, Portugal; jgc@fct.unl.pt

[3] Instituto de Tecnologia Química e Biológica António Xavier, Universidade NOVA de Lisboa, Av. da República, 2780-157 Oeiras, Portugal

* Correspondence: vanessap@ibet.pt

Abstract: Natural disasters (such as earthquakes, floods, heatwaves and landslides), isolation and war affect the water access of millions of people worldwide. Developments in the areas of membrane filtration, photolysis and photocatalysis are important for safe water production and water re-use applications. This work aimed to test alternative ways to ensure effective disinfection of wastewater effluents: light-emitting diodes that emit at different wavelengths, photocatalytic membranes, and the combination of the two solutions. The different treatment processes were tested at the laboratory scale to assess their performance in the removal and inactivation of water quality indicator bacteria and fungi present in wastewater effluents. The membranes were found to be effective to retain the microorganisms (rejection values higher than 96%), while three small ultraviolet C light-emitting diodes that emitted light at 255 and 265 nm showed an excellent performance for inactivation (higher than 2.5-log inactivation of total coliforms and *Escherichia coli* after 10 min of exposure in real wastewater effluents). When photocatalytic membranes are used, ultraviolet A light-emitting diodes ensured effective treatment of the retentate (higher than 65%). The combination of these two processes is extremely promising since it ensures not only the production of a high quality permeate that can be reused, but also the treatment of the retentate.

Keywords: indicator bacteria and fungi; wastewater effluent; light-emitting diodes; ceramic membranes; photocatalytic membranes; combined treatment; retention and disinfection

Citation: Bernardo, J.; Sério, J.; Oliveira, B.; Marques, A.P.; Huertas, R.; Crespo, J.G.; Pereira, V.J. Towards a Novel Combined Treatment Approach Using Light-Emitting Diodes and Photocatalytic Ceramic Membranes. *Water* **2022**, *14*, 292. https://doi.org/10.3390/w14030292

Academic Editors: José Antonio Mendoza-Roca and Jesus Gonzalez-Lopez

Received: 4 November 2021
Accepted: 12 January 2022
Published: 19 January 2022

1. Introduction

Research developments in the areas of disinfection and filtration have been considered essential to ensure water purification in the coming decades [1].

Ultraviolet (UV) radiation and advanced oxidation processes are known to be effective for the inactivation of a broad range of microorganisms and degradation of chemical compounds e.g., [2–6]. Light-emitting diodes (LEDs) are promising alternatives to the conventionally used low and medium pressure mercury lamps due to their long lifetime, lower cost, lower energy consumption, low maintenance, mercury free composition and the wide range of wavelengths available to optimize treatment [7]. In the last few years, several studies have suggested the use of UV LEDs as an alternative solution for water disinfection.

Jarvis et al. [8] showed that a full-scale UV LED reactor was at least as effective as a mercury UV reactor for the inactivation of *Cryptosporidium* surrogates. Beck et al. [9] tested flow-through UV reactors containing either a low-pressure mercury lamp or UV LEDs emitting at 276 nm to disinfect bacteria, viruses, and protozoan pathogens that remained in the domestic wastewater and septic tank permeate after filtration. The results demonstrated

that the UV LED reactor achieved up to 3.5-log reduction of MS2 virus in wastewater at a low flow rate of 0.01 L/min, and for the UV LED inactivation of septic tank effluent, up to 1.5-log reduction of MS2 was attained. In a study conducted by Song et al. [10], the inactivation of *Escherichia coli* bacterium and MS2 virus in synthetic laboratory waters as well as *E. coli* and total coliforms in real wastewater was investigated by continuous and pulsed irradiation using UV LEDs. The results showed comparable inactivation for all microorganisms examined by continuous and pulsed UV LED irradiation at 265 nm under equivalent UV fluence values.

The efficiency of UV-C LEDs that emit light at 255 and 265 nm was recently studied and compared in terms of the inactivation of filamentous fungi spiked in surface water matrices [11,12]. These studies confirmed that the LEDs that emit at 265 nm (closer to the maximum peak absorption of DNA) performed better in terms of inactivation and addressed the effects of the different wavelengths on the morphology of the spores, cellular membrane permeability, enzymatic activity, dimmer formation, proteome response and reactivation potential of the tested fungi (*Aspergillus fumigatus*, *Aspergillus niger* and *Aspergillus terreus*).

To ensure effective disinfection, a combined treatment approach can be considered using hybrid systems, where membrane filtration retains the microorganisms as well as chemical compounds, whereas UV radiation inactivates and degrades the pollutants retained by the membrane.

If proven effective, this hybrid treatment process may help minimize two problems usually associated with membrane filtration processes: the production of a highly concentrated retentate and fouling.

Ceramic membranes have been widely proposed for water and wastewater treatment due to their filtration performance and long-term stability because of their chemical, mechanical and thermal resistance [13]. Ceramic membranes are therefore good candidates to test in hybrid treatment processes that combine membrane filtration with photolysis or photocatalysis since the membrane surface that retains the pollutants needs to be resistant to the UV light and to the reactive oxygen species produced.

Silicon carbide membranes have been proposed due to their high water permeability, fouling resistance, good performance under harsh conditions and wide range of applications [14]. As an example, these membranes were tested for the treatment of a secondary wastewater effluent and proved to be effective for the removal of suspended solids (99%), colloidal particles (96%) and chemical oxygen demand (83%) [15].

In hybrid treatment processes, membranes can be used unmodified or further modified to gain photocatalytic properties that, in combination with UV light, through the production reactive oxygen species such as hydroxyl radicals, may help overcome membrane fouling and improve filtration efficiency [13].

Among the heterogeneous catalysts tested, TiO_2 nanoparticles revealed to be the most promising materials [16]. To be catalytically active, TiO_2 requires irradiation with a source in a wavelength range lower than 390 nm. Even though the sol-gel method has been widely proposed for the synthesis of TiO_2-based materials, it usually involves the use of organic solvents which are hazardous. The use of photocatalytic membranes modified without using organic solvents presents several economic and environmental advantages. In a recent study, photocatalytic surfaces prepared at a low temperature and under solvent-free conditions exhibited a narrow pore size distribution and homogeneity without cracks [17]. These surfaces were characterized in terms of pore size and degradation of methylene blue [17]. Given their porous structure, the microfiltration photocatalytic membranes produced using solvent-free conditions are expected to effectively retain microorganisms such as bacteria and fungi that could then be inactivated by photocatalysis. The retention of microorganisms (present at occurrence levels in real water matrices) using ceramic membranes modified using a solvent-free procedure combined with LEDs that emit light at different wavelengths to ensure inactivation by photocatalysis has not been previously described.

A hybrid reactor that allows combined treatment using membranes and photocatalysis [18] was recently assembled and tested for the combined treatment of wastewaters, achieving removals of total organic carbon, chemical oxygen demand and phenolic compounds of approximately 90% in the first 20 min of treatment [18]. Even though the submerged photocatalytic membrane reactor and the modified membranes represent a step forward towards the development of new advanced treatment technologies able to cope with several water and wastewater contaminants, it implies the use of large membranes and large photolysis systems (either UV mercury lamps or LED panels) that can irradiate the surface of the membranes. Before this hybrid system is used, which will imply the modification of large membrane areas and the development of LED panels, the effectiveness of unmodified and modified ceramic membranes to retain microorganisms present in the water at occurrence levels need to be confirmed, the inactivation with LED systems that emit at different wavelengths needs to be tested and the combined treatment needs to be evaluated.

The objectives of this work were therefore to evaluate the efficiency of LEDs that emit light with different wavelengths at the laboratory scale and test the effect of filtration and the two processes combined. Photocatalytic ceramic membranes previously modified, using a solvent-free procedure, and characterized [17] were also used to evaluate their combined retention and disinfection potential. The processes were tested in terms of their ability to retain and inactivate water quality indicator bacteria (total coliforms and *E. coli*) and fungi present at occurrence levels in real wastewater effluents.

2. Materials and Methods

2.1. Matrix

Several samples of treated wastewater were collected in sterile bottles at a wastewater treatment plant. The wastewater effluent matrix used was characterized in several sampling events conducted over the course of a year in a previous study [19]: pH = 7.22 ± 0.15; total suspended solids = 34 ± 15 mg/L; chemical oxygen demand = 97 ± 29 mg/L O_2. The samples were stored at 4 °C and analyzed in the same day of collection. This is an important issue since previous tests showed that the concentration of microorganisms decreased over time, even when stored at 4 °C. Therefore, every test was conducted with fresh samples, and was collected and analyzed in the same day.

2.2. Experimental Setup

The laboratory systems used are depicted in the Supplementary Material section (Figure S1).

In this work, several experiments were performed with the aim to:

- Test LEDs with different wavelengths without the use of photocatalytic surfaces (Figure S1a);
- Test the efficiency of filtration without light (Figure S1b) comparing non-modified silicon carbide membranes and photocatalytic membranes modified with silicon dioxide and titanium dioxide [17];
- Test both solutions, LEDs with different wavelengths combined with non-modified and modified membranes (Figure S1c).

2.2.1. Light-Emitting Diodes

The inactivation experiments were performed in a Class II biological safety cabinet. The LED reactor used in this study was a PearlLab Beam reactor (AquiSenseTechnologies, Charlotte, NC, USA) that contains a triple wavelength UVinaire™, a control box, a UV homogenizing (collimating) tube, a wavelength selector, and an AC-DC adapter (12 V 90 W). The selected wavelengths were: 255 and 265 nm for inactivation and 365 nm to activate photocatalytic surfaces (Figure S1a). The average intensity of each LED wavelength was measured using a radiometer (ILT 950 UV Spectroradiometer, International Light Tecnologies, Inc., Peabody, MA, USA). These measurements were performed at a

distance of 4 cm (the same distance used in the experiments) and the results obtained were 55.66 μW/cm^2, 250.30 μW/cm^2 and 5284.89 μW/cm^2 for 255 nm, 265 nm and 365 nm wavelengths, respectively. The average irradiance values in the wastewater were determined after considering different correction factors (reflection factor, Petri factor, water factor, and divergence factor) [20]. The following correction factors were used: 0.98 for the reflection factor, 0.90 for the Petri factor and 0.64 for the divergence factor. The water factor, which considers the decrease in irradiance due to sample absorbance at the wavelengths of interest, was determined for the wastewater.

In a refrigerated and stirred vessel, 50 mL of sample was subject to the different LED wavelengths for 1, 5 and 10 min of exposure. The samples were then analyzed in terms of total coliforms, *E. coli* and fungi.

Furthermore, to verify the stability of the selected microorganisms without being subject to any light sources, a control sample was placed in a dark environment during the elapsed experimental time and analyzed at the beginning and at the end of the experiment.

2.2.2. Membrane Modification

Silicon carbide membranes (from LiqTech with a diameter of 4.5 cm) provided support for the deposition of titanium dioxide films with photocatalytic properties. To produce the photocatalytic films, the sol-gel technique was used without the addition of solvents and with the addition of tetraethyl orthosilicate (TEOS 98 %, Merck KGaA, Darmstadt, Germany) and Degussa P25 titanium dioxide particles (30–90 nm diameter, Evonik Industries, Essen, Germany). The detailed modification procedure followed was previously described by Huertas et al. [17]. Huertas et al. [17] proposed a solvent-free process for the development of photocatalytic microfiltration membranes. In the mentioned study [17], the membranes were only tested in terms of their photocatalytic performance using methylene blue, but their estimated porous characteristics showed that a high retention of microorganisms such as bacteria and fungi could be expected. The only difference in the modification was the deposition method (dip coating was used in this study while drop casting was used in the previous study [17]). Dip coating (previously described by Fraga et al. [18]) was the selected method because, if the results of this paper are promising, larger membranes will need to be modified using this technique. The characterization of the membranes used in this study is provided in the Supplementary Material section.

2.2.3. Filtration Experiments with and without LED Inactivation

These experiments were performed in a dead-end filtration unit (Nalgene®, Rochester, NY, USA) connected to a vacuum pump (Cole-Parmer, Vernon Hills, IL, USA). The entire system was sterilized to test the efficiency of the retention of microorganisms in the non-modified and modified membranes (Figures S1 and S2). Feed, permeate and retentate samples were analyzed. Due to the much higher levels of flux in the non-modified membranes (around 37 mL/min compared to 0.2 mL/min for the modified membranes), the experiments varied from 10 min for the non-modified membranes and 80 min for the modified membranes. The effectiveness of filtration was also tested in combination with each of the three LED wavelengths.

The system was placed and operated inside a black box for security reasons.

2.3. Methods for Detection of the Target Microorganisms

2.3.1. Bacterial Analysis

The concentration of total coliforms and *E. coli* in the samples was analyzed using the Colilert-18/Quanti-Tray® (IDEXX, Westbrook, ME, USA) method (ISO 9308-2: 2012) as previously described [21,22]. The protocol consisted of adding the kit substrate to 100 mL of the sample in sterile bottles (direct sample and decimal dilutions from 1:10 to 1:100,000) and then shaking until completely mixed. The mixture was then placed inside a Quanti-Tray® (IDEXX, Westbrook, ME, USA), sealed, and incubated at 35 ± 0.5 °C for 18–22 h. The detection of total coliforms indicated the presence of the enzyme β-galactosidase which

metabolizes one of the substrate nutrients and changes its color to yellow. Regarding *E. coli*, the detection occurs since β-glucuronidase metabolizes another nutrient from the substrate and creates fluorescence that was measured with a UV lamp that emits at 366 nm. The results were obtained by counting the number of positive wells that were converted into a most probable number of total coliforms and *E. coli* present in the samples.

2.3.2. Mycological Analysis

To determine the concentration of filamentous fungi and yeasts present in treated wastewater, 100 mL of each sample (direct and diluted) was filtered through a membrane with a pore size of 0.22 µm (PALL, Port Washington, NY, USA). Each membrane was placed in a Petri dish with malt extract agar culture medium, a general medium that favors fungi growth. The medium was supplemented with the antibiotic chloramphenicol (Oxoid, Hampshire, UK) to avoid bacterial growth. The plates were incubated at 27 °C for 5 days in the dark [23]. The number of colonies was then counted to determine the number of colony-forming units per milliliter. The species that grew in the general culture medium for fungi used in this study were not isolated nor identified at the species level.

2.4. Data Analysis

Log inactivation for total coliforms, *E. coli* and fungi were calculated as log (C_0/C). Microorganism concentrations before and after UV exposure are denoted as C_0 and C, respectively.

The membrane performance was evaluated by the apparent rejection values that compare the permeate quality with the feed. The rejection percentage of bacteria (total coliforms, *E. coli* and fungi) was determined for the filtration experiments using Equation (1):

$$\text{Rejection } (\%) = (1 - \frac{C_p}{C_f}) \times 100 \quad (1)$$

where C_p and C_f are the concentrations of the target microorganisms in the permeate and feed samples, respectively.

The combined treatment performance was evaluated by comparing the target microorganism mass (concentration × volume) in the retentate and feed solutions. The percent retentate treatment of bacteria (total coliforms, *E. coli* and fungi) was determined for the filtration and photolysis combination experiments using Equation (2):

$$\text{Retentate treatment } (\%) = (1 - \frac{C_r}{C_{f,i}}) \times 100 \quad (2)$$

where C_r and $C_{f,i}$ represent the most probable number of the target microorganisms in the reference volume of retentate and feed in the beginning of each experiment, respectively.

3. Results and Discussion

3.1. Experiments with Light-Emitting Diodes

Figure 1 presents the inactivation results obtained with time for the different target microorganisms. The control samples, placed in the dark, maintained the concentration levels of the microorganisms during the experimental time. The UV fluence values determined for each wavelength at different exposure times and the inactivation rate constants are presented in the Supplementary Material section.

The results in Figure 1 show that three small LEDs that emit at 255 and 265 nm reach excellent levels of inactivation in terms of total coliforms and *E. coli*. The 365 nm wavelength revealed to be ineffective in achieving inactivation by direct photolysis. This result was expected due to the low capacity of absorption of light by the DNA at this wavelength. However, this wavelength may be used to activate photocatalytic surfaces, so it was important to verify its inactivation effect by direct photolysis.

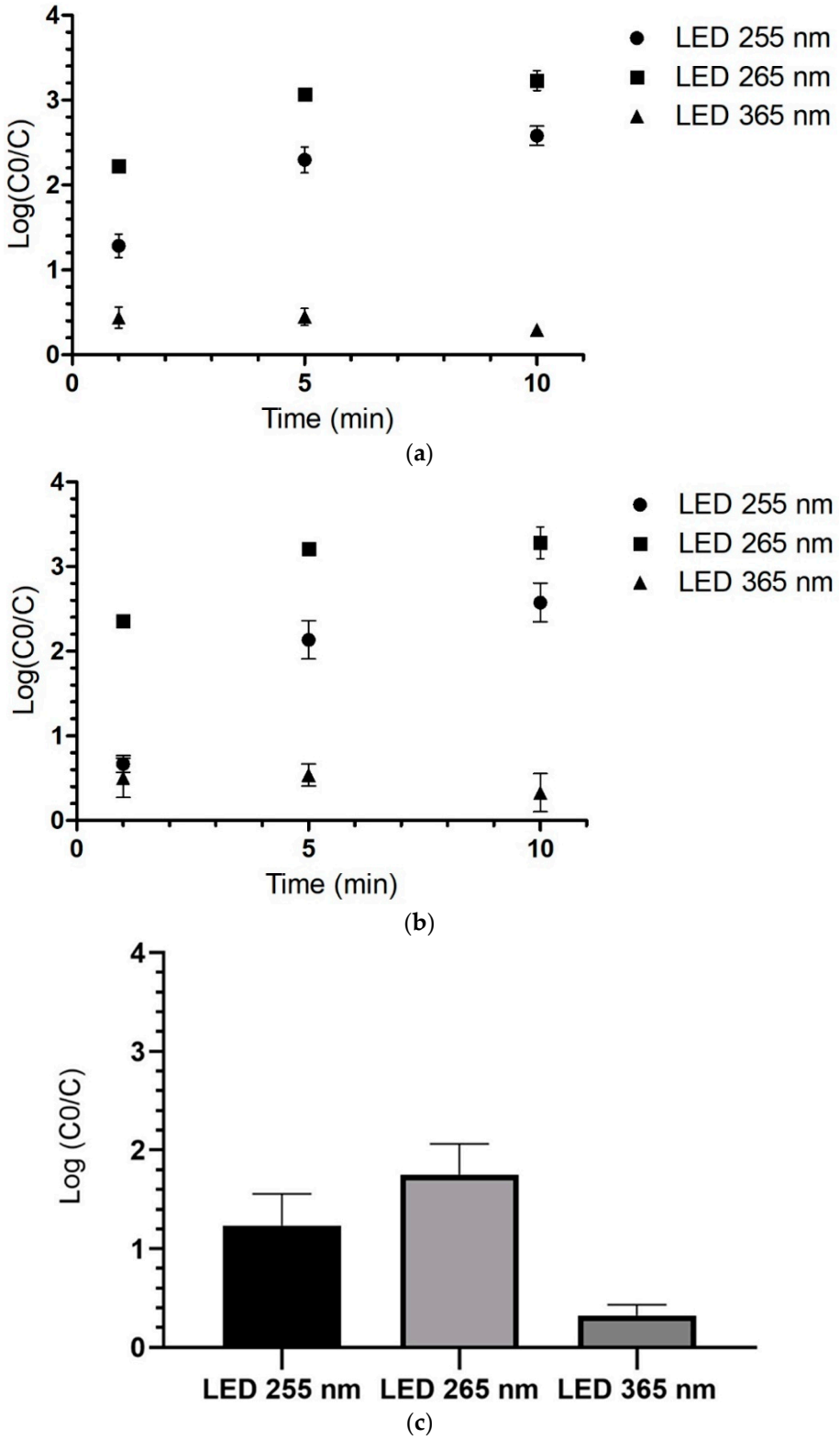

Figure 1. Log inactivation using LEDs that emit at 255, 265 and 365 nm after 10 min of exposure (that correspond to 12, 58 and 1653 mJ/cm^2, respectively); (**a**) total coliforms, (**b**) *E. coli* and (**c**) fungi. Data are expressed as means of two independent experiments. Error bars represent duplicate results.

Other studies have reported the inactivation of microorganisms using LED systems. Oliveira et al. [11,12] reported the inactivation of filamentous fungi of three different species of *Aspergillus* (*A. terreus*, *A. fumigatus* and *A. niger*) in real water matrices and concluded that

LEDs that emit at 265 nm were more efficient than LEDs that emit at 255 nm. Song et al. [10] showed that UV-C LEDs that emit at 265 nm were able to inactivate total coliform and *E. coli* in wastewater and that LED treatments were more efficient than LP-UV treatment for the inactivation of pathogenic bacteria (*Pseudomonas aeruginosa* and *Legionella pneumophila*) and surrogate species (*Bacillus subtilis* spores, bacteriophage Qβ, *E. coli*). Li et al. [24] and Sholtes et al. [25] demonstrated that UV-C LEDs and LP-UV show similar inactivation kinetics for *E. coli* B and MS2 coliphage. Jarvis et al. [8] tested the hypothesis that a full-scale UV LED reactor can match the *Cryptosporidium* inactivation efficiency of conventional mercury UV reactors. The results showed that the full-scale UV LED reactor was at least as effective as conventional mercury UV reactors at the water-quality and drive-current conditions considered. Nevertheless, comparisons between the bench- and full-scale UV LED reactors indicated that improvements in the hydraulic flow profile and power output of the full-scale reactors could help to further improve the efficiency of UV LED reactors for municipal drinking water disinfection.

3.2. Membrane Filtration

Extremely high rejections of the target microorganisms were obtained for all the target microorganisms with the non-modified and modified membranes (Figure S4 in the Supplementary Material section). This was expected due to the porous characterization of the unmodified and modified microfiltration membranes used [17].

None of the target microorganisms were detected in the permeate samples of the modified membrane (Figure S4). As expected, the high permeate quality is maintained in the assays where filtration was combined with LEDs that emit light at different wavelengths (Figure S5).

Furthermore, from the retentate analysis, we observed an extremely high efficiency of treatment with the LEDs that emit at 255 and 265 nm (Figures 2 and 3).

The combination of LEDs that emit at 365 nm with photocatalytic surfaces highly increased their efficiency, leading to a higher treatment of retentate compared with the combination of LEDs that emit at 365 nm with non-modified membranes (Figures 2 and 3). The difference in the results obtained for the treatment of the retentate is due to the production of highly reactive oxygen species by the photocatalytic membranes. The mechanisms of inactivation were not addressed in this study because the study was conducted at occurrence levels in a real wastewater effluent. To understand the mechanisms of inactivation, future studies could be performed with this combined treatment and fortified matrices. However, many inactivation studies have already been conducted using titanium dioxide in suspension or fixed to an inert support and have proposed that the cell membrane and cell wall are the main sites of attack by the reactive radicals, involving lipid peroxidation of the cell wall followed by lesions on the microbial cell walls and leakage of cell content [26].

Previous studies reported inactivation results consistent with this work. Xiong and Hu [27] showed the enhancement of the inactivation of *E. coli* with the increase in UV light intensity using a UV-A LED 365 nm/TiO_2 water treatment. Biancullo et al. [28] tested the efficiency of UV-A LED 381 nm/TiO_2 treatment on urban wastewater and reported a 2-log decrease in the bacterial load. Claro et al. [29] demonstrated that the combination of UV-A LED 385 nm with different photocatalysts (TiO_2, SiZnO, N-SiZnO, and F-N-SiZnO) led to a high inactivation efficiency exceeding 95% using raw river water. Oliveira et al. [30] reported that a newly designed photocatalytic membrane reactor, combining filtration with UV photolysis/photocatalysis under a LP-UV lamp that emits at 254 nm using ceramic modified membranes, was used to treat filtered surface water inoculated with *A. fumigatus*. The results showed that photocatalysis was able to cause the deformation of spores and led to changes in membrane permeability and enzymatic activity.

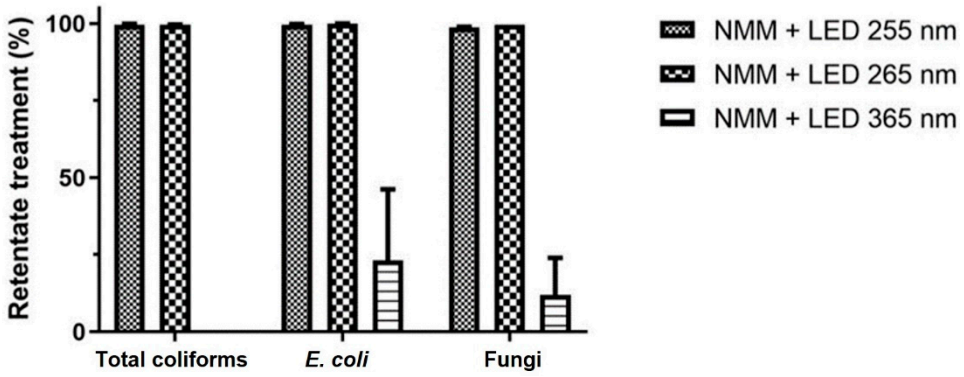

Figure 2. Retentate treatment percentage of total coliforms, *E. coli* and fungi after dead-end filtration of the non-modified membrane with three different LEDs (255, 265 and 365 nm) inactivation.

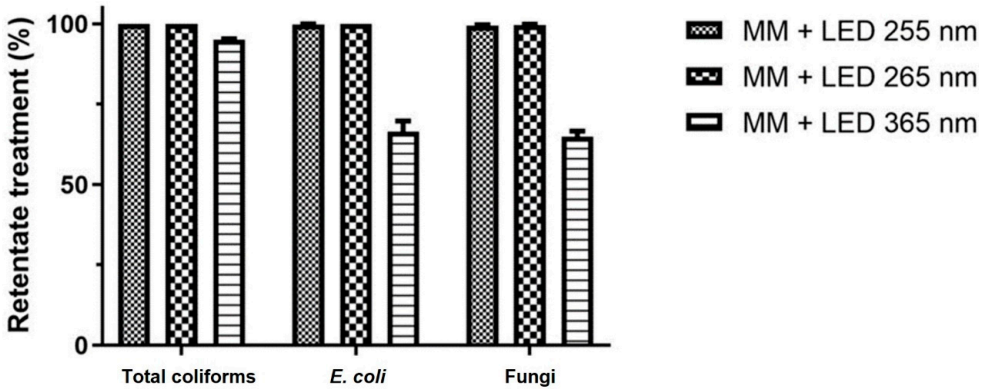

Figure 3. Inactivation percentage of the retentate in terms of total coliforms, *E. coli* and fungi after dead-end filtration of the modified membrane with LEDs that emits at 255, 265 and 365 nm.

4. Conclusions

The results obtained in this work show that:

- Three small LEDs that emit light at 255 and 265 nm achieved extremely high inactivation levels (higher than 2.5-log inactivation) of total coliforms and E. coli after 10 min of exposure in real wastewater effluents.
- The combination of membrane filtration and LEDs that emit at 255 and 265 nm achieve an extremely high quality of permeate (water produced) while guaranteeing an extremely high treatment of the retentate, one of the issues associated with the membrane processes.
- Even though a considerable permeability decrease was observed in the silicon carbide membranes modified with titanium dioxide and silicon dioxide, all the permeate samples were free from contamination with the fecal indicators. The combination of photocatalytic ceramic membranes modified using a solvent-free procedure with LEDs that emit light at higher wavelengths improved the retentate treatment due to the production of highly reactive oxygen species (retentate treatment percentages higher than 65% were obtained).

Future work will include the modification of commercially available high-flux, flat-sheet silicon carbide membranes with a higher surface area that filter from the outside to the inside. The membranes will be tested in a novel hybrid reactor [18]. LED panels that emit in the UV-C and UV-A wavelengths tested in this study will be built so that the non-modified and modified photocatalytic membranes can be easily irradiated which will decrease fouling and increase the feed/retentate treatment due to direct and indirect photolysis.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/w14030292/s1, Figure S1. Experimental setup used in the different assays: (a) LED system;

(b) Membrane filtration system; (c) Combined treatment system. Figure S2. Scanning electron microscopy images obtained for the non-modified and modified membrane: top surface (up) at ×3000 magnification and cross section (down) at ×500 magnification. Figure S3. Scanning electron microscopy with energy-dispersive X-ray spectroscopy mapping obtained for the Si and Ti atoms for non-modified (up) and modified (down) membranes. Table S1. Atomic composition in percentage of C, O, Si and Ti atoms analyzed for membranes (non-modified and modified) before filtration assays. Table S2. UV fluence values (mJ/cm^2) for each wavelength and exposure time. Table S3. Inactivation rate constants. Figure S4. Rejection percentage of total coliforms, *E. coli* and fungi after dead-end filtration using the non-modified and modified membranes. Figure S5. Rejection percentage of total coliforms, *E. coli* and fungi after dead-end filtration of the non-modified membrane (a) and the modified membrane (b) with LEDs that emits at 255, 265 and 365 nm.

Author Contributions: Conceptualization, V.J.P.; methodology, V.J.P., B.O., R.H. and A.P.M.; formal analysis, J.B., J.S., A.P.M. and R.H.; resources, V.J.P. and J.G.C.; writing—original draft preparation, J.B., V.J.P. and A.P.M.; writing—review and editing, J.B., J.S., B.O., A.P.M., R.H., J.G.C. and V.J.P.; supervision, V.J.P.; funding acquisition, V.J.P. and J.G.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Fundação para a Ciência e a Tecnologia through the fellowship SFRH/BD/111150/2015 and project PTDC/EAM-AMB/30989/2017, the Associate Laboratory for Green Chemistry—LAQV (UIDB/50006/2020 and UIDP/50006/2020). iNOVA4Health—UIDB/04462/2020 and UIDP/04462/2020, a program financially supported by Fundação para a Ciência e Tecnologia/Ministério da Ciência, Tecnologia e Ensino Superior, through national funds is acknowledged. Funding from INTERFACE Programme, through the Innovation, Technology and Circular Economy Fund (FITEC), is gratefully acknowledged.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Shannon, M.A.; Bohn, P.W.; Elimelech, M.; Georgiadis, J.G.; Mariñas, B.J.; Mayes, A.M. Science and technology for water purification in the coming decades. *Nature* **2008**, *452*, 301–310. [CrossRef] [PubMed]
2. Hijnen, W.A.M.; Beerendonk, E.F.; Medema, G.J. Inactivation credit of UV radiation for viruses, bacteria and protozoan (oo)cysts in water: A review. *Water Res.* **2006**, *40*, 3–22. [CrossRef] [PubMed]
3. Linden, K.G.; Shin, G.-A.; Lee, J.-K.; Scheible, O.K.; Shen, C.; Posy, P. Demonstrating 4-log Adenovirus Inactivation in a Medium-Pressure Ultraviolet Disinfection Reactor. *J. AWWA* **2009**, *101*, 90–97. [CrossRef]
4. Linden, K.G.; Shin, G.; Sobsey, M.D. Comparative effectiveness of UV wavelengths for the inactivation of *Cryptosporidium parvum* oocysts in water. *Water Sci. Technol.* **2001**, *43*, 171–174. [CrossRef]
5. Rosenfeldt, E.J.; Linden, K.G. Degradation of Endocrine Disrupting Chemicals Bisphenol A, Ethinyl Estradiol, and Estradiol during UV Photolysis and Advanced Oxidation Processes. *Environ. Sci. Technol.* **2004**, *38*, 5476–5483. [CrossRef]
6. Keen, O.S.; Linden, K.G. Degradation of Antibiotic Activity during UV/H_2O_2 Advanced Oxidation and Photolysis in Wastewater Effluent. *Environ. Sci. Technol.* **2013**, *47*, 13020–13030. [CrossRef]
7. Song, K.; Mohseni, M.; Taghipour, F. Application of ultraviolet light-emitting diodes (UV-LEDs) for water disinfection: A review. *Water Res.* **2016**, *94*, 41–349. [CrossRef]
8. Jarvis, P.; Autin, O.; Goslan, E.H.; Hassard, F. Application of Ultraviolet Light-Emitting Diodes (UV-LED) to Full-Scale Drinking-Water Disinfection. *Water* **2019**, *11*, 1894. [CrossRef]
9. Beck, S.E.; Suwan, P.; Rathnayeke, T.; Nguyen, T.M.H.; Huanambal-Sovero, V.A.; Boonyapalanant, B.; Hull, N.M.; Koottatep, T. Woven-Fiber Microfiltration (WFMF) and Ultraviolet Light Emitting Diodes (UV LEDs) for Treating Wastewater and Septic Tank Effluent. *Water* **2021**, *13*, 1564. [CrossRef]
10. Song, K.; Taghipour, F.; Mohseni, M. Microorganisms inactivation by continuous and pulsed irradiation of ultraviolet light-emitting diodes (UV-LEDs). *Chem. Eng. J.* **2018**, *343*, 362–370. [CrossRef]
11. Oliveira, B.R.; Barreto Crespo, M.T.; Pereira, V.J. Small but powerful: Light-emitting diodes for inactivation of *Aspergillus* species in real water matrices. *Water Res.* **2019**, *168*, 115108. [CrossRef]
12. Oliveira, B.R.; Marques, A.P.; Asif, M.; Barreto Crespo, M.T.; Pereira, V.J. Light-emitting diodes effect on *Aspergillus* species in filtered surface water: DNA damage, proteome response and potential reactivation. *Environ. Pollut.* **2021**, *287*, 117553. [CrossRef]
13. Gu, Q.; Ng, T.C.A.; Bao, Y.; Ng, H.Y.; Tan, S.C.; Wang, J. Developing better ceramic membranes for water and wastewater Treatment: Where microstructure integrates with chemistry and functionalities. *Chem. Eng. J.* **2022**, *428*, 130456. [CrossRef]
14. Eray, E.; Candelario, V.M.; Boffa, V.; Safafar, H.; Østedgaard-Munck, D.N.; Zahrtmann, N.; Kadrispahic, H.; Jørgensen, M.K. A roadmap for the development and applications of silicon carbide membranes for liquid filtration: Recent advancements, challenges, and perspectives. *Chem. Eng. J.* **2021**, *414*, 128826. [CrossRef]

15. Eray, E.; Boffa, V.; Jørgensen, M.K.; Magnacca, G.; Candelario, V.M. Enhanced Fabrication of Silicon Carbide Membranes for Wastewater Treatment: From Laboratory to Industrial Scale. *J. Membr. Sci.* **2020**, *606*, 118080. [CrossRef]
16. Fujishima, A.; Rao, T.N.; Tryk, D.A. Titanium dioxide photocatalysis. *J. Photochem. Photobiol. C Photochem. Rev.* **2000**, *1*, 1–21. [CrossRef]
17. Huertas, R.M.; Fraga, M.C.; Crespo, J.G.; Pereira, V.J. Solvent-Free Process for the Development of Photocatalytic Membranes. *Molecules* **2019**, *24*, 4481. [CrossRef]
18. Fraga, M.C.; Huertas, R.M.; Crespo, J.G.; Pereira, V.J. Novel Submerged Photocatalytic Membrane Reactor for Treatment of Olive Mill Wastewaters. *Catalysts* **2019**, *9*, 769. [CrossRef]
19. Cristóvão, M.B.; Bernardo, J.; Bento-Silva, A.; Ressureição, M.; Bronze, M.M.R.; Crespo, J.G.; Pereira, V.J. Treatment of anticancer drugs in a real wastewater effluent using nanofiltration: A pilot scale study. *Sep. Purif. Technol.* 2022; *under review*.
20. Bolton, J.R.; Linden, K.G. Standardization of methods for fluence (UV dose) determination in bench-scale UV experiments. *J. Environ. Eng.* **2003**, *129*, 209–215. [CrossRef]
21. Warden, P.S.; DeSarno, M.S.; Volk, S.E.; Eldred, B.J. Evaluation of Colilert-18 for detection and enumeration of fecal coliform bacteria in wastewater using the U.S. Environmental Protection Agency Alternative Test Procedure Protocol. *J. AOAC Int.* **2011**, *94*, 1573–1580. [CrossRef]
22. Yakub, G.P.; Castric, D.A.; Stadterman-Knauer, K.L.; Tobin, M.J.; Blazina, M.; Heineman, T.N.; Yee, G.Y.; Frazier, L. Evaluation of Colilert and Enterolert Defined Substrate Methodology for Wastewater Applications. *Water Environ. Res.* **2002**, *74*, 131–135. [CrossRef] [PubMed]
23. Pereira, V.J.; Fernandes, D.; Carvalho, G.; Benoliel, M.J.; San Romão, M.V.; Barreto Crespo, M.T. Assessment of the presence and dynamics of fungi in drinking water sources using cultural and molecular methods. *Water Res.* **2010**, *44*, 4850–4859. [CrossRef]
24. Li, G.Q.; Wang, W.L.; Huo, Z.Y.; Lu, Y.; Hu, H.Y. Comparison of UV-LED and low pressure UV for water disinfection: Photoreactivation and dark repair of *Escherichia coli*. *Water Res.* **2017**, *126*, 134–143. [CrossRef]
25. Sholtes, K.A.; Lowe, K.; Walters, G.W.; Sobsey, M.D.; Linden, K.G.; Casanova, L.M. Comparison of ultraviolet light-emitting diodes and low-pressure mercury-arc lamps for disinfection of water. *Environ. Technol.* **2016**, *37*, 2183–2188. [CrossRef]
26. Foster, H.A.; Ditta, I.B.; Varghese, S.; Steele, A. Photocatalytic disinfection using titanium dioxide: Spectrum and mechanism of antimicrobial activity. *Appl. Microbiol. Biotechnol.* **2011**, *90*, 1847–1868. [CrossRef] [PubMed]
27. Xiong, P.; Hu, J. Inactivation/reactivation of antibiotic-resistant bacteria by a novel UVA/LED/TiO_2 system. *Water Res.* **2013**, *47*, 4547–4555. [CrossRef]
28. Biancullo, F.; Moreira, N.F.F.; Ribeiro, A.R.; Manaia, C.M.; Faria, J.L.; Nunes, O.C.; Silva, A.M.T. Heterogeneous photocatalysis using UVA-LEDs for the removal of antibiotics and antibiotic resistant bacteria from urban wastewater treatment plant effluents. *Chem. Eng. J.* **2019**, *367*, 304–313. [CrossRef]
29. Claro, E.M.T.; Bidoia, E.D.; de Moraes, P.B. A high-performance doped photocatalysts for inactivation of total coliforms in superficial waters using different sources of radiation. *J. Environ. Manag.* **2016**, *177*, 264–270. [CrossRef] [PubMed]
30. Oliveira, B.R.; Sanches, S.; Huertas, R.M.; Barreto Crespo, M.T.; Pereira, V.J. Treatment of a real water matrix inoculated with *Aspergillus fumigatus* using a photocatalytic membrane reactor. *J. Membr. Sci.* **2020**, *598*, 117788. [CrossRef]