



Assessing textile wastewater treatment in an anoxic-aerobic photobioreactor and the potential of the treated water for irrigation

Zaineb Dhaouefi^{a,b,c,d}, Alma Toledo-Cervantes^a, Dimas García^{a,e}, Ahmed Bedoui^{c,d}, Kamel Ghedira^c, Leila Chekir-Ghedira^{c,d}, Raúl Muñoz^{a,*}

^a Department of Chemical Engineering and Environmental Technology, School of Industrial Engineering, Valladolid University, Dr. Mergelina s/n., Valladolid 47011, Spain

^b Faculté des Sciences de Tunis, Université de Tunis El Manar, Tunisia

^c Unité de recherche des substances naturelles bioactives et biotechnologie UR12ES12, Faculté de pharmacie, Université de Monastir, Tunisia

^d Laboratoire de Biologie Cellulaire et Moléculaire, Faculté de Médecine Dentaire, Université de Monastir, Tunisia

^e Centro para la Investigación de los Recursos Acuáticos de Nicaragua, CIRA/UNAN-Managua, Apdo. Postal 4598, Nicaragua

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ABSTRACT

The potential of an anoxic-aerobic photobioreactor for the treatment of synthetic textile wastewater (STWW) was evaluated in terms of carbon and nutrient removal, water and toxicity reduction. The potential of the treated water for irrigation of *Raphanus sativus* was also assessed. Despite the low C/N ratio of the STWW, microalgae-bacteria symbiosis supported steady-state removal efficiencies of total organic carbon, total nitrogen and total phosphorus of $48 \pm 3\%$, $87 \pm 11\%$ and $57 \pm 5\%$, respectively, at a hydraulic retention time of 10 days. In addition, a consistent decolorization of the STWW was observed, with disperse orange-3 and blue-1 removals of $\sim 80\%$ and $\sim 75\%$, respectively. This effective STWW treatment was confirmed by the reduction in genotoxicity potential and enhanced growth of *R. sativus* during irrigation with treated water. Finally, the heavy metal content of the soils irrigated with treated water was lower than that of STWW irrigated soils, which highlights the potential of this anoxic-aerobic photobioreactor for the treatment of textile wastewater and water reuse. However, more studies are still needed to elucidate the long-term effects of the treated water over the final consumer health.

1. Introduction

The textile and clothing industry is one of the oldest and largest industries worldwide, and unfortunately, one of the most polluting industrial sectors due to the large volumes of wastewater generated and the inherent presence of hazardous dyes and chemicals [1]. The flow-rate and chemical composition of a textile wastewater depend on the type of fabric, dyeing process and chemicals/auxiliaries used during dyeing. Therefore, textile wastewaters exhibit a wide range of concentrations of chemical oxygen demand (COD), biochemical oxygen demand (BOD) and heavy metals, pH, turbidity, color and salinities [2]. Surprisingly, up to 50% of the dyes applied remain in the process water during industrial dyeing, which causes a significant visual impact despite being present at low concentrations (< 1 ppm) [3].

In this context, the toxic, mutagenic or carcinogenic nature of textile

wastewaters, along with the dye-mediated light absorption, severely deteriorates aquatic ecosystems during their uncontrolled discharge [4,5]. Approximately 70% of the dyes used worldwide are azo and anthraquinone dyes due to their multiple ranges of colors [6]. These dyes are typically used as dispersed dyes in polyester fibers and present hydrophobic properties. However, their effective solubilization requires the use of a high amount of chemicals and auxiliaries at high temperature. In this context, the wastewater containing the non-adsorbed-dyes fraction is extremely recalcitrant due to the complex aromatic structures of both the azo and anthraquinone dyes and the degradation products generated during dyeing. In addition, the sequestration of heavy metals by some chemicals used as dyes solubilization agents eventually increases wastewater toxicity [7,8].

Physicochemical technologies (e.g. Fenton's oxidation) and the combination of physicochemical and biological technologies (e.g.

Abbreviations: AP, alkaline phosphatase; BOD, biochemical oxygen demand; β -gal, β -galactosidase; COD, chemical oxygen demand; DO, dissolved oxygen; HRT, hydraulic retention time; IC, inorganic carbon; ICP-AES, Inductively Coupled Plasma-Atomic Emission Spectrometer; IF, the induction factor; LED, light emitting diode; RE, removal efficiency; SPSS, statistical package for the social sciences; SRT, sludge retention time; STWW, synthetic textile wastewater; TN, total nitrogen; TOC, total organic carbon; TP, total phosphorus; TSS, total suspended solids

* Corresponding author.

E-mail address: mutora@iq.uva.es (R. Muñoz).

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anaerobic-ozonation and aerobic-adsorption treatments) can effectively reduce the toxicity of textile wastewaters [9,10]. Nonetheless, their high chemical and energy requirements, prohibitive operating costs and generation of secondary pollution, which can jeopardize their economic and environmental sustainability, limit the full-scale implementation of physicochemical technologies [11]. In contrast, biotechnologies have emerged as a cost-effective and environmental friendly alternative to physicochemical technologies for textile wastewater treatment, although biological processes are often limited by the low biodegradability of azo and anthraquinone dyes [12,13]. Conventional biotechnologies such as activated sludge processes exhibit high operating cost derived from the intensive aeration required, while anaerobic bacterial processes rarely sustain a complete dye mineralization. In this context, algal-bacterial symbiosis powered by solar radiation has emerged as a cost-effective technology capable of simultaneously removing nutrients and mineralizing recalcitrant organic pollutants from wastewater [14]. However, a complete nutrient removal in algal-bacterial systems often requires a high C/N ratio, which is not the case of textile wastewaters. The cost-effective treatment of wastewaters with a low C/N ratio has been recently achieved in an innovative anoxic-aerobic photobioreactor configuration where the biodegradable carbon is oxidized in the anoxic chamber using the $\text{NO}_3^-/\text{NO}_2^-$ produced from NH_4^+ nitrification in the photobioreactor [15,16]. In this context, on one hand microalgae provide, via photosynthesis, the O_2 required in the photobioreactor for nitrification and the aerobic heterotrophic biodegradation of the remaining organic pollutants [16]; and bacteria release growth-promoting factors and CO_2 that can improve microalgal growth [17,18]. Furthermore, some studies have shown the potential of symbiotic algal-bacterial consortia to decolorize dyes and metabolize the aromatic amines typically released during the physicochemical oxidation of dyes, which are even more hazardous than their parents' dyes [19,20,21]. Despite the high potential of algal-bacterial processes for wastewater treatment, only few studies have focused on the bioremediation of textile wastewaters [22–24].

This research aimed at evaluating the performance of a novel anoxic-aerobic algal-bacterial photobioreactor during the treatment of a synthetic textile wastewater characterized by alkaline pH (12.1 ± 0.1), low C/N ratio (0.9), and COD and BOD concentrations of 387 ± 1.0 and $136 \pm 18 \text{ mg-O}_2 \text{ L}^{-1}$, respectively. The influence of flue gas CO_2 supplementation and wastewater feeding regime on carbon and nutrient removal, water decolorization and toxicity reduction was assessed. Finally, the impact of raw and treated wastewater irrigation on *Raphanus sativus* growth and soil properties was comparatively evaluated.

2. Materials and methods

2.1. Synthetic textile wastewater composition

The synthetic textile wastewater (STWW) here used mimicked the composition of a real textile wastewater (Table 1). The STWW was weekly prepared and stored at 4°C prior utilization. The soaping agent, antcrease and dispersing agent were kindly provided by CHT/BEZEMA group (Beitlich GmbH, Spain), which manufactures textile additives. Disperse blue-1 (1,4,5,8-Tetraaminoanthraquinone) and disperse orange-3 (4-(4-Nitrophenylazo)aniline) were purchased from Sigma-Aldrich (Madrid, Spain), while the rest of the chemicals were obtained from Panreac (Barcelona, Spain).

2.2. Biodegradability of the STWW

The biodegradability of the STWW was assessed in 250 mL Erlenmeyer flasks containing 190 mL of STWW inoculated with 10 mL of aerobic activated sludge from Valladolid wastewater treatment plant (Spain). The Erlenmeyer flasks were incubated at 200 rpm and $25 \pm 1^\circ\text{C}$ under dark conditions for 10 days. The dissolved total

Table 1
Synthetic textile wastewater composition [13].

Parameter	Concentration (mg L^{-1})	Function
COTOBLANC KRS	330	Soaping agent
BIAVIN BPA	330	Antcrease
MEROPAN DA	170	Dispersing agent
Trisodium phosphate	300	Electrolyte
Sodium hydroxide	1000	Alkaline agent
Sodium hydrosulfite	900	Reducing agent
Acetic acid	170	Acid generation
Ammonium sulfate	600	Electrolyte
Disperse blue-1 (1,4,5,8-Tetraaminoanthraquinone)	12	Dyeing
Disperse orange-3(4-(4-Nitrophenylazo)aniline)	20	Dyeing
pH	12.1 ± 0.1	
Chemical oxygen demand ($\text{mg-O}_2 \text{ L}^{-1}$)	387 ± 1	
Biochemical oxygen demand, BOD_5 ($\text{mg-O}_2 \text{ L}^{-1}$)	136 ± 18	
Carbon/nitrogen (C/N) ratio	0.90	

organic carbon (TOC), inorganic carbon (IC) and total nitrogen (TN) concentrations were analyzed at days 0, 3, 5 and 10.

2.3. Photobioreactor set-up and operation

The experimental set-up consisted of a 0.9 L anoxic bioreactor interconnected to a 2.7 L photobioreactor and a 1 L settler via internal and external recirculation of the liquid broth and settled biomass, respectively (Fig. 1). The anoxic reactor was continuously stirred at 300 rpm and operated under dark conditions to prevent microalgae photosynthesis. The cultivation broth from the anoxic tank overflowed directly into the photobioreactor, while the internal at and external recirculation rates were set at 4 and 0.5 folds the STWW flowrate. The enclosed jacketed glass photobioreactor (AFORA, Spain) was continuously stirred at 300 rpm and maintained at $25 \pm 1^\circ\text{C}$ via water recirculation from a thermostatic bath (Huber, Offenburg, Germany). A cylindrical arrangement of LED lamps stripes (F30 W-12 V) located around the photobioreactor provided a light irradiation of $400 \pm 51 \mu\text{mol m}^{-2} \text{ s}^{-1}$ under a 12 h:12 h light/dark illumination regime. The effluent from the photobioreactor was pumped into the settler. The anoxic reactor and photobioreactor were initially filled with an algal-bacterial consortium from a previous culture treating domestic wastewaters in a similar denitrification-nitrification configuration [25]. The concentration of the microalgae inoculum, composed of 68.4% of *Acutodesmus obliquus*, 21.4% of *Scenedesmus quadricauda* and 10.2% of *Scenedesmus tenuispina*, was $1.73 \cdot 10^{10}$ cells L^{-1} . The process was operated at an algal-bacterial sludge retention time (SRT) of 11 days and a HRT of 10 days ($\text{HRT}_{\text{anoxic tank}} = 2.5$ days, $\text{HRT}_{\text{photobioreactor}} = 7.5$ days) along the three operational stages. However, while STWW was continuously fed at 0.36 L day^{-1} in stage I and II, a 12 h STWW feeding at 0.72 L day^{-1} was set in stage III (which resulted in a total daily STWW input of 0.36 L throughout the 3 operational stages). The pH of the STWW during stage I was decreased from 12.1 ± 0.1 to 7.3 ± 0.1 by addition of chloridric acid (37% v/v). A CO_2 -absorption column of 0.3 L was interconnected to the photobioreactor to provide additional inorganic carbon for nitrifying bacteria and microalgae growth, and pH control, during stages II and III (Fig. 1). The absorption column was operated by co-currently pumping the photobioreactor cultivation broth and synthetic flue gas composed of 20% of CO_2 and 80% of N_2 at 1.1 mL min^{-1} during the illuminated periods at a liquid:gas ratio of 10. The oxygen required for the oxidation of the organic matter and ammonium in the photobioreactor was provided by algal photosynthesis during the illuminated period, while air was sparged at 20 mL min^{-1} through a sintered glass diffuser at the bottom of the photobioreactor during the dark periods in stage I and II. However, photobioreactor aeration was carried out exclusively during

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