Science of the Total Environment 648 (2019) 601–608

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Comparison of different detoxification pilot plants for the treatment of industrial wastewater by solar photo-Fenton: Are raceway pond reactors a feasible option?

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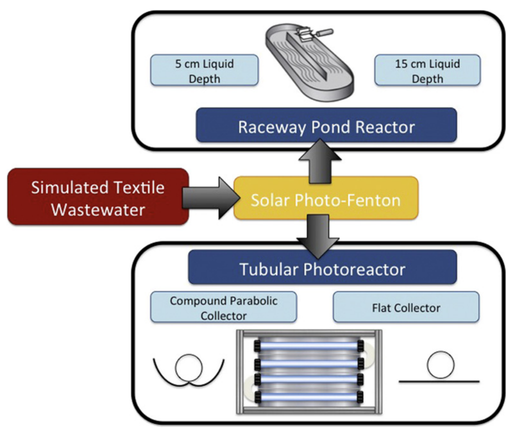
HIGHLIGHTS

- Treatment capacities of different pilot plants for solar photo-Fenton are compared.
- Raceway pond is used to treat industrial wastewater at different liquid depths.
- Flat collector and CPC photoreactor are used to treat industrial wastewater.
- Economic treatment capacity is defined for photoreactor comparison.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 25 April 2018
Received in revised form 24 July 2018
Accepted 10 August 2018
Available online 15 August 2018

Editor: Paola Verlicchi

Keywords:
Solar photoreactor
Photo-Fenton
Dye wastewater
CPC
Raceway

ABSTRACT

This paper represents a first approach in the study of photoreactor selection to treat industrial wastewater using solar photo-Fenton. In this context, simulated textile industry effluent containing a mixture of four dyes at different initial dissolved organic carbon (DOC) concentrations (45, 90, 180 and 270 mg/L) was treated by using three different solar reactor geometries: (i) tubular (5 cm diameter) provided with compound parabolic collector (ii) tubular (5 cm diameter) provided with flat collector and (iii) open channels forming raceway ponds with two liquid depths (5 and 15 cm). For comparison purposes, mineralisation percentages over 75% and chronic toxicity reduction were set as treatment goals. Regardless of the initial DOC concentration, negligible differences in terms of treatment time and hydrogen peroxide consumptions were found between the flat collector and compound parabolic collector photoreactors. Conversely, the treatment in the raceway pond reactors always resulted in higher values. In spite of this, when the photoreactors were compared in terms of treatment capacity (mg of DOC removed/m^2 min) the raceway pond reactor at 15 cm of liquid depth presented the best results, with values as much as two or three times higher than those of the tubular reactors, except for the wastewater with 270 mg of DOC/L for which the raceway pond reactor at 5 cm liquid depth became the best option. When the treatment capacity is modified to include the photoreactor investment (mg of DOC removed/€ m^2) the differences between the

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https://doi.org/10.1016/j.scitotenv.2018.08.143

raceway pond reactor at both liquid depths and the photoreactors with solar collectors increased by as much as two orders of magnitude, which demonstrates the potential application of the former for the treatment of industrial wastewater by solar photo-Fenton.

1. Introduction

Any industrial application of a photocatalytic process requires a photoreactor, which is distinctive from classic reactors because the rates of photochemical reactions are functions of the position in the reaction zone. This is due to the unavoidable non-uniformity of the radiation field caused by photon absorption (homogeneous media) or absorption and scattering (heterogeneous media) (Alfano et al., 2009). Water treatment photoreactors can essentially be divided into two different types: (i) Concentrating reactors (ii) Non-concentrating reactors (Malato et al., 2007). Within the first category, the most used photoreactors were probably parabolic-trough collector (PTCs), which consist of a structure that supports a reflective concentrating parabolic surface. Although the PTC has been proved effective for wastewater treatment (Spasiano et al., 2015), its use has been neglected due to high cost. On the other hand, Compound Parabolic Collectors (CPC) considered non-concentrating devices or low concentrating devices (1–1.5 sun), are the most extensively used photoreactors in this field. CPCs present two connected parabolic mirrors and an absorber tube in the axis. Thanks to the design of the reflector surface most of the incident radiation, including the diffuse component, can be used so that the light is reflected through the upper part of the tube, favouring the complete illumination of the system (Goswami et al., 1997).

In spite of the importance of radiation distribution in the tubes, it has been reported that the photo-Fenton process can reach photosaturation from a certain UV radiation level (Carra et al., 2014a) and also that the production of radical excess can be counter-productive for the process (Cabrera Reina et al., 2017). These statements are extremely important as photo-Fenton could be also efficiently run with more simple photoreactors selecting adequate operation conditions. In this sense, very interesting works have been recently carried out using non-concentrating low cost raceway pond reactors (RPR) for the treatment of contaminants of emerging concern (μg/L–ng/L) (De Obra et al., 2017; Arzate et al., 2017; Carra et al., 2014b). The results have proved that, for micropollutant removal by solar photo-Fenton, UV light excess likely occurs when using tubular reactors and, therefore, increasing the path length could be a good solution to improve process efficiency.

Industrial wastewater treatment requires a much higher amount of radicals as the pollutants are present in the order of mg/L and, consequently, the above-explained considerations are not valid for this particular case. So far, the CPC photoreactor is considered the best choice when treating industrial (toxic and non-biodegradable) wastewater by solar photo-Fenton although some key points could also support the use of different detoxification plants. Industrial wastewater is characterised by its important variability. A wide range of characteristics accounting for inorganic salts, dissolved organic carbon (DOC) concentration, volatile compounds, suspended solids, or colour, among many others can be found depending, mainly, on the industry sector (Doumic et al., 2015; Chatzisymeon et al., 2006).

Although major efforts are being made to study new photoreactor configurations, this research field seems to be still in its infancy, especially considering that most of the photoreactor development carried out in the 90s was done based on heterogeneous (TiO_2) photocatalysis experiments. While 333 scientific papers can be found when searching for the terms “photo-Fenton + CPC” in a specific search engine (www.sciencedirect.com), only 24 were found when searching for “photo-Fenton + Raceway”. Furthermore, intermediate configurations between RPRs and CPC photoreactors like the use of simpler solar collectors, for instance the flat collector (FC), remain almost unexplored.

This paper represents a first step in the study of photoreactor selection to treat industrial wastewater using solar photo-Fenton. In this context, simulated textile industry effluent containing a mixture of four dyes at different initial DOC concentrations (45, 90, 180 and 270 mg/L) was treated using three different solar detoxification pilot plants: (i) CPC photoreactor (ii) FC photoreactor and (iii) RPR. Two liquid depths (5 and 15 cm) were used for RPR while 5 cm tube diameter was used for tubular reactors. With the aim of achieving mineralisation percentages over 75% and reducing chronic toxicity, at least, to slight levels, the photoreactor performances were compared in terms of treatment time, hydrogen peroxide consumption, accumulated energy (Q_{UV}) and treatment capacity (TC) expressed as the mass of mineralised DOC per unit of time and photoreactor surface unit. Also a new parameter, the economic treatment capacity (TC_E) was defined.

2. Materials and methods

2.1. Reagents and analytical methods

Textile industry effluent was simulated by dissolving a mixture of four dyes with each compound providing equal DOC concentration to wastewater. The dyes were purchased from Sigma-Aldrich, in tap water: (i) Acid Red 1 (CAS Number 3734-67-6, Molecular Weight (MW) 509.42 g/mol), (ii) Acid Yellow 17 (CAS Number 6359-98-4, MW 551.29 g/mol), (iii) Reactive Black 5 (CAS Number 17095–24-8, MW 991.82 g/mol) and (iv) Reactive Orange 16 (CAS Number 12225-83-1, MW 617.54 g/mol). Their chemical structures are shown in Fig. 1. The catalyst, Fe(II), was added in the reaction bulk as $FeSO_4{\cdot}7H_2O$ (99%), which was obtained from Fluka. The oxidant agent, hydrogen peroxide (H_2O_2; 33% w/v), was provided by Sigma-Aldrich. Sulphuric acid (95–97%), used for pH adjustment, was purchased from J.T. Baker.

The decontamination process was followed measuring the DOC concentration using a Shimadzu-V_{CHP} TOC analyser fitted with an autosampler system. The hydrogen peroxide concentration was determined with the ammonium metavanadate colorimetric method proposed by Nogueira et al. 2005 (Nogueira et al., 2005) while iron species concentration in water was measured by means of the o-phenanthroline standardised spectrometric method (ISO 6332). Absorbance measurements were carried out with a Hach Lange DR 5000 spectrophotometer.

2.2. Solar detoxification pilot plants

The experiments were carried out in three types of photoreactors installed at CIESOL, Centro de Investigaciones de la Energía Solar, Almería (Spain):

a) CPC photoreactor: this solar detoxification plant consist of two twin loops each containing two Pyrex glass tubes fitted onto the focus of two CPC mirrors, tilted 37° (local latitude). The total illuminated surface per loop is 0.42 m^2, the illuminated volume is 4.77 L and total volume is 7 L. The simulated industrial effluent is recirculated through the system (reservoir tank, piping and glass tubes) by means of a centrifugal pump.
b) FC photoreactor: this solar detoxification plant follows exactly the same scheme as the previous one but using a flat mirror, also tilted 37° (local latitude), instead of CPCs.