



Comparison of two excilamps and two reactor configurations in the UV-H₂O₂ removal process of amaranth



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ABSTRACT

Nowadays dyes are used in many industrial activities and their presence in wastewater is quite common and involves different environmental and health problems. In addition, dyes are usually recalcitrant compounds and conventional treatment are not appropriate for their removal. So there is great interest in finding alternative removal processes, as the one based on excimer lamp technology for the removal of amaranth dye described in the present work. Two excilamps and two reactor configurations have been tested: a batch reactor with KrCl or XeBr excilamp and a KrCl flow-through photoreactor. After comparing the results for both lamps in the batch reactor, the KrCl excilamp has proven to be more efficient both in terms of conversions achieved and energy consumption and, by this, it has been selected to be used in the flow-through photoreactor. For both reactor configurations, several experimental series were done to analyze the influence of the different operational variables on the photoprocess and the optimal mass ratio between hydrogen peroxide and dye has been established. Also, it has been proven that the use of Fenton reagent improves the process efficiency, particularly in the case of the XeBr lamp.

In addition, a kinetic model, based on a previous one developed by the authors for a flow-through photoreactor and slightly modified to can also apply it to batch reactors, has been applied. The model has been validated with an excellent fitting of the experimental data for all series and both reactor configurations.

1. Introduction

With the aim to increase sales, companies tend to offer more attractive products, something that can be achieved by means of color, since it allows us to differentiate the products, makes them more striking or customize them. Industries such as cosmetics, food or textile are just some examples of this tendency. Dyes can also be used for more technical purposes, such as those found in pharmacy or laboratories.

The main problem derived from the multiple applications of dyes is the presence of colored compounds in wastewater that will end up being discharged into the natural environment. Despite the apparently innocuous aesthetic effects on the receptor medium, the transmission of solar energy decreases by the presence of dyes and this affects the photosynthesis and reduces the concentration of oxygen in the ecosystem. In addition, some of these compounds may be toxic to organisms of the aquatic medium [1,2], affecting their development or behaviour [3,4]. Another important aspect of dyes is that, in some cases, the products formed during their decomposition can become more toxic than the dyes themselves.

Azo dyes are the most common and numerous dyes and among them

amaranth has been the one selected for this research. It is used in the coloration of textile fibers, leather, paper, thermoplastics, wood, cosmetics, inks and in the food industry with the number E123 of the International Numbering System (INS) [5]. Amaranth also has applications in biological research, such as detecting changes in membranes potential [5]. According to some recent research studies, as one with *in vitro* human lymphocytes [6], it seems that amaranth can be toxic and, by this, it is not authorized anymore in EEUU as additive in the food, pharmaceutical and cosmetic industry. As a consequence, amaranth can be considered as a pollutant which must be removed from wastewaters.

Different physical, chemical and biological methods have been used for the removal of amaranth from wastewaters. Adsorption treatments have been tested with good results with adsorbent materials such as zeolites or activated carbon [7]. However this technique involves the pollutant change of phase and by this, although almost total removal from wastewater can be achieved, the pollutant remains in the adsorbent, which must be treated further. Membrane processes have been applied as well among the physical treatments, using nanofiltration and reverse osmosis with polysulfone and polyethylamine membranes [8,9]. In this case the main disadvantage is the high energy

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Nomenclature

<i>A</i>	Amaranth	$k_L a$	Volumetric mass transfer coefficient, (min^{-1})
<i>a</i>	Parameter defined in Eq. (4.2), ($\text{mg L}^{-1} \text{min}^{-2}$)	k_r	Pseudo first order kinetic constant, (min^{-1})
<i>a'</i>	Parameter defined in Eq. (4.4), ($\text{mg}^{-1} \text{L min}$)	<i>r</i>	Correlation coefficient, (dimensionless)
<i>b</i>	Parameter defined in Eq. (4.2), ($\text{mg L}^{-1} \text{min}^{-2}$)	r_A	Reaction rate, ($\text{mg L}^{-1} \text{min}^{-1}$)
<i>b'</i>	Parameter defined in Eq. (4.4), (min)	r_{dif}	Mass transfer rate, ($\text{mg L}^{-1} \text{min}^{-1}$)
<i>c</i>	Parameter defined in Eq. (4.2), ($\text{mg L}^{-1} \text{min}^{-1}$)	<i>t</i>	Reaction time, (min)
<i>d</i>	Parameter defined in Eq. (4.2), ($\text{mg L}^{-1} \text{min}^{-1}$)	$t_{1/2}$	Half-life reaction time, (min)
E_A	Energy received in the time unit by amaranth, (W min^{-1})	<i>V</i>	Volume of bulk solution, (mL)
ϵ	Quantum yield defined in Eq. (3.6), ($\text{mg L}^{-1} \text{W}^{-1}$)	V_R	Volume of photoreaction zone in the film, (mL)
<i>I</i>	Intensity of radiation, (W)	X_A	Conversion of amaranth, (dimensionless)
k_{c1}	Kinetic constant of direct photolysis in the film, (min^{-1})	[<i>A</i>]	Concentration of amaranth at time <i>t</i> , (mg L^{-1})
k_{c2}	Kinetic constant with hydrogen peroxide in the film, ($\text{mg}^{-1} \text{L}^{-1} \text{min}^{-1}$)	[<i>A</i>] ₀	Initial concentration of amaranth, (mg L^{-1})
k_E	Proportionality constant defined in Eq. (3.5), (dimensionless)	[<i>A</i>] _{im}	Concentration of amaranth in the film, (mg L^{-1})
		[H_2O_2]	Concentration of hydrogen peroxide at time <i>t</i> , (mg L^{-1})
		[H_2O_2] ₀	Initial concentration of hydrogen peroxide, (mg L^{-1})
		[H_2O_2] _{im}	Concentration of hydrogen peroxide in the film, (mg L^{-1})
		[<i>Prod</i>] _{<i>i</i>}	Concentration of product <i>i</i> , (mg L^{-1})

consumption and also the concentrate obtained that must be treated as well. Biological treatments have been used with bacterial culture such as *Pseudomonas* [10], fungi [11] or peroxidase enzymes extracted from *Arundo donax*, *Typha angustifolia* or *Phragmites australis* [12], however, due to the toxicity of pollutants, some of these treatments can be applied only for low dye concentrations.

Chemical methods like coagulation-flocculation, ionic exchange or electro-coagulation have proven to be effective for the removal of azo dyes like amaranth [13–15]. Among the chemical methods, oxidation processes offer the important advantage that total mineralization of the pollutant can be achieved in most cases. Subcritical and supercritical water oxidation have been used attaining removal efficiencies of azo dyes between 80–90% [16,17], although extreme temperature and pressure conditions are normally required for these treatments.

Advanced oxidation processes (AOPs) offer an interesting alternative that has been attracting increasing interest over the last years. AOPs are based on the generation of strong oxidizing agents, like HO•, allowing oxidation and total mineralization of water pollutants even at very low concentrations due to their high reactivity and low selectivity [18]. Both photochemical and non-photochemical AOPs such as ozonation [19], Fenton treatment [20] or UV-Fenton [21] have been successfully used for amaranth removal from wastewater.

Among the photochemical AOPs, excimer technology has been selected for the present work because in the literature there are no references about the use of this technology in the removal of this dye. Excimer lamps or excilamps are new UV sources based on transitions of exciplex (rare gas halides) or excimer molecules (rare gas or halogen dimmers) formed as a result of an electric discharge. They are attractive alternatives to commonly used mercury lamps because they have some important advantages including the absence of elemental mercury, long lifetime (from 1000 to 10,000 h), geometric freedom and high photon flux. Their main advantage is the emission in a narrow-band UV radiation, nearly monochromatic and matching the dissociation energies of bonds of the main organic compounds, especially if barrier discharge lamps are used since they provide a narrower emission spectrum. From an environmental point of view, the absence of toxic mercury is a remarkable characteristic [22–25]. Over the last years excimer technology has been applied to the removal of different organic pollutants that can be found in industrial wastewaters, using different lamp configurations [26–37].

In addition, different kinetic studies on oxidation treatments of amaranth and other dyes can be found in the literature [38–41]. In most processes the progress curves can be successfully fitted to a pseudo first order kinetic model and, commonly, the first order kinetic parameter shows a dependence on the operational variables, mainly the concentration of pollutant, which is not the expected behaviour for a true

first order kinetics. In a previous work [42] the authors studied the photodegradation of the methylene blue dye using an exciplex KrCl flow-through photoreactor, and a kinetic model, that explained the dependence of the first order kinetic parameter on the operational variables, was developed and successfully validated.

In the present paper, a comparative study is carried out, firstly in a batch reactor with two different excilamps of KrCl and XeBr, to test their efficiency in the removal of amaranth. Once the best lamp is selected, the second part of the study is focused on the photodegradation of amaranth by using an exciplex KrCl flow-through photoreactor. This is the reactor configuration that allows us to work, in the future, not only in discontinuous but also in semicontinuous or continuous mode, as a previous step for a further scale-up of the process. In addition, the previously developed kinetic model, with some small modifications to be suitable for batch reactors, is checked and validated with the data obtained from both reaction systems.

2. Materials and methods

2.1. Reagents

Amaranth (85–95% w/w) was purchased from Sigma-Aldrich Fine Chemicals, hydrogen peroxide (33 % w/v) was purchased from Panreac, ferrous sulphate was purchased from Probus, catalase solution (1340 U/mg solid) and aluminium potassium sulphate ($\geq 98.0\%$) were purchased from Sigma-Aldrich Fine Chemicals. Other chemicals were of analytical grade and were used without further purification.

2.2. Materials

The equipment used for the experiments consist in two barrier discharge excilamps (purchased from the Institute of High Current Electronics of the Siberian Branch, Russian Academy of Sciences). The XeBr and KrCl excilamps emitting maximum UV radiation at 283 and 222 nm, respectively, were of cylindrical geometry covered by a metal case having an UV exit window with an area of 75 cm². The exit window was oriented at a fixed distance of 3 cm over a vessel of 100 mL of capacity and 4.5 cm external diameter. The average radiation intensity delivered to the solution was 17.12 and 2.47 mWcm⁻² for XeBr and KrCl excimer lamps, respectively.

Also a flow-through KrCl photoreactor with an irradiation zone length of 30 cm and an internal diameter of 2 cm, which provides an irradiation area of 188.5 cm² and with an average radiation intensity of 2.38 mWcm⁻² was used in the experiments.

Data of radiation intensity, both for the two excilamps and for the flow-through photoreactor, were provided by the manufacturer.

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