TREATMENT OF TEXTILE DYE WASTEWATER BY SOLAR-POWERED ELECTROOXIDATION

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ABSTRACT

Wastewaters represent one of the biggest problems today as they contain many pollutants, some of which are well-known pollutants, while there are also those whose impact on human health and the ecosystem has not been fully investigated. There are many persistent pollutants that cannot be removed by conventional wastewater treatment technologies, which causes great concern regarding their impact on human health. For this reason, the study, development and implementation of advanced technologies in wastewater treatment are of great importance. In recent years, new advanced oxidation processes based on electrochemical technology - electrochemical advanced oxidation processes (EAOPs), have been developed for the treatment of persistent pollutants. EAOPs are known not only as very efficient technologies but also as large consumers of electricity. In this research EAOP were used for the treatment of textile dye wastewater. In order to eliminate their biggest drawback, instead of being powered by the distribution network, the electrochemical reactor was powered by solar panel. A battery was used to store the energy obtained from the solar panel to ensure continuous operation and avoid energy fluctuations. The key parameters that affect the efficiency of the process, the economic feasibility and the potential for real-scale application were examined.

Keywords: Green energy, EAOPs, electrooxidation, dye removal.

INTRODUCTION

Global industrialization and urbanization have significantly increased both the volume and the complexity of wastewater. Many pollutants, especially micropollutants such as pesticides, pharmaceutical residues and dyes are resistant to removal by conventional wastewater treatment methods (coagulation, biological and chemical oxidation, etc.), thereby posing risks to both ecosystems and human health [1]. Therefore, there is a growing demand for more efficient wastewater treatment (WWT) systems. When selecting an appropriate WWT technology, several criteria must be considered, including efficiency, sustainability, compliance with environmental regulations, and economic viability [2]. By developing advanced technologies, such as advanced oxidation processes (AOPs) it is possible to produce high-quality water. AOPs are based on the formation of highly reactive hydroxyl radicals (OH·) and other oxidants, which degrade pollutants either through mineralization or by transforming them into less toxic by-products [3], [4]. These processes include UV/H_2O_2, ozonation, Fenton and photo-Fenton reactions, as well as electrochemical approaches - are among the most effective methods for wastewater treatment containing organic pollutants.

AOPs in which OH· formation is initiated by electricity are called electrochemical advanced oxidation processes (EAOPs). These processes are based on the electrolytic in situ generation of OH· in an electrochemical reactor (ECR) and include processes such as electrochemical oxidation, electro-Fenton, photoelectrochemical and sono-electrochemical technologies, and capacitive deionization [5]. The most popular and most researched EAOP is electrochemical oxidation/electrooxidation (EO), in which pollutants are oxidized/degraded either directly on the surface of the anode, or indirectly through intermediates. The main advantages of EAOPs include their wide applicability, the complete mineralization of organic pollutants, the potential for integration with solar photovoltaic and thermal energy, the additional production of useful by-products (e.g., H_2), the possibility of automation, ease of handling, the simplicity of equipment, and safe operation [3], [6]. On the other hand, the main drawbacks are their high energy consumption, sensitivity to wastewater composition (e.g., chlorides induce the formation of toxic by-products), and the low conductivity of some wastewaters, which requires the addition of supporting electrolytes (salts). Furthermore, electrode activity may decrease and electrode lifetime may be shortened due to the deposition of organic matter on their surface [7], [8], [9].

Economic analysis of electrochemical WWT systems shows that the main costs are the initial investment and the electricity consumption during the electrolysis process. Investment costs include the design and construction of the ECR system, the provision of electrolytes in the case of low conductivity wastewater, and the procurement of appropriate electrode materials. However, the biggest challenge for the widespread application of EAOP technologies remains the high energy consumption, which also represents the most significant financial burden [10]. Since, energy requirements represent the main limitation for large-scale application of EAOPs the focus has shifted to integrating renewable energy sources. In this sense, solar photovoltaic systems have been developed to allow the direct powering of electrochemical reactors with photovoltaic (PV) panels or PV–battery systems [11]. The direct use of electricity from the PV array to power the ECR could provide energy autonomy for solar EAOPs as it does not require any connection to the grid [12], [13]. In addition, the use of intermediate components for energy storage (batteries) is being investigated for achieving more efficient integrated technologie. Management of PV energy and storage (e.g., redox flow batteries) increases stability and treatment efficiency [14]. It is important to note that PV systems produce direct current (DC), which can be directly used by the EAOPs system.

Although it is still under development for optimization and further improvements, numerous results on independent, so-called "off-grid" PV modules that drive the electrochemical degradation of various organic pollutants at the laboratory/pilot level are readily available in the literature. Successful pilot and industrial-scale implementations exist in textile effluents, olive mill wastewater, hospital effluents, and municipal sewage [15], [16]. These cases show that scale-up is

feasible, but site-specific testing is essential due to variations in pollutant load and water chemistry.

Laboratory testing is a crucial step prior to any scale-up process, as it enables a realistic assessment of the applicability of AOPs depending on the contaminant type, water matrix composition, and intended process scale. To ensure practical relevance, experiments should be conducted in real water matrices and under the same conditions.

From a benchmarking perspective, laboratory experiments provide the basis for evaluating new solutions in terms of cost, energy efficiency, and reliability compared to established processes, supporting decisions on whether to proceed to demonstration or pilot scale. Since traditional scale-up is expensive and time-consuming, a structured three-step approach (theoretical application screening, lab-scale performance testing, and cost and energy comparison with a benchmark process) offers a systematic framework to develop scalable and industrially relevant AOPs [17].

Recent research on wastewater treatment highlights several practical insights for policymakers and plant operators when considering AOPs and EAOPs, more precisely PV-AOPs and PV-EAOPs. EAOPs show strong potential when combined with other conventional WWT methods (e.g., biological treatment). This hybrid approach reduces energy demand and improves biodegradability of effluents, enabling compliance with discharge limits even for persistent organic pollutants and pharmaceuticals [18], [19]. Therefore, it should avoid stand-alone deployment and instead plan for pre/post-treatment integration. Moreover, emerging approaches (e.g., solar STEP processes) enable synergies such as hydrogen production alongside wastewater treatment [6], [20]. These systems should consider not only as treatment units but also as strategic assets within circular economy models.

Energy cost remains a key barrier to the widespread application of these technologies. Studies consistently show that powering electrochemical systems with solar PV or hybrid solar-battery setups improves sustainability and reduces operating costs [3], [14]. Challenges also include electrode fouling, by-product formation, and the need for supporting electrolytes [7], [18]. Proactive maintenance, the selection of durable electrode materials, and process monitoring are critical managerial priorities.

Another important tool in the analysis of the sustainability of WWT systems, in addition to energy analysis, is life cycle assessment (LCA). This analysis can be used to assess resource consumption in a WWT plant and its overall impact on the environment from the very beginning (design and construction of the plant) until the end of the plant's operating life. There are several studies available in the literature on the application of LCA in AOPs for wastewater treatment [8], [21], [22], [23], [24], [25], which conclude that LCA is a useful tool to support decision-making by providing a detailed overview of environmental impacts.

In summary, practice-oriented insights emphasize combining processes, using renewable energy sources, relying on LCA for decision-making, and strategically considering local conditions and long-term benefits, especially regarding energy use and environmental impacts.

METHODOLOGY

The experimental part of the research consists of the EO process powered by electrical energy obtained from a PV array (PV-EO) for the removal of textile azo dye from synthetic wastewater (SWW) prepared in the laboratory. For the research purposes, two different reactors were used. The first reactor (ECR 1) with a smaller capacity was used to determine the key parameters for the EO process, and the second reactor with a larger capacity (ECR 2) was used to optimize the operating parameters. ECR 1, made of glass, had a capacity of 0.5 dm^3, and contained three electrodes of the same dimensions (6 x 5 x 0.2) cm. ECR 1 operates in batch mode. The ECR 2 made of acrylic has a capacity of 1.5 dm^3 of wastewater, with possibility of both batch and recirculating operation modes. The reactor contains eight electrodes (4 anodes and 4 cathodes) of the same dimensions (10 x 12.3 x 0,3) cm and distance between electrodes d=8 mm.

ECR 1 and ECR 2 were powered by electricity obtained from the monocrystalline silicon PV solar panel with a nominal power of 170W, a maximum current of 8.33 A, and a maximum voltage of 20.41 V (SolarFAM, Netherland). The panel was placed on the roof of the Faculty of Technology University of Banja Luka (at an angle of 45°, south oriented). Solar charge controller (MPPT) (SolarFAM, Netherland) was connected between PV and the 12 V battery (MK Powered, 12 V/74 Ah) to ensure that the maximum available electric energy was always drawn from solar cells. The battery was connected directly to output DC voltage regulator to control the current supply to the ECR. The experimental setup for PV-EO process in ECR 1 and ECR 2 is shown in Figure 1.

All experiments were performed at an ambient temperature (≈20°C) with initial SWW volume of 0.4 dm^3 in case ECR 1 and 1 dm^3 of batch and 1.5 dm^3 for recirculating operating mode, in case ECR 2. For the preparation of SWW, Bemacid Red E-TL (BR) dye (CHT GmbH, Germany) was used and sodium chloride, 99.5% NaCl (Lachner, Czech Republic) was added as a supporting electrolyte to increase electrical conductivity of SWW. The initial dye concentration was approximately 100 mg/L. The exact concentration before and after treatment was determined spectrophotometrically (PerkinElmer, LAMBDA 25).

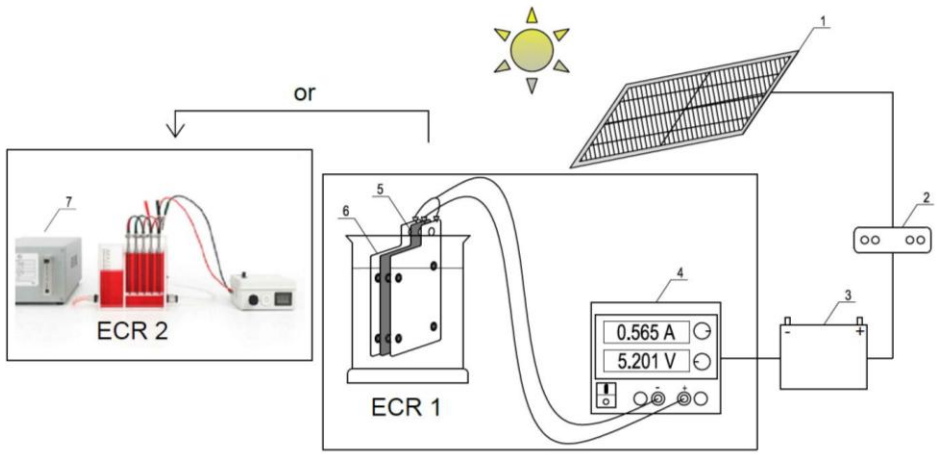


Figure 1. Scheme of PV-EO experimental setup (1- solar panel, 2- MPPT; 3- battery, 4- voltage and current regulator, 5- anode, 6- cathodes, 7- peristaltic pump)

As the electrodes in ECR 1, the following materials were used:

- stainless steel, EN 1.4301 (SS) and graphite felt (GF) were used as cathodes;
- mixed metal oxides (MMO) were used as anodes: Ru mixed oxide on Ti substrate coated with 6g Ru/m^2 (MMO 1) and Ir-Ru mixed oxide on Ti substrate coated with 6g Ru-Ir/m^2 (MMO 2), (Metakem, Germany);
- boron-doped diamond (BDD 1) (Metakem, Germany).

As the electrodes in ECR 2, the following materials were used:

- graphite felt (GF) cathodes;
- boron-doped diamond (BDD 2) anodes (YASA ET (Shanghai) Co., Ltd, China)

Before each experiment, the electrodes were appropriately cleaned depending on the type of material. Different combinations of anode–cathode pairs were placed in the ECR, connected to the output DC regulator, and the current density was adjusted to the desired value.

RESULTS

The results of BR removal from SWW by the PV-EO processes are expressed in terms of removal efficiency (E, %) and calculated according equation (1), where ci and cf are the initial and final concentrations of BR, expressed in mg/L.

$$E = \frac{c_i - c_f}{c_i} \cdot 100\% \quad (1)$$

When operating costs are estimated, in the literature they are most often expressed as the energy required to remove a certain mass of pollutant from the wastewater (kWh/kg of pollutant) or the energy required to treat a specific volume of wastewate (kWh/m^3). Specific energy consumption (W_{sp}) represents one of the most important technological indicators of ECR operation, because it affects the overall cost of treatment. W_{sp} is calculated according to equation (2), where I is current [A], U is voltage on the electrochemical reactor [V], t is the time [h], m is the mass of the removed pollutant [kg], and V is the volume of wastewater [m^3].

$$W_{sp} = \frac{U \cdot I \cdot t}{m_{pollutant}} \left[\frac{kWh}{kg_{pollutant}}\right] \quad \text{or} \quad W_{sp} = \frac{U \cdot I \cdot t}{V_{wastewater}} \left[\frac{kWh}{m^3}\right] \quad (2)$$

In this research, the impact of several important parameters on the efficiency of the EO process was examined, along with the simultaneous calculation of energy consumption. The first part of the research was carried out on a reactor of smaller dimensions (ECR 1), while the second part of the study represents a scale-up using a reactor of larger dimensions (ECR 2).

DISCUSION

The results of the research are presented into two parts. In the first part, the efficiency of PV-EO treatment in a smaller capacity reactor (ECR 1) is presented, while in the second part, the scale-up results are given in a larger capacity reactor (ECR 2).

Efficiency of BR degradation in ECR 1

The choice of anode material is of critical importance for the EO process, since OH· or other oxidizing species are formed on the anode surface. The nature of the anode material affects not only the efficiency of the process, but also the selectivity of the electrode reaction. The anodes used in EO can be classified as “active” and “inactive”. “Active anodes” (low oxygen evolution overpotential) are good electrocatalysts for oxygen evolution (e.g., IrO_2, Pt, RuO_2), and “inactive anodes” (high oxygen evolution overpotential) are poor electrocatalysts for the oxygen evolution [5]. Both active (MMO) and inactive (BDD) anodes were used in this research. The selection of the cathode material in the EO process refers to the selection of a stable, inert material that should prevent accumulation of impurities during the reduction of wastewater on the cathode surface. In the most research, stainless steel or different carbon materials are chosen as the cathode material [26]. Figure 2 shows the efficiency of different electrode pairs in BR degradation during EO treatment. Based on the obtained results shown in Figure 2, it can be observed that treatment is highly efficient, as high dye removal efficiency was achieved in a short time (5 min). The highest efficiency is shown by the BDD anode, while GF proved to be efficient compared to SS. In further research, the most efficient electrode pair was used: BDD - GF.

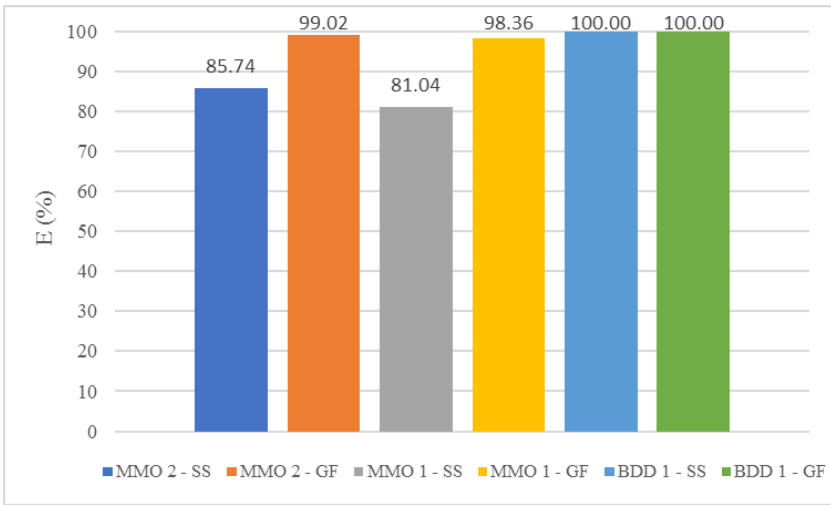


Figure 2. The impact of electrode material on BR dye removal efficiency (t= 5 min; j= 5 mA/cm^2; c_{NaCl}= 1 g/L)

In order to reduce the investment costs of the treatment, reducing the concentration of the supporting electrolyte was considered (Figure 3). Supporting electrolyte is necessary when the conductivity of the wastewater is low. Electrical conductivity is directly related to the voltage on the ECR. When the water conductivity is higher, the voltage on the ECR is lower and, consequently, Wsp is lower, in accordance with equation (2). NaCl was chosen as supporting electrolyte because it is readily available and inexpensive, but also because the generation of active chlorine and other reactive chlorine species (hypochlorite, hypochlorous acid) can occur via indirect oxidation [6], [7], [27]. In this way, in addition to the OH· formed in situ in ECR by the electrolysis of water, these reactive chlorine species enhance the rate of oxidation and the decomposition of organic pollutants in wate. From Figure 4, it can be seen that reducing the NaCl concentration decreases removal efficiency, requiring a longer time for complete dye removal.

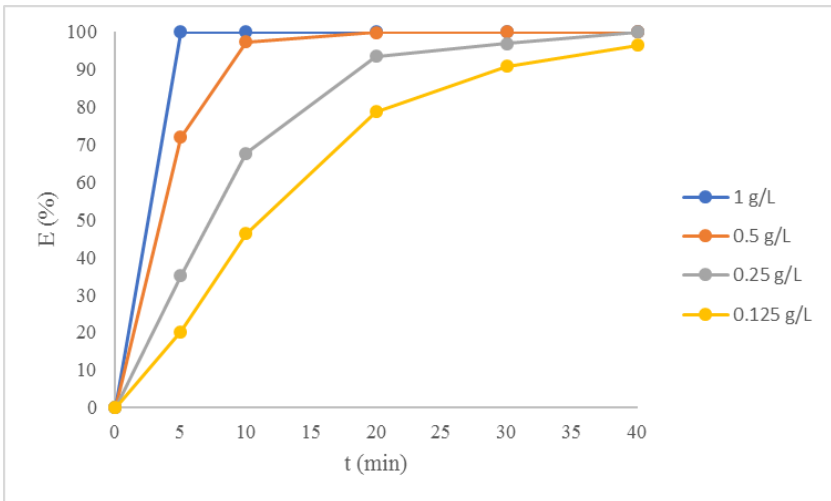


Figure 3. The impact of supporting electrolyte concentration on BR dye removal efficiency at different treatment time (BDD - GF electrode pair; j= 5 mA/cm^2)

As the concentration of supporting electrolyte decreases, the voltage on the ECR increases as expected, which consequently leads to an increase in energy consumption, according to equation (2). Table 1 shows the energy consumption for achieving more than 95% dye removal during the treatment at different concentrations of NaCl.

Table 1. Energy consumption at different concentrations of NaCl

NaCl concentration (g/L)	E (%)	Treatment time (min)	W_{sp} (kWh/kg)	W_{sp} (kWh/m^3)
1	100	5	1.15	0.26
0.5	97.33	10	4.62	1.05
0.25	100	40	17.13	4.00
0.125	96.5	40	20.55	4.20

In EAOPs powered by electricity from the distribution network, extending the treatment time is not desirable, because more energy is consumed, making the process more expensive. In that case, supporting electrolyte concentration is increased in order to reduce the voltage on the reactor and shorten the treatment time, thereby reducing the treatment cost. However, since in these experiments the ECR was supplied with electricity from a solar panel, it allowed the use of lower NaCl concentrations and longer treatment times. In this way, the cost related to the addition of supporting chemicals is reduced. For this reason, the supporting electrolyte concentration of 0.5 g/L for the continuation of the research.

Another important parameter that affects the efficiency of wastewater treatment and energy consumption is the current density. In general, the pollutant degradation rate or the efficiency of mineralization increases with increasing current density up to a certain optimal value. High current densities do not always enhance the oxidation rate or improve pollutant removal efficiency due to the occurrence of the oxygen evolution reaction and/or mass transport limitations. If the current density is above the optimal value it can also lead to an increase in the rate of the OH· consumption, resulting in higher energy consumption [28], [29]. The impact of current density (1.25, 2.5 and 5 mA/cm^2) on the dye removal efficiency by EO was shown in Figure 4.

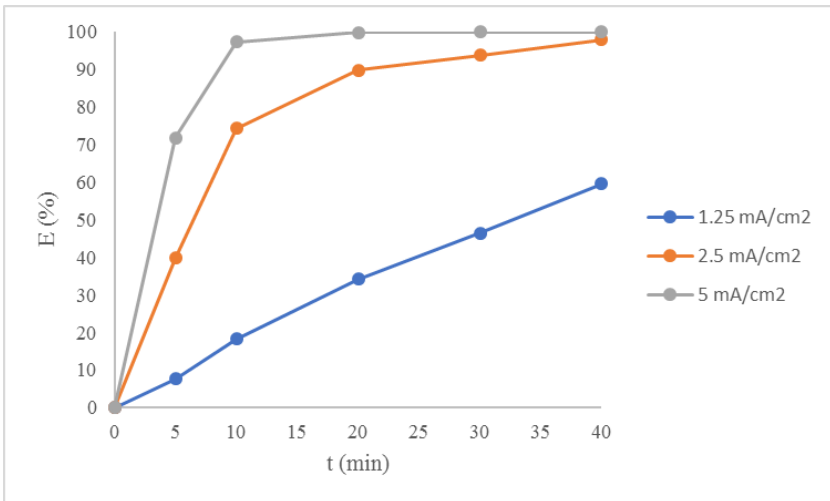


Figure 4. The impact of current density on BR dye removal efficiency at different treatment time (c_{NaCl}= 0.5 g/L; j= 2.5 mA/cm^2; BDD - GF electrode pair)

At higher current densities (2.5 and 5 mA/cm^2), differences in the achieved efficiencies are observed, but for 40 minutes of treatment, almost the same degradation efficiency is achieved. At the lowest tested current density (1.25 mA/cm^2) the process is significantly less efficient. Table 2 shows energy consumption during the treatment at different current densities.

Table 2. Energy consumption at different current densities

Current density (mA/cm^2)	E (%)	Treatment time (min)	W_{sp} (kWh/kg)	W_{sp} (kWh/m^3)
5	97.33	10	4.62	1.05
2.5	98.01	40	2.81	0.68
1.25	59.51	40	2.03	0.26

The current density of 2.5 mA/cm^2 was chosen for further research in ECR 2, because the energy consumption is almost half that at higher current densities, and the efficiency is still satisfactorily high for a relatively short EO treatment time.

Efficiency of BR degradation in ECR 2

After the most important parameters (electrode material, current density, supporting electrolyte concentration) were examined in the ECR 1 and the optimal conditions were selected, the research was continued with a scale-up using a larger-capacity reactor - ECR 2. The goal was to confirm the efficiency of the PV-EO treatment in ECR 2 under the experimental conditions previously selected in ECR 1

The ECR 2 reactor can operate in two modes: batch mode (BM) and recirculation mode (RM). When the reactor operates in BM, the maximum wastewater volume is 1 dm^3, and in RM, it is 1.5 dm^3. It was investigated whether the reactor operating mode affects the efficiency of BR dye removal (Figure 5).

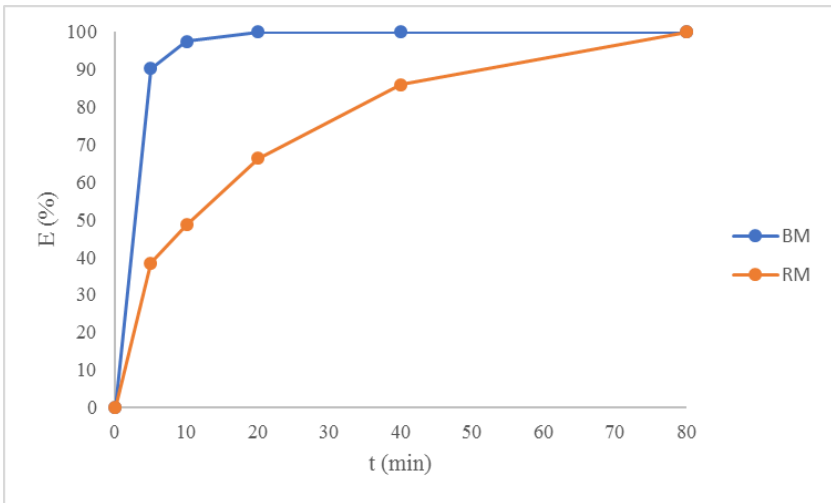


Figure 5. The impact of reactor operation mode on BR dye removal efficiency at different treatment time (BDD - GF electrode pair; c_{NaCl}= 0.5 g/L; j= 2.5 mA/cm^2; 50 dm^3/h flow for RM)

The experiment showed that efficient PV-EO treatment can be achieved in a larger reactor, with a larger amount of wastewater, using the process parameters previously selected as optimal in ECR 1. In ECR 2, 97.5% dye removal was achieved in 10 minutes, and 100% in 20 minutes,

compared to ECR 1 (Figure 4) where, under the same conditions, 98.0% dye removal was achieved in 40 minutes. More electrodes in the reactor results in faster dye degradation.

Based on the diagram shown in Figure 5, it is noticeable that the reactor operating mode has a significant impact on treatment efficiency, particularly at shorter treatment times. Complete (100%) dye removal is achieved in BM in 20 minutes, while in RM, 80 minutes of treatment were necessary for complete dye removal. The lower efficiency in RM is probably due to the shorter contact time with the electrode surface.

Table 3 shows the energy consumption for PV-EO treatment in ECR 1 and ECR 2 in batch operation mode, at c_{NaCl} = 0.5 g/L and j = 2.5 mA/cm^2.

Table 3. Comparison of energy consumption for ECR 1 and ECR 2

Reactor type	E (%)	Treatment time (min)	W_{sp} (kWh/kg)	W_{sp} (kWh/m^3)
ECR 1	98.01	40	2.81	0.68
ECR 2	97.47	10	15.32	1.28

Table 3 clearly shows that the energy consumption is higher in ECR 2, which is expected considering the larger wastewater volume, the greater number of electrodes in the reactor, and the larger electrode surface area.

In order to consider the industrialization of a process, it is useful to estimate the operating cost in terms of energy consumption (kWh/m^3) for treating 1 m^3 of wastewater [30]. The price of electricity varies from country to country. For example, in 2024 the electricity price for industrial use (non-household consumers) in Europe was between 0.0767 - 0.2578 € per kWh [31]. Treatment of 1 m^3 of wastewater containing BR dye as a pollutant, under experimentally determined treatment conditions (c_{NaCl}= 0.5 g/L, j= 2.5 mA/cm^2) costs between 0.098 – 0.323 € for 10 minutes of treatment. In accordance with equation (2), energy consumption can be calculated per 1 kg of removed pollutant. In this case, the energy consumption for removing 1 kg of BR dye under the specified conditions amounts to 1.17 – 3.94 €. This can still be considered as cost-effective, but it should be noted that some treatments could be significantly more expensive. For more polluted wastewater, the treatment would take longer, leading to higher energy consumption.

In order to use energy more efficient and further simplify the process, direct connection of the PV array to the ECR can be implemented, avoiding the use of storage battery systems. This clearly contributes to more efficient energy use (no energy losses in battery charge-discharge cycles) and reduces the investment and maintenance costs of the storage system. But, changes in the intensity of solar radiation during the day lead to significant energy fluctuations, so the ECR is powered by a variable energy source. This power profile can affect the performance and efficiency of WWT [32].

CONCLUSION

The results presented in this paper demonstrate that the degradation of the Bemacid Red E-TL textile dye can be efficiently performed by the electrooxidation process. Moreover, the electrochemical reactor can be powered by renewable energy from a solar panel, making the process efficient, sustainable, and cost-effective. By optimizing the process parameters, such as the concentration of the supporting electrolyte and the applied current density, the treatment duration can be controlled. Shorter treatments would require higher concentrations of supporting electrolyte and higher current density. In this regard, shorter treatments increase costs due to additional chemicals and higher energy requirements, while longer treatments reduce these costs but may shorten electrode lifetime. Process parameters need to be adjusted depending on the type and amount of treated wastewater. However, it can be concluded that EAOPs in combination with

green renewable energy sources have great potential for implementation and scaling up the process to a commercial level.

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DECLARATIONS OF INTEREST STATEMENT

The authors affirm that there are no conflicts of interest to declare in relation to the research presented in this paper.

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