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Free Chemicals - Sonoelectrochemical Treatment as a Biocompatible Procedure for Decolourization of Anthraquinone Dyes

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Abstract

Dye compounds are known as one of the most important environmental pollutants. Up to now, several methods have been offered to eliminate these pollutants from industrial wastes, especially in the textile industry. A method that has received a great deal of attention in recent years is the sono-electro-chemical method. In this study, using the Central Composite Design, the interactive effects of four essential variables such as pH, initial dye concentration, time, and applied potential on sonoelectrochemical decolorization of Basic Blue 47 were optimized and investigated. Based on the obtained results in the absence of oxidant agents, the best index for decolorization in optimal conditions includes pH of 9.0, initial dye concentration of 303.3 μM, time of 93 min, and potential of 0.81 V corresponding to the design by the CCD was 98%. It was experimentally found to be 96.0 %, and the Chemical Oxygen Demand (COD) was 97 %.

Keywords: Sonoelectrochemical, Basic Blue 47, Free Chemicals, Degradation.

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1. INTRODUCTION

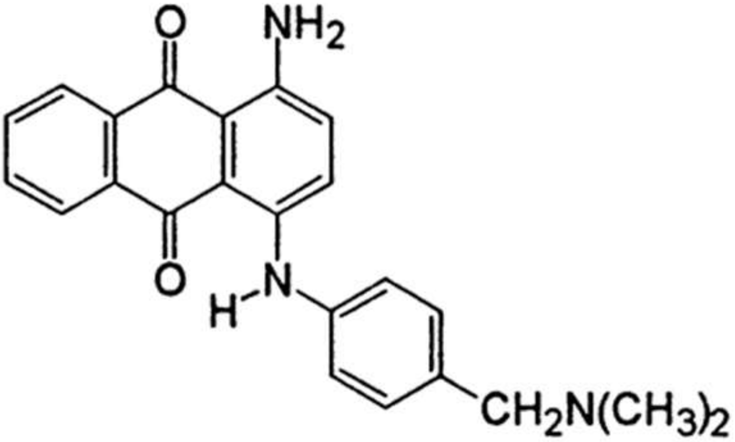
Textile industries are growing in developing countries. Textile industries are among the largest water-consuming and waste-producing industries [1-2]. In different stages of the textile industry, the dyeing process uses a large amount of water for dyeing, repairing, and washing purposes. The waste produced in textile industries originate from dyeing and supplementary processes and can be a significant threat to the environment. The wastewater with a COD above 1600 mg/L and a high dye concentration is classified as a very potential wastewater [3-4]. Around one million tons of dye are annually produced worldwide, while Azo and Anthraquinone dyes account for 65-75% of the total textile dyes. [5]. In addition, this wastewater contains a complex mixture of pollutants like heavy metals, additives, detergents, and surfactants. The release of these agents in wastewater makes the treatment process more difficult. The discharge of dye wastewater into the environment results in carcinogenicity and mutation due to the presence of toxic elements such as benzidine, naphthalene, and other undesirable aromatic compounds [6-8]. The removal of textile dyes from industry's wastewater before they are discharged into hydrologic systems is of paramount importance because of the complex structure, artificial origin, and the resistant nature of textile dyes [9-10]. There are various processes such as physical [11-12], chemical [13-14], and biological [15-16] methods for decolorizing the textile industry's wastewater. These methods face several limitations because of the presence of resistant organic chemicals in wastewater as well as problems related to costs, productivity, and sludge production. These restrictions can be overcome by advanced oxidation processes [17-21]. One of the most advanced oxidation processes is provided by sonoelectrochemical technology, which is used to decompose and treat dye solutions. In this process, hydroxyl radicals are released as a result of the cavitation phenomenon caused by ultrasonic vibrations inside the liquid at a high temperature and pressure and by the decomposition of compounds [22-23]. This attractive combinatory technology has received lots of attention owing to such advantages as adaptability to the environment, use of clean energy, recovery of heavy metals, treatment of highly toxic wastes, no generation of secondary pollution in the environment, and being more economical [24-25]. Therefore, the sonoelectrochemical decomposition of pollutants is a favorable method for treating industrial wastes particularly the wastes of textile industries [26, 27]. The aim of this study is to evaluate the efficiency of the sonoelectrochemical process in the decomposition of C.I. Basic Blue 47 from aqueous environments without hydrogen peroxide. In this research, the independent variables are solution pH, initial dye concentration, applied potential, and optimized time using the CCD method.

2. EXPERIMENTAL

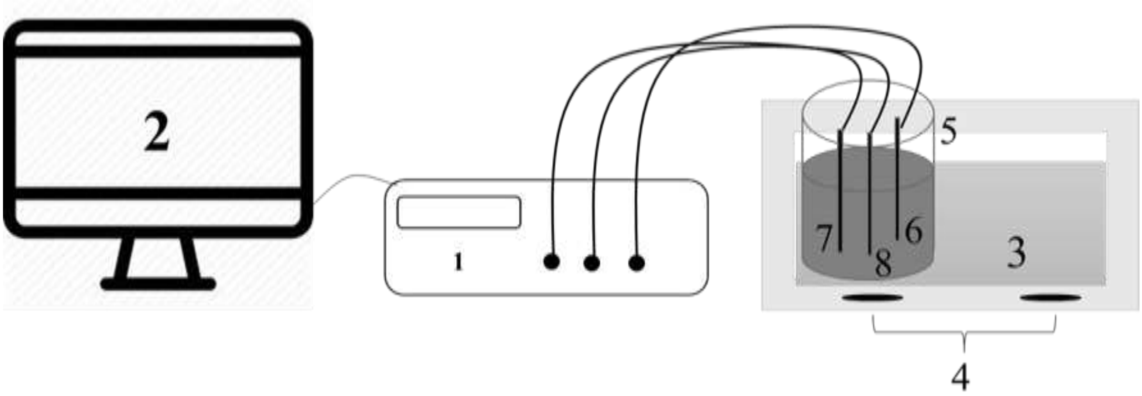
2.1 Materials and Methods

The C.I. Basic Blue 47 (BB47) dye (Scheme 1) was supplied by the Ciba Company and put to use with no purification. The dye has maximum absorbance (λ_{max}) at 617 nm. The instruments used in this study included a spectrophotometer (UV-Vis- Carry 100 made in Japan), a pH meter of model 876 made by Metrohm, Switzerland, a galvanostat-potentiostat device, a microAutolab from the Netherlands, three-electrode cells including carbon, platinum, and calomel electrodes made by Azar Electrode, Iran, and Euronda S.P.A model 4D (i.e. an ultrasonic generator) made by Vicenza, Italy.

The sonoelectrochemical tests were done in a non-separated electrolyte cell made from a Perix sheet. The working and auxiliary electrodes were placed in the cell vertically and parallel to each other at a distance of 2.5 cm. Carbon electrodes and platinum electrodes were used as working electrodes (anode) and counter electrodes (cathode) respectively. The saturated calomel electrode was used as a reference electrode. (See the setup used for decolorization experiments in Scheme 2).



Scheme 1. The Chemical structure of Basic Blue 47.



Scheme 2. Experimental sonoelectrochemical setup used for decolorization. (1) Electro analyzer apparatus; (2) Personal Computer (3) ultrasonic bath; (4) ultrasound Transducer; (5) Pyrex vessel; (6) working electrode (carbon); (7) reference electrode (saturated calomel); (8) auxiliary electrode (platinum plate).

In each experiment 100 mL of a BB47 solution was prepared with different initial concentrations. Various parameters such as the initial pH of the solution, the applied potential, the decolorization time, and the dye solution concentration were optimized simultaneously by the CCD method. At first, using the CCD software, as many as 30 tests were designed based on the four mentioned variables. Table 2 provides the corresponding data. Based on the conditions in Table 2, the decolorization tests were repeated four times, and then the averages from the dye solutions were determined without the presence of oxidants. Thereafter, the value of decolorization was calculated using Eq. (1):

$$\mathbf{Decolorization} = \frac{\mathbf{C_0 - C_t}}{\mathbf{C_0}} \times \mathbf{100} \qquad (\mathbf{1})$$

where C_t and C_0 are the concentrations of the dye at the time t and 0 (μM) respectively. It should be noted that the concentration of the remaining dye was calculated through measuring the absorption value at the maximum wavelength (λ_{max}=617 nm) using the spectrophotometer device and a calibration curve. The COD value was determined by the dichromate closed reflux method.

$$\mathbf{\%\ COD\ removal} = \frac{\mathbf{COD_0 - COD_t}}{\mathbf{COD_0}} \times \mathbf{100} \qquad (\mathbf{2})$$

where COD_0 and COD_t (in ppm) are the chemical oxygen demand at 0 (the initial time) and t (the reaction time) respectively.

Table 1. The conditions designed for achieving the optimal conditions using the CCD method

Run	**pH**	**[Dye] /μM**	**Time / min**	**Applied Potential / V**	**Decolorization%**
1	4.25	752.5	91.25	0.7125	**42**
2	8.75	752.5	91.25	0.3375	**46.46**
3	4.25	257.5	33.75	0.7125	**40**
4	6.5	505	62.5	0.525	**58**
5	6.5	505	120	0.525	**62**
6	8.75	257.5	91.25	0.7125	**63.06**
7	6.5	505	62.5	0.525	**54.2**
8	6.5	505	62.5	0.525	**53.6**
9	8.75	752.5	33.75	0.3375	**38.99**
10	6.5	505	62.5	0.9	**59.5**
11	6.5	10	62.5	0.525	**56.92**
12	4.25	752.5	91.25	0.3375	**40.12**
13	6.5	505	62.5	0.525	**52**
14	8.75	257.5	33.75	0.7125	**51.6**
15	4.25	257.5	91.25	0.3375	**60**
16	4.25	752.5	33.75	0.7125	**41.93**
17	8.75	752.5	33.75	0.7125	**40.33**
18	8.75	257.5	33.75	0.3375	**50.33**
19	4.25	257.5	33.75	0.3375	**38.8**
20	2	505	62.5	0.525	**35.21**
21	6.5	505	62.5	0.525	**52.65**
22	6.5	505	62.5	0.15	**53.14**
23	8.75	257.5	91.25	0.3375	**53.15**
24	4.25	752.5	33.75	0.3375	**32**
25	8.75	752.5	91.25	0.7125	**59.21**
26	6.5	1000	62.5	0.525	**54.95**
27	11	505	62.5	0.525	**75**
28	6.5	505	62.5	0.525	**51**
29	4.25	257.5	91.25	0.7125	**73**
30	6.50	505.0	5.0	0.53	**35**

2.2 Design of the Experiment

With regard to the design of the experiment and the optimization of the dye construction conditions using the sonoelectrochemical system, the simultaneous effects of different factors such as pH, the initial dye concentration, time and the applied potential on the decolorization of BB47 by the CCD and with the help of Design Expert 8.0.2 Software were analyzed. The range and levels of variables are provided in Table 1. Each of the four factors was calculated as 11 according to the alpha value. For this design and based on these four factors, 30 tests were designed. Table 1 provides the conditions of each test. To minimize the variables that were uncontrollable for any reason, the sequence of tests was chosen randomly.

Indeed, to resolve the problem of interaction of different variables and to reduce the cost required for the optimization of conditions for the dye removal, the CCD method was selected. In general, the overall aim of the test design was to determine the effective factors influencing the reaction and their interactions with the minimum number of tests, thereby reporting the best result for achieving the maximum impact factor corresponding with statistical procedures [28].

Table 2. ANOVA results of RSM models provided for decolorization of BB47

Source	**Sum of squares**	**Df**	**Means square**	**F value**	**p-value Prob>F**
Model	3802.64	14	271.62	17.58	<0.0001*
A-pH	1.13	1	1.13	0.073	0.073
B-Dye	1.94	1	1.94	0.13	0.7280
C-time	861.13	1	861.13	55.37	<0.0001
D-Potential	170.67	1	170.67	11.04	0.0046
AB	31.92	1	31.92	2.07	0.1712
AC	29.59	1	29.59	1.92	0.1866
BC	72.00	1	72.00	4.66	0.0475
A^2	2.23	1	2.23	0.14	70.94
B^2	0.16	1	0.16	0.010	0.9213
C^2	392.58	1	392.58	25.42	0.0001
ABC	210.83	1	210.83	13.64	0.0022
A^2B	136.76	1	136.76	8.85	0.0094
B^2C	82.64	1	82.64	5.35	0.0354
A^3	319.71	1	319.71	20.69	0.0004
Residual	231.79	15	15.45		
Lack of fit	201.85	10	20.19	3.37	0.0960**
Pure error	29.94	5	5.45		
Cor total	4034.43	29			

* Significant and ** Not significant. The model F-VALUE of 17.58 implies the model is significant. There is only a 0.01% chance that the “model F-VALUE” this large could occur due to noise.

3. RESULTS AND DISCUSSION

3.1. Mathematical Model for Prediction of Decomposition of BB47

The regression model of the coded factors for the performance of the decomposition of BB47 is indicated in Eq. (3), obtained by the CCD method where the coded values of A, B, C, and D are the variables of pH, the dye solution concentration, the time, and the applied potential respectively.

$$\textbf{\textit{Decolorization}}\ \% = +53.26 - 0.38A - 0.49\,B + 10.37\,C + 2.67\,D + 1.41\,AB - 1.36\,AC - 2.12\,AD - 0.28\,A^2 - 0.75\,B^2 - 3.75\,C^2 + 3.63\,ABC - 5.06\,A^2B - 3.94\,B^2C + 2.58\,A^3 \tag{3}$$

ANOVA is required for the significance test and the validity of the model provided in Table 1. For ANOVA, an F-value lower than 0.0001 implies significance. According to Table 1, an F-value equal to 17.58 demonstrates that the model is significant. The ability of this model was evaluated by the correlation coefficient of $R^2=0.999$, suggesting that over 99.9% of the sample change was in accordance with the presented model and had an only 0.1% deviation from the total. A value lower than the coefficient of variation (CV%=7.81) implies that the tests are accurate and reliable. “Adeq Precision” measures the signal-to-noise ratio. A ratio larger than 19.607 is desirable.

3.2. The Effect of Independent Variables

Figure 1 illustrates the simultaneous effect of pH and the applied potentials on the decomposition of BB47, using 384.4 μM BB47 and within 71.8 minutes. As it can be seen, as the pH increases from 2.0 to 11.0, the decolorization percentage also improves. Based on the result, it can be suggested that as the solution pH increases, the hydroxyl group (OH^-) concentration grows gradually. In an alkaline pH, the main anion is OH^- which converts into $OH^\bullet$ radicals by losing an electron at the anode surface. On the other hand, the presence of ultrasonic waves enhances the migration of hydroxyl anions towards the anode [29]. Therefore, once the pH rises, the number of hydroxyl radicals is increased as the main factor in the dye decomposition.

Figure 1 also shows that when the applied potential increases from 0.15 to 0.71 V, the decolorization rises with fast slope and then continues by gentle slope. It seems that an increase in the voltage is directly related to the further production of hydroxyl radicals. Indeed, the potential and the number of free radicals are tightly correlated [29].

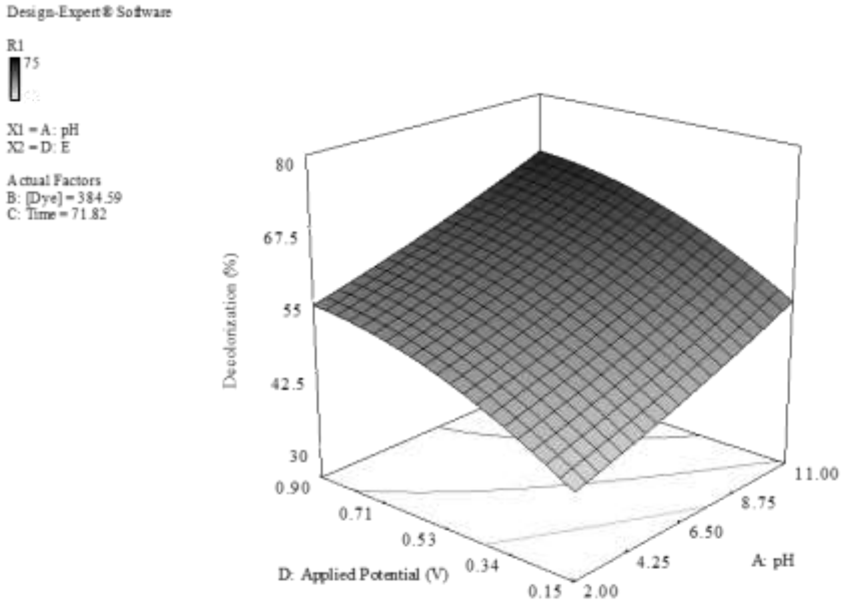


Figure 1. The 3D diagram showing the effect of pH and various potentials on decolorization value at the constant dye concentration of 505 μM and the contact time of 62.5 minutes

Figure 2 shows the simultaneous effect of initial BB47 concentration and time on the decolorization percentage. As it can be observed, within the constant pH of 8.0 and the applied potential of 0.8 V, when dye concentration decreases from 10.0 μM to 675.0 μM, the decolorization percentage shows a rising trend and decreases in higher dye concentration. Based on the literature, it is evident due to the rise of the dye solution concentration, the number of effective contacts between dye molecules and electrode surface as well as exploded bubbles, which due to contact of water with the ultrasonic waves are generated and grew. On the other hand, when the number of dye molecules further increases, the consumption of hydroxyl radicals by dye molecules becomes more than free radical production. The reduction in the number of free radicals in proportion to the number of dye molecules diminishes the system efficiency for dye decomposition purposes [29, 30].

As Figure 2 illustrates, once time increases from 5.0 to 120.0 min, the decolorization percentage is also enhanced. It seems that with an increase in time, free radicals such as •OH and HOO• will be increased, which promotes the discoloration rate [22, 29]. Also, in our case, more dye molecules can move to the electrode surface and oxidize on it.

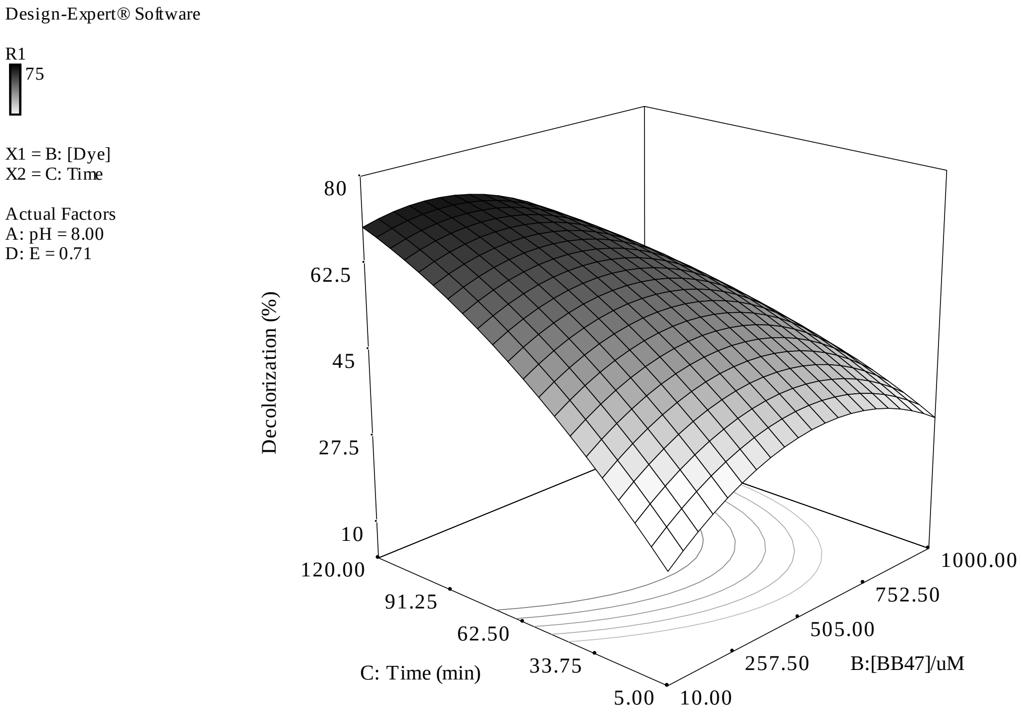


Figure 2. The three-dimensional diagram showing the effect of time and dye concentration on decolorization value at pH=8.0 and at the voltage of 0.81 V

3.3. UV/Visible Absorption Spectra OF BB47 Solution

The change in the UV/Visible absorption spectra for BB47, as a function of the time of the sonoelectrochemical degradation, was recorded (Fig. 3), using the applied potential of 0.81 V, and pH of 9.0 for 90 minutes at intervals of 10 minutes.

As it can be seen, one main peak in the UV region (260 nm) and a band in the visible region (650 nm) were observed at the absorption spectra of BB47 (Fig. 3). The disappearance of the visible band was probably due to the fragmentation of the anthraquinone structure by oxidation, and the change of absorbance in the UV region was considered evidence of aromatic fragment degradation in dye molecules.

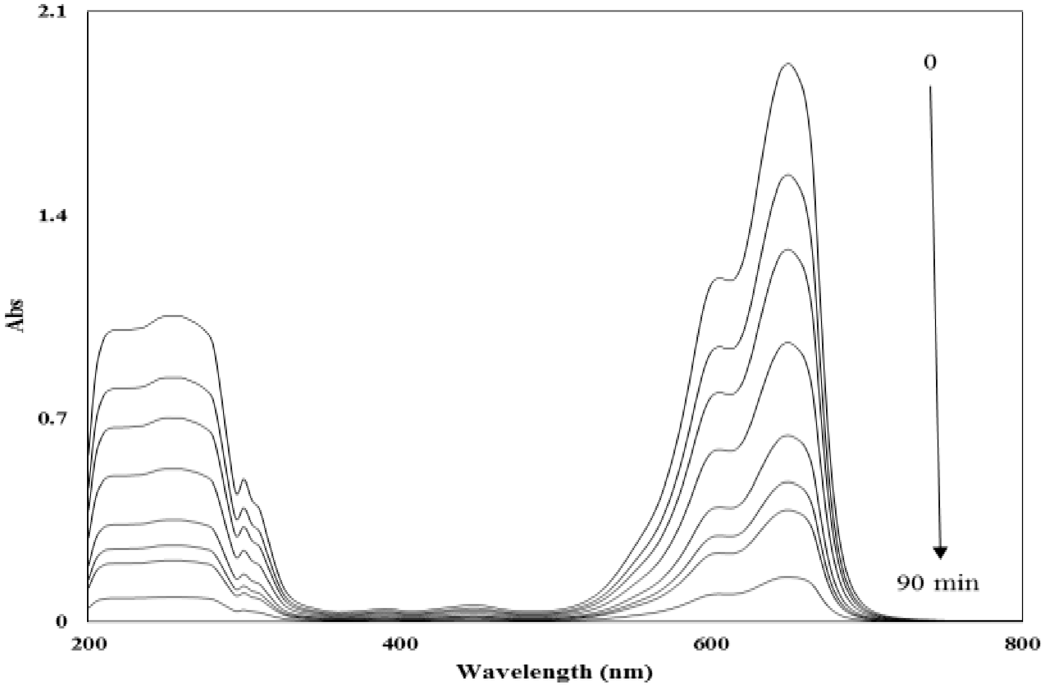


Figure 3. UV–vis spectra of aqueous solution containing BB47 during sonoelectrochemical process (pH = 9.0, 303.0 μM BB47, applied potential = 0.81 V).

3.4. Decolorization of BB47 with Different Systems

A specific series of experiments was designed under optimum conditions for the decolorization of BB47 using different methods. The experiments were performed using sonoelectrochemistry, electrochemistry, and sonochemistry process. Table 3 shows the decolorization efficiency of BB47 in the mentioned processes. As compared to other techniques, the sonoelectrochemical process resulted in a higher removal percentage (96 %).

Sonoelectrochemical oxidation for decolorization of BB47 (96 %) was more efficient than electrochemical oxidation (23 %) and sonochemical oxidation (46.6 %) after 93 min. This may be due to the general improvement in the hydrodynamics and movement of species toward the surface of electrodes [9, 22].

The results also indicate that decolorization of BB47 occurs at a higher rate through the sonoelectrochemical method in comparison to other methods. In the first case, a decolorization efficiency of 96% was observed in 93 minutes under optimum conditions, whereas in the electrochemistry during the same time, only a decolorization efficiency of 23 % was recorded. However, a decolorization efficiency of 46.6 % was achieved by sonochemistry. This decolorization can be due to the synergetic effect of ultrasound in electrochemical treatment. Similar results were gained by Radi *et al.* (2015) in the decolorization of Reactive Blue 49 using ultrasound-assisted electrochemical oxidation. Their results confirmed that ultrasound improves the production of free radicals, resulting in the generation of additional hydroxyl radicals to react with the substrate. In addition, ultrasonic wave irradiation can eliminate the impurity layer at the surface of the electrode and increase its activity, as well as increase the reaction rates for mass transfer [22].

It should be noted that in these experiments has not been used for decolorization of any oxidant chemical and only is used from electric power as a renewable and biocompatible resource. Finally, sonoelectrochemical degradation was found to be efficient in decolorizing simulated textile effluents. The results show that ultrasound was significantly enhanced in the electrochemical oxidation.

3.5. Energy Consumption

The consumption of Energy (EC (kW h m−3)) is an essential factor for estimating the feasibility of the electrochemical process for the removal of pollutants. It should be clear that the energy consumption was considered for the particular setup used, without any concerns about a design that can lead to low operational cost. It is defined as the required energy of electrochemical oxidation for the removal of 1.0 g of BB47 and can be calculated from EC=UIt/1000V. Here, U is the applied potential (V), I is the applied current (A), t is the time of electro-oxidation (h), and V is the volume of the working solution (m^3). In general, the best results were found for the ultrasound-assisted electrochemical process, considering the constant current density (15 mA cm−2) and applied voltage (0.81 V). The longer the time, the higher will be EC for decolorization treatment.

3.6. The Combination *of Ultrasound-Electrochemical Process in COD Removal*

An optimal formulation for decolorization of BB47 was determined to minimize the chemical oxygen demand (COD). To determine the reduction of COD of the solution containing BB47, COD (before treatment) and final COD (after sonoelectrochemical oxidation) of the aqueous dye solutions were measured. Thus, a considerable percentage of COD removal proved to have occurred. Under optimum operating conditions, a COD reduction of 96% was achieved. The above combination is considered to have an important role in reducing and eliminating COD. The optimization results are shown in Table 3 in the order of desirability.

Table 3. The comparison of different methods for decolorization of BB47 under optimal conditions.

Method	**[Dye] (μM)**	**Time (min)**	**pH**	**Applied Voltage (V)**	**Decolorization (%)**	**EC (kW h m^3)**
Sonoelectrochemistry	303.3	93	9.0	0.81	96	0.25

Electrochemistry	303.3	93	9.0	0.81	23	8.28*
Sonochemistry	303.3	93	9.0	-	46.6	-

* A long time requires for only 75 % decolorization of BB47.

4. CONCLUSION

The results of the present study show that sonoelectrochemical and optimized use of CCD can be efficiently made to decolorize and degrade BB47. Based on the achieved model, each of the parameters is altered within an allowable range so that the optimal response, i.e. the maximum value of decolorization, is achieved. It was observed that an increase in the initial pH, the applied potential, and the duration of the sonoelectrochemical process would enhance the percentage of dye degradation. It is concluded that the percentage of dye degradation decreases with an increase in the initial concentration of the dye molecules. It was found that optimal conditions would include a pH of 9.0, dye concentration of 303.3 μM, duration of 93 minutes, and potential of 0.81 V. The decolorization percentage predicted by the model was 92.8% in these conditions, while it was found to be 92.34% after the experimental test. A high decolorization efficiency of more than 92% was also achieved without an oxidizing agent, which would damage the environment. The closeness of these responses is indicative of the appropriacy of the presented model. According to these results, the sonoelectrochemical process can serve as a valuable alternative to conventional physicochemical methods for the treatment of wastewater.

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