Decolourization of Disperse Blue 3 Dye by Electro Coagulation Process Using Al and Fe Electrodes –Application of the Artificial Neural Network Model

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ABSTRACT: *Contamination in wastewater is a major issue in the present world, Disperse blue 3 dye (DB_3) removal was studied by an electrocoagulation process using Al and Fe electrodes. The experiments were performed with synthetic solutions in batch mode. The effect of the operating parameters like the electrolysis time, current density, initial pH, conductivity, inter-electrode distance, and initial dye concentration, has been investigated. The results show high discoloration efficiency, reaching 98 and 96% with Al and Fe electrodes respectively. The optimum condition of the EC process was electrolysis times of 70 and 30 min, current densities of 139 and 93 mA/cm², initial pH 5, the conductivity of 5.67 mS/cm, and inter-electrode distance of 1.5 cm. The Artificial Neural Network (ANN) technique was used to model the experimental data of the current density. The feed-forward neural network model was optimized by using the Levenberg-Marquardt algorithms. A comparison between the predicted and experimental data gave high correlation coefficients (0.99977 and 1) with the minimum MSE value ($1.55.10^{-7}$ and $1.31.10^{-5}$) respectively for Al and Fe electrodes.*

KEYWORDS: *Electro-coagulation; Disperse blue 3; Al and Fe electrodes; Artificial neural Network.*

INTRODUCTION

Industrial wastewaters represent a heavy source of environmental pollution. It affects both the water quality as well as the microbial and aquatic flora. With competing demands on limited water resources, the awareness of issues involved in water pollution has led to a considerable public debate about the environmental effects of the industrial effluents discharged into the aquatic medium.

This industrial discharge, including the textile effluents, is a cause of human illnesses. About 40% of textile dyes contain organically bound chlorine, which is a known carcinogen. Chemicals evaporate into the air and get absorbed by our skin and cause allergic reactions in children before birth.

Due to this chemical pollution, the normal functioning

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of cells is interrupted. This may cause an alteration in the physiology and biochemical mechanisms of animals, resulting in the impairment of important functions like respiration, osmoregulation, reproduction, and even mortality [1]. However, due to the toxic nature and harmful effect of synthetic dyes on all forms of life, interest in natural dyes has resumed worldwide. Nevertheless, even natural dyes are rarely low-impact.

In practice, dyes are classified into different application classes: disperse, acid, basic, direct, vat, fiber reactive, sulfur, In accordance with US EPA, by using general dye chemistry as the basis for classification, textile dyestuffs are grouped into 14 categories or classes: (1) acid dyes, (2) direct (substantive) dyes, (3) azoic dyes, (4)disperse dyes, (5) sulfur dyes, (6) fiber reactive dyes, (7) basic dyes, (8) oxidation dyes, (9)mordant (chrome) dyes,(10) developed dyes, (11) vat dyes,(12) pigment, (13) Optical/Florescent Brightener and (14) Solvent Dyes [2].

For convenience, all the above dyes can be grouped into three categories namely Days for cellulose fibers, Days for protein fibers, and Dyes for synthetic fibers. In this logic, this work addresses the dispersed dye (Disperse blue 3).

Recently, different methods were used for the treatment of wastewater contaminated by dyes, including biological, physical, and chemical methods such as adsorption [3-4], oxidation [5], and electro-coagulation [6], combined processes such as electro-coagulation/ adsorption [7] and flocculation [8]. One of the methods developed to overcome the drawbacks of conventional treatments of water and wastewater is Electro-Coagulation (EC). However, Avsar et al. stated that the main disadvantage of the conventional EC consists of the formation of an impermeable oxide film on the cathode [9], resulting in high energy consumption and low efficiencies [9]. This prevents the effective current transfer between the anode and cathode so that the performance of the EC reactor decreases. One of the suggested solutions consists of the polarity changing of electrodes resulting in the so-called ―self-cleaning‖ of electrodes. The use of the alternating current in EC systems delays the passivation of cathode and anode deterioration, phenomena met in direct current systems, and thus, ensures a reasonable lifetime of electrodes [10].

The electrochemical technique in which particles generated by the anode combine to free active coagulation factors in the solution. Metal ions are generated in the anodic compartment while hydrogen concomitantly evolved at the cathode [11]. Hydrogen can help mass particles float and come out of water [12]. The resulting metal ions, *i.e.* Al^{3+}(ag) and Fe^{3+}(ag), undergo some reaction to produce hydroxides or polyhydroxide, which strongly absorb dispersed compound molecules, causing coagulation [13].

The present paper is devoted to the removal of Disperse blue 3 (DB_3: Molecular formula: $C_{17}H_{16}N_2O_3$) by electrocoagulation using Al and Fe electrodes. The effect of the current density, electrocoagulation time, electrode distance, pH, and conductivity on the electrocoagulation efficiency is discussed. All these parameters are important in designing large-scale plants for the industrial application of EC. On the other hand, the experimental data of the variation of the current density during the elimination of DB_3 by electrocoagulation were modeled with the Artificial Neural Network approach (ANN).

EXPERIMENTAL SECTION

Materials

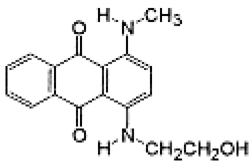
Disperse blue 3 was purchased from Boufarik textile plant (Algeria) whose main characteristics are given in Table1. The DB_3 solution was freshly prepared before experimental runs, by dissolving accurate amounts in ultra-purified water.

Electrocoagulation process

Typically, EC equipment consists of DC-powered cathode and anode electrodes, which are partially submerged in a tank that contains a contaminated solution. These electrodes can vary in shape, size, and number, but rectangular-shaped plates are often used [14]. The most common metals used to manufacture EC electrodes are Al and Fe because they are cost-effective, widely available, and non-hazardous[14-15]; the experimental setup is illustrated in Fig. 1. The anode and cathode (effective electrode area: 21.5 cm²) were positioned vertically and fixed at a distance of 1.5 cm. electrodes were two Al or Fe plates are connected to digital DC power supply (P.S-A 305D, 0-30V; 0-3A; DAZHENG).

The electrodes were polished with fine-grained emery paper and were dipped in sulphuric acid to remove the rust and other impurities which were then cleaned using a scrub and finally rinsed with distilled water and dried before

Table 1: main characteristics of Disperse blue 3.

Name	Molecular formula	Structure formula	Molecular weight	CAS Number	Synonyms
DB_3	$C_{17}H_{16}N_2O_3$		296.32 g/mol	2475-46-9	Acetate Brilliant Blue 4B * Acetoquinone Light Pure Blue R * Altocyl Brilliant Blue B * Amacel Blue BNN *. Cibacet Blue F3R

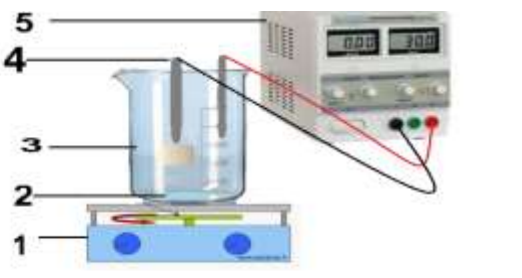


Fig. 1: A schematic diagram of the electrocoagulation reactor (1-agitator 2-magnetic stir bar 3-EC reactor 4-électrode 5-DC power supply).

using it for the experiments, all the runs were performed at room temperature (24 ± 2 °C) [16]), Additionally, the process's conductivity was controlled by adding NaCl, needs to be mentioned that sodium chloride was applied as the electrolytes because chlorides are widely present in various industrial wastewater with high concentrations [17-18]. The experiments were carried out in batch mode. In each run, the model wastewater solution (1 l, 50 mg/L of DB_3) was placed in the electrochemical cell. During the runs, the reactor unit was continuously stirred (200 rpm) by a magnetic stirrer. Coagulation has started once the current density has been set to the desired value. The pH and conductivity of the solution were measured by C861 consort multi-parameter analyzer. The process had a duration of 70 min of Al and 30 min of Fe, and 5 mL of this solution were collected every 5 min with a syringe (the process did not need to stop). At the end of the electro-coagulation, the sample was left to settle by centrifugation (5000 rpm, 10 min), and then analyzed. The DB_3 concentration was determined at λ_{max}= 639 nm using a Jenway 6305.UV/VIS spectrophotometer.

The color removal efficiency Y (%) was calculated from the relation:

$$Y(\%) = \left({}^{(C0 - Ct)}/_{C0}\right) \times 100 \quad (1)$$

Where C_o is the initial dye concentration before electrocoagulation (mg/L) and C_t is the concentration after t_{min}.

Artificial Neural Networks (ANNs)

Artificial neural networks (ANNs) are among the main tools used in machine learning. As the 'neural' part of their name suggests, they are brain-inspired systems that are intended to replicate the way that we humans learn. ANNs consist of input and output layers, as well as (in most cases) a hidden layer consisting of units that transform the input into something that the output layer can use. They are excellent tools for finding patterns that are far too complex or numerous for a human programmer to extract and teach the machine to recognize.

ANNs can be used as an alternative to the polynomial regression-based modeling tool, which provides the modeling of complex nonlinear relationships. The ANN model is potentially more accurate by including all experimental data [19]. Furthermore, various industrial applications of ANNs exist, e.g., modeling and identification, optimization, and process control.

RESULTS AND DISCUSSION

Effect of time of electrolysis on the efficiency of DB_3 removal

The reaction time influences the treatment efficiency of the electrolytic process. During electrolysis, the anodic electro-dissolution led to the release of coagulating species. The DB_3 removal efficiency depends directly on the concentration of metal ions produced on the electrodes. This is due to the fact that with an increase in retention time, all coagulated species settle down easily to give clear supernatant liquid and sludge. But providing a retention time more than the optimum retention time results in the reduction of pollutant removal efficiency as the adsorbed pollutant desorbs back into the solution [20-21]. So, the effect of time of electrolysis was studied

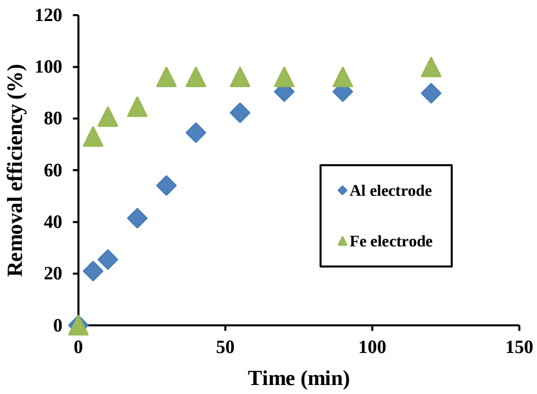


Fig. 2: Variation of DB_3 removal efficiency with time: initial concentrations= 50 mg/l. Distance inter-electrode = 1.5 cm, conductivity = 5.67 mS/cm, initial pHi= 5.44, and current density (139 mA/cm^2 of Al, 93 mA/cm^2 of Fe).

at a constant current density of 139 mA/cm² and initial pH 5.44 (Fig. 2); an increase in the electrolysis time from 5 to 90 min. yields an increase in the dye removal efficiency from 21 to 90% respectively, at the Al electrode. In the case of the Fe electrode, the removal efficiency increases from 67 to 96% in the interval (5 – 30 min.).

According to *Shokri* [22] the electrocoagulation process removal percent of Acid orange 5 versus time was improved from 36.5 to 99.3%, with an increase in electrolysis time from 10 to 60 min. at the optimum condition, the removal of COD (85.5%) was lower than the removal of Acid Orange 5 (99.3%). The differences between the removal of COD and Dye were low because of the less formation of intermediate products, at a constant current density of 2.0 mA/cm^2 and initial pH of 7.

Effect of current density on the efficiency of Disperse blue 3 removal

The magnitude of current density determines the amount of anode metal ions that dissolve during electrolysis and the production rate of bubbles at the cathode, therefore, the current density is among the main factors that affect electrochemical treatment [23]; to investigate its effect on the Disperse blue 3 elimination, a set of experiments were conducted in solutions containing constant dye concentration with a current density ranging from 2.32 to 139 mA/cm². The pH and the conductivity of the solution as well as the distance between electrodes were kept constant. (Fig. 3) illustrates the effect of current density on the Disperse blue 3 removal efficiencies at Al and Fe electrodes.

The reaction at the Al electrode decreases from 90% for a current of 139 mA /cm² down to 22% at 2.32 mA/cm² after 70 min. while at Fe electrode increases the removal efficiency up to 96% at 139 mA /cm² and 65% at 2.32 mA/cm² after 30 min. It can be analyzed that the current density has fewer effects on the final total DB_3. While the dissolving rate of Al and Fe electrodes increases at high densities. This is due to oxidation to Al^{3+} and Fe^{3+} which passes to the wastewater at higher current densities, thus promoting the formation rate of $Al(OH)_3$ and $Fe(OH)_3$ owing to the solubility products K_s, 10^{-32} and 10^{-38} respectively, In the electrocoagulation processes, the reaction rate is controlled by the current [24-25] by controlling hydroxide flocs formation. *Mohajeri et al.* reported that they used the electrical current as 0–3 A from the experimental conditions [26]. The current density determines the coagulant dose rate, bubble production rate, and flocs growth, which can influence the treatment efficiency of the electrocoagulation. In addition, it was demonstrated that the bubbles' amount increases while their size decreases.

In all experiments presented below, the current densities of 139 and 93 mA/cm^2 are considered optimal in the present work for Al and Fe electrodes respectively. The results show that increasing current density increases the DB_3 removal due to the increasing rate of anodic dissolution.

Effect of initial pH:

Initial pH plays a fundamental role in the performance of EC process and its change during the treatment is linked to the process efficiency [27]. It determines the ionic character of the metal hydroxides and DB_3 molecules in solution and in this way it will have a significant influence on the removal mechanism. In order to investigate the pH effect on the Disperse blue 3 elimination, the tests were performed by adjusting the initial pH in the interval (1 – 12) at the optimal conditions (electrodes distance 1.5 cm, current density 139 mA/cm² for Al: 70 min., 93 mA/cm² for Fe: 30 min.). The pH effect on electrocoagulation is illustrated in (Fig. 4).

The results with the Al electrode showed that when pH of DB_3 solutions was set between 5 and 12, the DB_3 removal efficiency peaks at pH 5. On the other hand, pH of Disperse blue 3 for acidic zones, corresponding to pH low than 3, a significant drop in removal efficiency was observed, according to *Mouedhen et al.* [28] reported that in pH

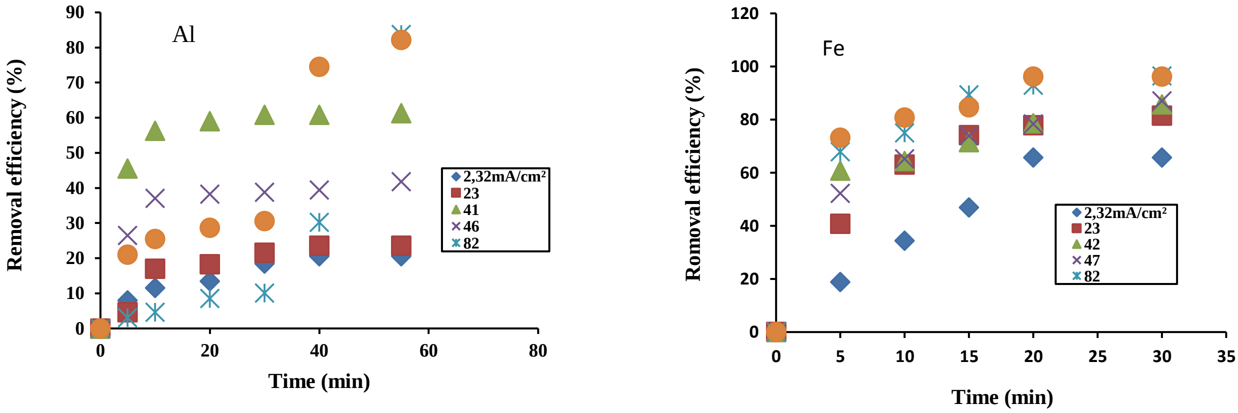


Fig. 3: Effect of the current density on the removal efficiency of Disperse blue 3; with Al and Fe electrodes: concentration = 50 mg/L. Distance inter-electrode = 1.5 cm, conductivity= 5.67 mS/cm, pHi= 5.44.; electrolysis time 70 min (Al) and 30 min (Fe).

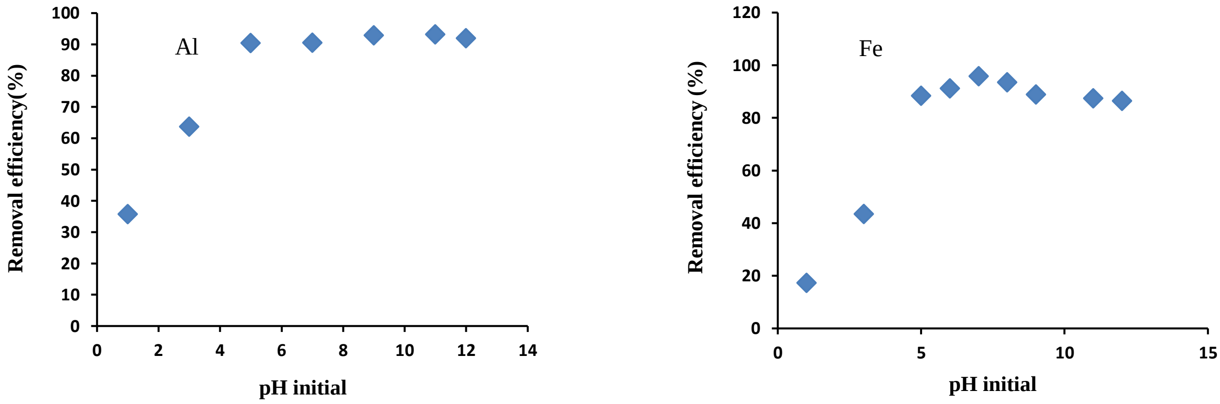


Fig. 4: Effect of initial pH on the removal efficiency of Disperse blue 3 with Al electrode (139 mA/cm^2, 70 min.) and Fe electrode (93 mA/cm^2, 30 min.). Concentration = 50 mg/l. Distance inter-electrode = 1.5 cm, conductivity= 5.67 mS/cm.

lower than 3, the released amount of Al during the electrolysis with constant charge per volume was significantly lower than in pH above 3. Acidic pH seemed to inhibit or decrease the dissolution rate of cathodes depending on the pH. For using an iron electrode when pH of dye solutions was between 6 and 9, the dye removal efficiency was optimal with a maximum pH 5.

Effect of conductivity

The effect of wastewater conductivity on Disperse blue 3 removal experiments was performed by adjusting the initial conductivity of 2.40 to 13.7mS/cm, under the following conditions: 70 and 30 of treatment time and a distance of 1.5 cm between the electrodes 5,5 initial pH, 139 and 93mA/cm² current density values of Al and Fe electrodes respectively. (Fig.5) shows the effect of conductivity on Disperse blue 3 removals with the Al and Fe electrode the removal efficiency rates are above 95% in all conductivity values of the Al electrode and 85% of the Fe electrode. While conductivity increased from 2.40 mS/cm to 13.7 mS/cm, and the voltage decreased from 32.1 Volt to 15.1 Volt, color removal capacity did not change much due to conductivity. This can be attributed to the reduction of voltage and anodic oxidation at high conductivity [29].

Effect of the interelectrode distance

Many authors have studied the effect of distance between the electrodes on pollutant removal. It may be concluded from these works, the evolution of the electrocoagulation efficiency as a function of interelectrode distance depends on the nature of the pollutant, electrode setup, hydrodynamic conditions, etc [30]. In the present study, the effect of interelectrode distance is investigated between 0.5 and 4 cm. The results are depicted in (Fig. 6).

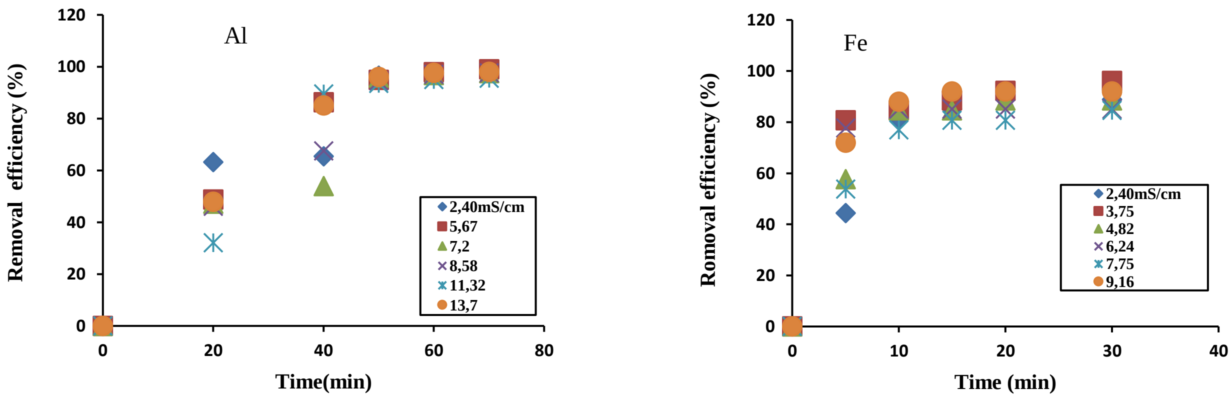


Fig. 5: Effect of conductivity on the removal efficiency of Disperse blue 3 with Al electrode (139 mA/cm^2, 70 min.) and Fe electrode (93 mA/cm^2, 30 min.); concentration= 50 mg/l. Distance inter electrode = 1.5 cm.

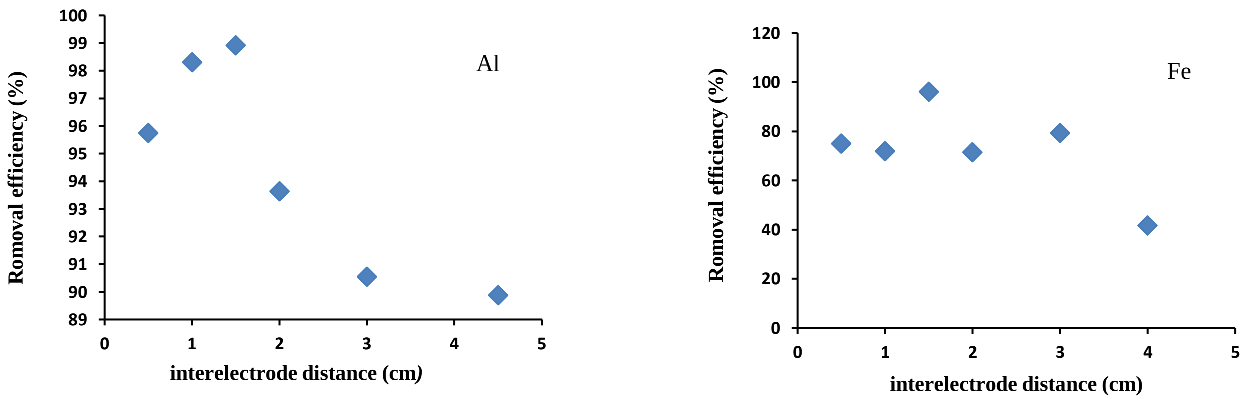


Fig. 6: Effect of inter-electrode distance on the removal efficiency of Disperse blue 3 with Al electrode (139 mA/cm^2, 70 min.) and Fe electrode (93 mA/cm^2, 30 min.); concentration= 50 mg/l. Conductivity= 5.67 mS/cm, initial pHi= 5.

It is observed that the best efficiencies were obtained with an interelectrode distance of 1.5 cm with DB_3 removal efficiency of 98% (Al) and 96% (Fe). In previous studies in the literature, the distance between electrodes is generally between 1 and 2 cm [31-32]. For the interelectrode distance of 0.5, 1, and 2, the dye removal efficiencies are 96, 97, and 94% (Al) and 75, 71, and 71% (Fe). At high distances, the DB_3 molecule and flocs interactions become weaker, leading to a decrease in the removal efficiency. The decrease in values was observed due to the decrease in IR drop which in turn lowered the production of electrocoagulation floc residue into the solution [33].

Effect of the DB_3 initial dye concentration:

The DB_3 solution with an initial concentration in the range (C_o: 50 – 900 mg/l), were treated by electrocoagulation using Al (139 mA/cm², 70 min) and Fe (93 mA/cm², 30 min.). (Fig. 7) shows that the removal efficiency decreases with raising the initial dye concentration C_o. After 70 min of operation, the efficiency decreases from 98 to 80% for Al, and from 95 to 87% for Fe when dye concentration augments from 50 to 700 mg/l. It is observed that the lower of DB_3 concentration, the better the decolorization efficiency. It was due to the insufficient hydroxyl and metal ions produced on the electrodes in the high dye concentrations and the constant current density [34].

The experimental data of the variation of the current density during the DB_3 removal by electro coagulation were modeled with the approach of Artificial Neural Network (ANN).

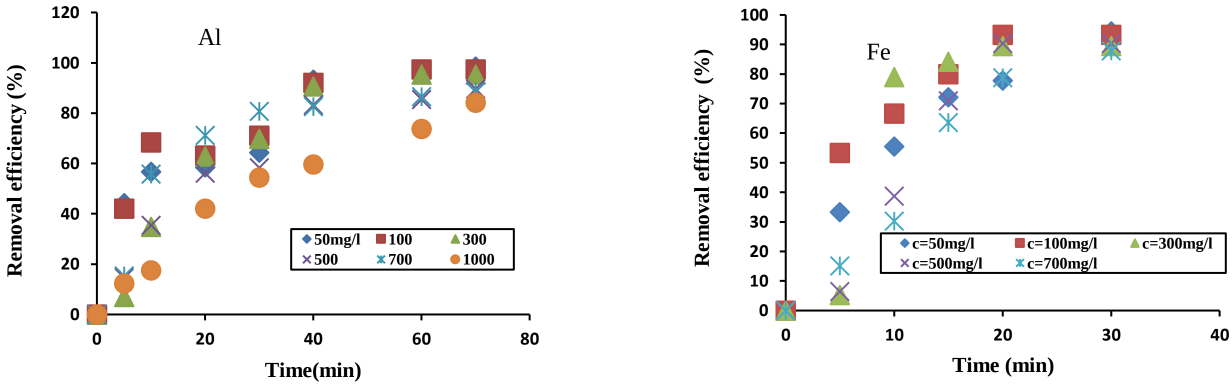


Fig. 7: Effect of initial dye concentration on the removal efficiency of Disperse blue 3 with Al electrode (139 mA/cm^2, 70 min.) and Fe electrode (93 mA/cm^2, 30 min. conductivity= 5.67 mS/cm, initial pHi=5). Distance inter-electrode = 1.5 cm.

ANN modeling

The neural models have been implemented using the programming package MATLAB mathematical (version 7.0, 2011b).

The backpropagation network employed in this modeling consisted of an input layer with four operational parameters including the current density, pH, voltage, and conductivity for times of 70 min and 30 min of Al and Fe electrodes respectively. ANN is determined by the number of layers, the number of neurons in each layer, and the nature of the transfer functions. The optimization of ANN topology is probably the most important step in the development of a model [35-37]. In order to determine the optimum number of hidden nodes, a series of topologies was used, in which the number of nodes varied from 2 to 10. The best result was obtained with 6 neurons in the hidden layer and a number of layers equaling two. The transfer function used for the input and the hidden layer is the log sigmoid transfer function given by:

$$f(x) = \frac{1}{1 - e^{-x}} \quad (2)$$

The function log sig produces outputs between 0 and 1 as the node's net input goes from negative to positive infinity. To determine the best back propagation training algorithm, 5 BP algorithms were tested; Table 2, shows a comparison of different BP training algorithms for Al and Fe electrodes respectively.

The first step of ANN modeling was to optimize a neural network with the aim of obtaining an ANN mode with a minimal dimension and minimal errors in training and testing, the maximum value of the correlation coefficient (R) of the testing set according to the Eqs.

$$MSE = \frac{\sum_{i=1}^{i=N} \left(y_{p,i} - y_{exp,i}\right)^2}{N} \quad (3)$$

$$R = \sqrt{1 - \frac{\sum_{i=1}^{i=N} \left(y_{p,i} - y_{exp,i}\right)^2}{\sum_{i=1}^{i=N} \left(y_{p,i} - y_m\right)^2}} \quad (4)$$

Where N is the number of data points, $y_{p,i}$ the network prediction, $y_{exp,i}$ the experimental response y_m the average of the experimental value, and i a data index.

Model selection

In this study, DB_3 was optimized by a (4-6-1-1) configuration, the training parameters used for the simulation of the sample ANN model are: TRAINLM, MSE, LOGSIG these are the training, performance, and transfer functions respectively in the simulation. The performance of all training algorithms and the maximum value of the correlation coefficient (R) of the testing was analyzed and the training algorithm train LM (Levenberg–Marquardt backpropagation) are found most suitable for the estimation of outputs that have the least mean square.

It was reported that the coefficients R obtained with ANN (Fig. 8) are 0.99977, 0.98313, and 1 for the test, training, and validation respectively, and minimal errors in training and testing is 1.55e-07. With Fe electrode the correlation obtained and the best validation performance (Fig. 8) are 1, 0.9958, and 1 for the test training, and validation respectively, and a minimal error of 1.3×10^{-5}. These results showed the best efficacy of the ANN methodology in the DB_3 elimination.

Table 2: Comparison of 5 backpropagation algorithms with 6 neurons in the hidden layer with the number of layers equal to two final current densities of Al and Fe electrodes at the time of 70min and 30 min respectively.

backpropagation (BP) algorithms	Algorithm code		Al and Fe electrodes					
			Epoch	MSE	R			Best linear equation of test
					Test	Training	Validation	
Conjugate Gradient with Powell/Beale Restarts	Train CGB	Fe	51	0.0987	1	0.99168	1	Output=0.83×target+14
		Al	50	0.206	0.99651	0.99769	0.99794	Output=0.96*target+3.1
Fletcher-Powell Conjugate Gradient	Train CGF	Fe	50	0.0692	1	0.99057	1	Output=1.5×target ±38
		Al	50	0.0719	0.99949	0.99792	0.99946	Output=0.94×target+4.7
Levenberg -Marquardt	TrainLM	Fe	50	1.31e-05	1	0.9958	1	Output=1×target+0.38
		Al	50	1.55e-07	0.99977	0.98313	1	Output=0.99×target+1.3
One Step Secant	Train OSS	Fe	51	0.0469	1	0.99142	1	Output=2×target±74
		Al	50	1.38	0.99973	0.99593	0.9966	Output=0.71×target+11
Scaled Conjugate Gradient	Train SCG	Fe	50	0.0443	1	0.99527	1	Output=1.3×target±20
		Al	58	0.212	0.99986	0.99568	0.8609	Output=1×target±3.6

CONCLUSIONS

In the present work, the removal of Disperse blue 3 from aqueous solutions by electrocoagulation with Al and Fe electrodes has been reported. The effects of the operation parameters like the electro-coagulation time, current density, initial pH, salt concentration, the distance between electrodes, and initial dye concentration. The results of Disperse blue 3 showed that the removal efficiency was enhanced from 22 to 90% by increasing the time from 5 to 70 min and from 67 to 96% of the time 5 to 60 min for Al and Fe respectively.

The electrolysis time is an important operation variable for dye elimination treatment efficiency. The electro-coagulation is strongly dependent on the solution pH with poor efficiency at low pH. The optimal value is revealed at pH 5. The optimal inter-electrode distance was 1.5 cm while the current density exhibits a strong effect on the dye removal. Higher efficiency was obtained at 139 and 93 mA/cm^2 for Al and Fe respectively. The Artificial Neural Network was applied to these parameters (current density, pH, voltage, conductivity, and time) to agree with the experimental data with a higher correlation coefficient for testing and the lowest MSE; beyond a threshold, no remarkable improvement may be achieved. The modeling results confirmed that the Artificial Neural Network could effectively reproduce experimental data and predict the behavior of the process.

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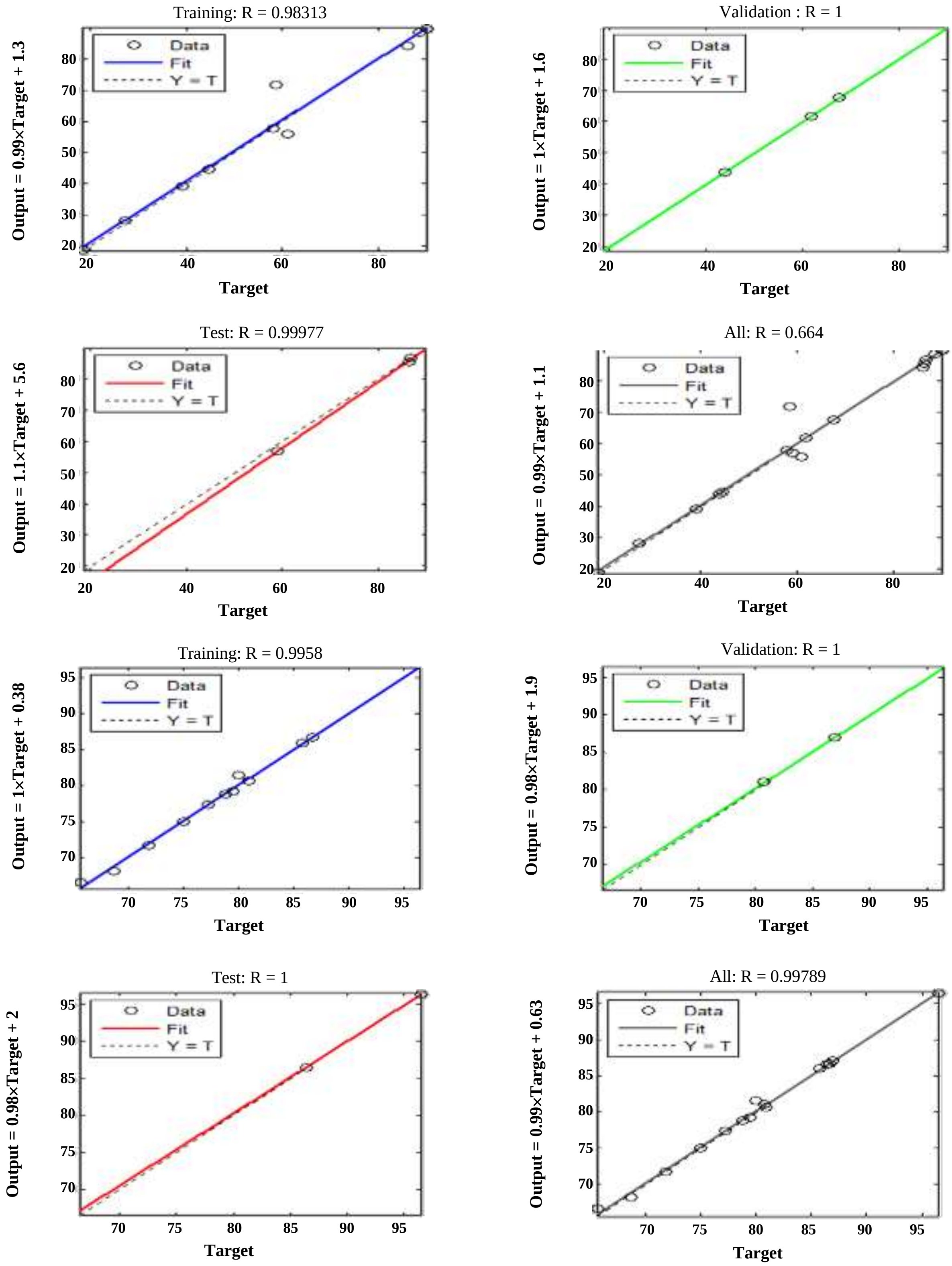

Fig. 8: The regression plots between the ANN estimations and experimental data base for Training, Validation, Test, and all data of Disperse blue 3 removals with Al and Fe electrode.

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