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Removal of Reactive Black 5 from Polluted Solutions by Electrocoagulation: Modelling Experimental Data Using Artificial Neural Networks

Bediha OYAR¹, Beytullah EREN^{*2}, Abdil ÖZDEMİR³

Abstract

The wide range of today's industry increases the diversity of pollutants in the wastewater characteristics. In particular, the wastewater of the textile industry is highly colored. Different techniques are used for color removal of dyes from wastewater. In this work, the removal efficiency of the textile dye (Reactive Black 5) at different current densities (48.5 A/m², 97.18 A/m², 194.36 A/m², 291.5 A/m², 388.7 A/m²) was investigated by electrocoagulation method. The dye concentration of wastewater prepared in the laboratory scale was adjusted to 100 mg/L. Two iron electrodes and 3 g NaCl were used in the electrocoagulation system. The samples which taken periodically were measured after the centrifugal processes with the UV spectrophotometer. The experimental results were also modelled with artificial neural networks (ANNs). As a result of the experiments, approximately 90-100% color removal efficiency was obtained. According to the modelling study, the ANNs can predict the color removal efficiency with coefficient of determination (R²) between the experimental and predicted output variable reached up to 0.99.

Keywords: Wastewater, electrocoagulation, textile dye (reactive black 5 (RB5)), color, artificial neural networks (ANNs).

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1. INTROCUCTION

Water is one of the most important elements among naturel resources[1]. Water consumption has increased as a result of population growth and technology development. As a result of this, the amount of wastewater has also increased and so the receiving environments have been exposed to more pollutants. Polluting of the existing water resources has an important place among environmental problems. Large amounts of wastewater are produced every year from domestic and industrial areas, and this global problem continues to grow day by day[2]. The characteristic of wastewater varies according to the source of wastewater. Industrial wastewater such as textile, food production, cosmetics, paper production has a large share in the color problem that occurs in the receiving environments due to the synthetic dyes[3–5]. Textile industry is especially known as a one of the quickly growing industry which uses a large amount water in the production process[3]. Textile wastewater is contained with considerable amount of pollutant and high level color due to organic, inorganic chemicals and dye which were used in production process[6]. Approximately 50.000 tons of dye are discharged by textile industry into receiving environments every year[7]. This type of wastewater is regarded as harmful for receiving environments due to its toxicity and high organic components[8]. Removal of color in wastewater is highly difficult due to their resistance to light and oxidation[9]. However, the color in textile wastewater causes a decrease in dissolved oxygen concentration in receiving environments[10]. Due to all these adverse effects, such wastewater must be treated before discharge to the receiving environment and the pollutant concentrations must be reduced below the limit values specified in the regulations[11].Removal of color from wastewater is crucial for human health and sustainable water quality. In the literature, it was seen that many physical, chemical, biological and advanced treatment methods such as adsorption, biosorption, chemical oxidation, photolysis, chemical coagulation, flotation, ion exchange, electro dialysis, ozonation, membrane filtration were used in color removal from wastewater[12–21]. However, many of these methods require

high investment and operating costs and also have various operational difficulties. In recent years, the interest in electrochemical methods has been increased considerably and is also preferred for color removal from wastewater. Electrochemical methods are frequently preferred due to the fact that they do not require chemical substances before and after wastewater treatment, produce less sludge, low space requirements, low initial investment cost and time savings[6,22].

Electroflotation, electroflocculation and electrocoagulation are some applications of electrochemical treatment methods[23]. In the literature, the electrocoagulation method, especially dye[6,11,19], selenium[24], heavy metal[25,26], polycyclic aromatic hydrocarbons (PAH)[27], phosphate[28], nitrogen[29], chemical oxygen demand (COD), oil and grease[30], drug[31] are used with high removal efficiency in the treatment of pollutants. The electrocoagulation method is also widely used because it is easily integrated into the treatment units and that simple equipment is sufficient for the treatment process.[32,33].

There are various parameters affecting the pollutant removal efficiency from wastewater by electrocoagulation method. The most important parameters effecting electrocoagulation process are pH, electrolyte concentration, current density, pollutant concentration, electrolysis time, electrode type.

Reactive dyestuffs especially used in textile industry cause black color formation in wastewater and degrade the water quality of the receiving environments. In this study, the effect of different current densities on the removal efficiency of Reactive Black 5 (RB5) by electrocoagulation was investigated. And also, the color removal efficiency was estimated by modeling the obtained experimental results with artificial neural networks (ANNs).

2. MATERIAL AND METHOD

2.1. ELECTROCOAGULATION

Electrocoagulation (EC) is a method developed to improve traditional methods in wastewater treatment[34]. This technology is a relatively low cost, easy to operate, environmentally friendly technology that requires a minimum of chemicals and produces a small amount of sludge. This method is a simultaneous operation, involving multiple chemical mechanisms[35].

The electrocoagulation process is considered to consist of 3 basic stages[7]. These stages are

- 1- Coagulant formation by oxidation of electrode (such as Al, Fe),
- 2- Stabilization of pollutants, particle suspension and emulsion breakage,
- 3- Collection of flocks in unstable phases[36].

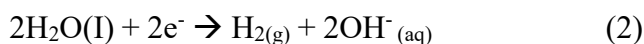
Generally, direct current anode and cathode electrodes are preferred in EC process. Electrodes can vary in shape, size, number and material[37]. Various electrode types such as stainless steel, iron, aluminum, boron doped diamond, titanium coating are used as electrode material in the EC process[13,38,39]. However, it is seen that iron and aluminum electrodes are generally used due to the low cost and yield obtained[8,40–42].

If iron electrode is used in EC process, the following reactions occur[35]:

Anode:



Cathode:



In electrocoagulation method, one of the important parameters affecting removal efficiency and treatment cost in color removal from wastewaters containing dye is current density. According to Faraday, the amount of cations dissolved from the anode is directly

related to the current density passing through the wastewater[8,43].

It is observed in studies that the pollutant removal increases as the current density increases[43–46]. The mechanism of electrocoagulation is shown in Figure 1 below.

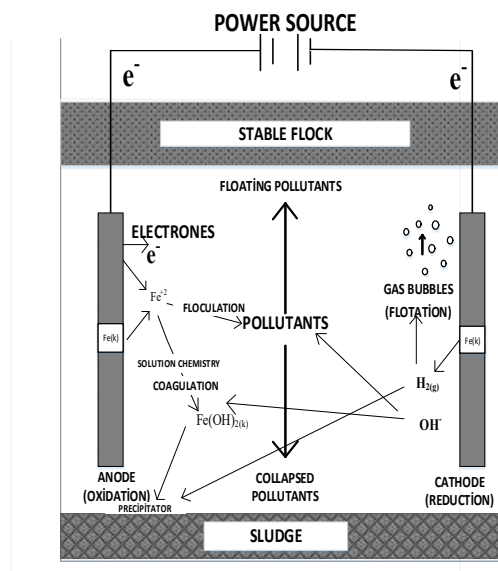


Figure 1. Mechanism of electrocoagulation[47]

2.2. EXPERIMENTAL MATERIALS

In this study, the removal of color (Reactive Black 5 (RB5) dye) at different current densities is investigated by using electrocoagulation method. The chemical bond structure of RB5 is shown in Figure 2. The solution used in experimental studies and containing dyestuff was prepared in the laboratory using ultra-pure water. For the calibration procedure, a calibration curve was created using 5 different concentrations of solution, 1 mg/L, 5 mg/L, 10 mg/L, 15 mg/L and 20 mg/L, and these is shown in Figure 3. Also, UV spectrum of absorption that is prepared for calibration is shown in Figure 4.

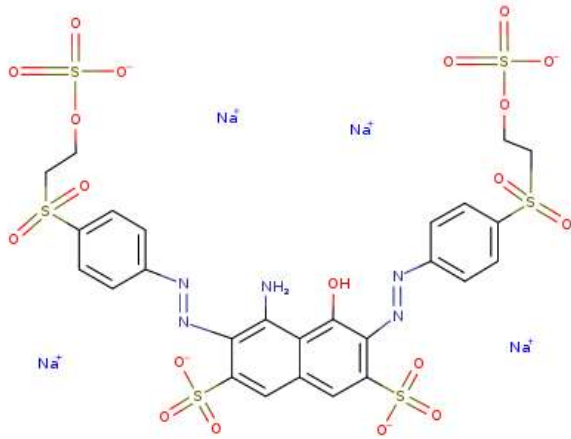


Figure 2. Chemical bond structure of Reactive Black 5

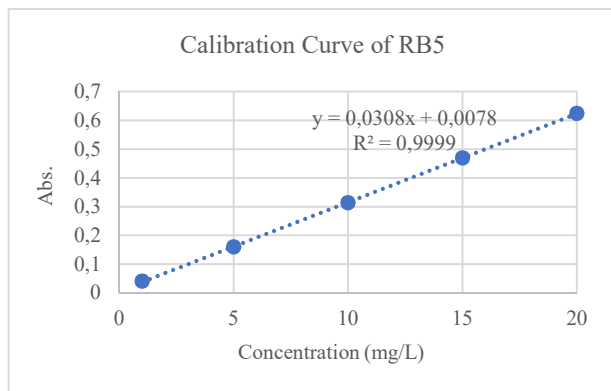


Figure 3. Calibration curve of RB5

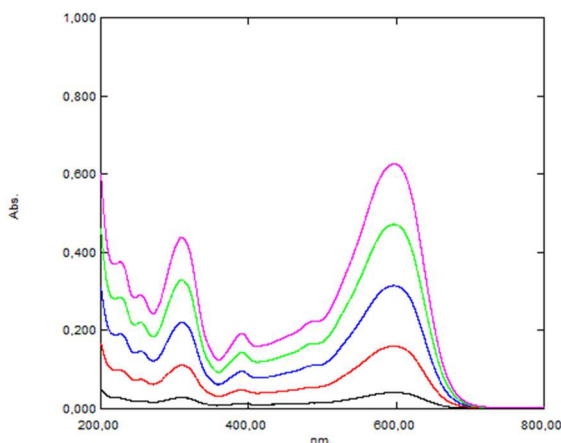


Figure 4. UV spectrum of RB5

2.3. EXPERIMENTAL METHOD

Electrocoagulation experimental setup used in this study was seen in Figure 5. The experimental setup consists of a 2000 ml glass beaker, two iron

electrodes, a direct current power supply and a magnetic stirrer. The distance between the electrodes used in the experiments is 2 cm and the active surface area is 10.9 x 9.8 cm². In the experiment, firstly 100 mg/L RB5 dye concentration was prepared in glass beaker. Secondly, 3 g of NaCl was added to dye solution as electrolyte. Finally, the current densities of 48.5 A/m², 97.18 A/m², 194.36 A/m², 291.5 A/m², 388.7 A/m², respectively, were provided to contact the solution for 60 minutes for each experiment. The working environment is acidic because the RB5 dye has a natural pH of 5.8 and no pH adjustment is made to the waste water sample[48].

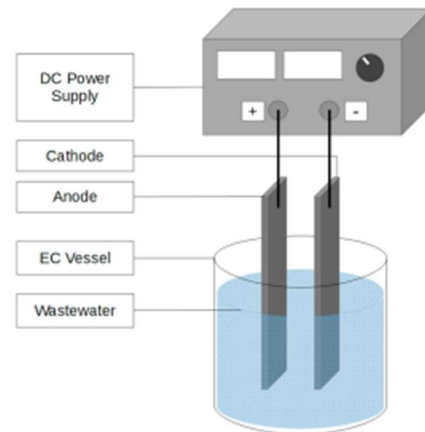


Figure 5. Electrocoagulation experimental setup[37]

5 ml samples were taken at 2, 4, 6, 8, 10, 12, 14, 16, 18, 20, 25, 30, 35, 45, 60 minutes in all current densities (48.5 A/m², 97.18 A/m², 194.36 A/m², 291.5 A/m², 388.7 A/m²). After the samples were centrifuged, the measurement of all samples, including calibration, was performed at UV-Vis recording spectrophotometer at intervals of 0.5 nm between 200-800 nm. The maximum absorbance value for Reactive Black 5 was found to be 600 nm as a result of the studies.

The removal efficiency of color was calculated using to below formula.

$$C_{\text{color}}(\%) = \frac{C_0 - C}{C_0} \times 100 \quad (3)$$

$C_{\text{color}}(\%)$: The removal efficiency of color (%)

C_0 : Initial dye concentration

C : Final dye concentration

2.4. COST ANALYSIS

Electricity consumption is taken into account while calculating the cost of color removal from waste water with electrocoagulation process. In measurement price of unit was accepted as 0,008 \$/kWh for the electricity consumption[49].

Amount of electricity consumption was calculated using equation below[33].

$$C_{\text{Energy}} = \frac{U \times I \times t}{v} \tag{4}$$

C_{Energy} : Consumption of electricity (kWh/m³)

U : Volt

I : Current

t : Electrolysis time (hour)

v : Solution volume (m³)

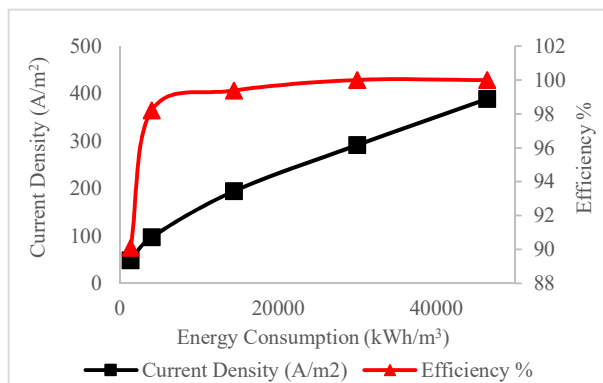


Figure 6. Effect of current density on consumption of electrical energy

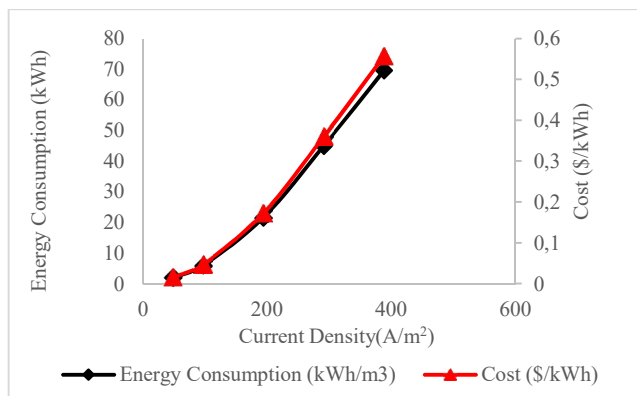


Figure 7. The relationship between energy consumption and cost

2.5. ARTIFICIAL NEURAL NETWORKS

In recent years, interest in modeling has increased in the process of estimating the status and optimum performance related to the treatment process in wastewater treatment plants. Artificial intelligence-based methods are often preferred in order to determine optimum performance in several disciplines covering water resources and environmental science. Artificial neural networks (ANNs) are the most remarkable modeling method among artificial intelligence-based methods. When compared with traditional mathematical methods, ANNs have advantages such as the lack of necessity to make mathematical definitions of events in the process, having the ability to predict using a limited number of experiments [50].

The nervous system of the human body has inspired the creation of ANNs, and these systems are important artificial intelligence systems capable of solving a number of complex problems[40,51–56].

ANNs have three layers including the input layer, the hidden layer(s) and the output layer.

In artificial neural networks, the input layer consists of independent variables, while the output layer consists of dependent variables. Between these two layers are hidden layers containing one or more neurons in layers[57]. ANNs create many connections between process elements to solve certain problems in harmony[34]. Schematic structure of the ANNs are showed in Figure 8.

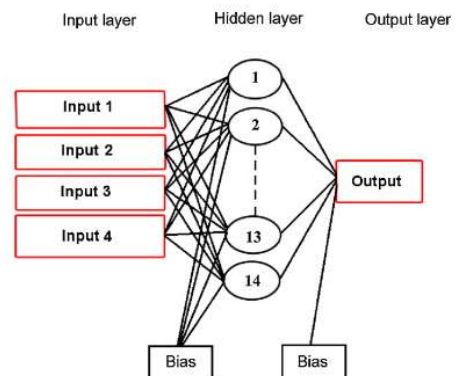


Figure 8. Structure of artificial neural network[58]

The electrocoagulation method provides high efficiency in pollutant removal from wastewater, as well as the fact that it contains complex chemical events throughout the EC process, raises the idea of modeling. Recently, Artificial neural networks (ANNs) successfully applied to solve many environmental engineering problems. Modeling of complex EC systems with ANNs make it a promising modeling technique [57,59].

3. RESULT AND DISCUSSION

3.1. Determination of the effect of current density on removal efficiency of RB5 dye

There are several operating parameters that is important in electrochemical processes. Electrode properties and current density is in among these parameters. The current density can be defined as

the amount of current passing through the unit electrode area. In this study, 5 different current densities were applied to find the most suitable current density in the removal of RB5 dye by electrocoagulation process. Experimental studies were carried out using an iron electrode. Current density that effect on electricity consumption was decided as 48.5 A/m², 97.18 A/m², 194.36 A/m², 291.5 A/m², 388.7 A/m². During experiment current density was adjusted by direct current power supply.

The effect of current density on removal efficiency of color is shown in Figure 9.

In Figure 9, It is seen that when the current density is 48.5 A/m², 90% removal efficiency is obtained in the 20th minute, and when the current density is 388.7 A/m², the same removal efficiency is reached in the 4th minute.

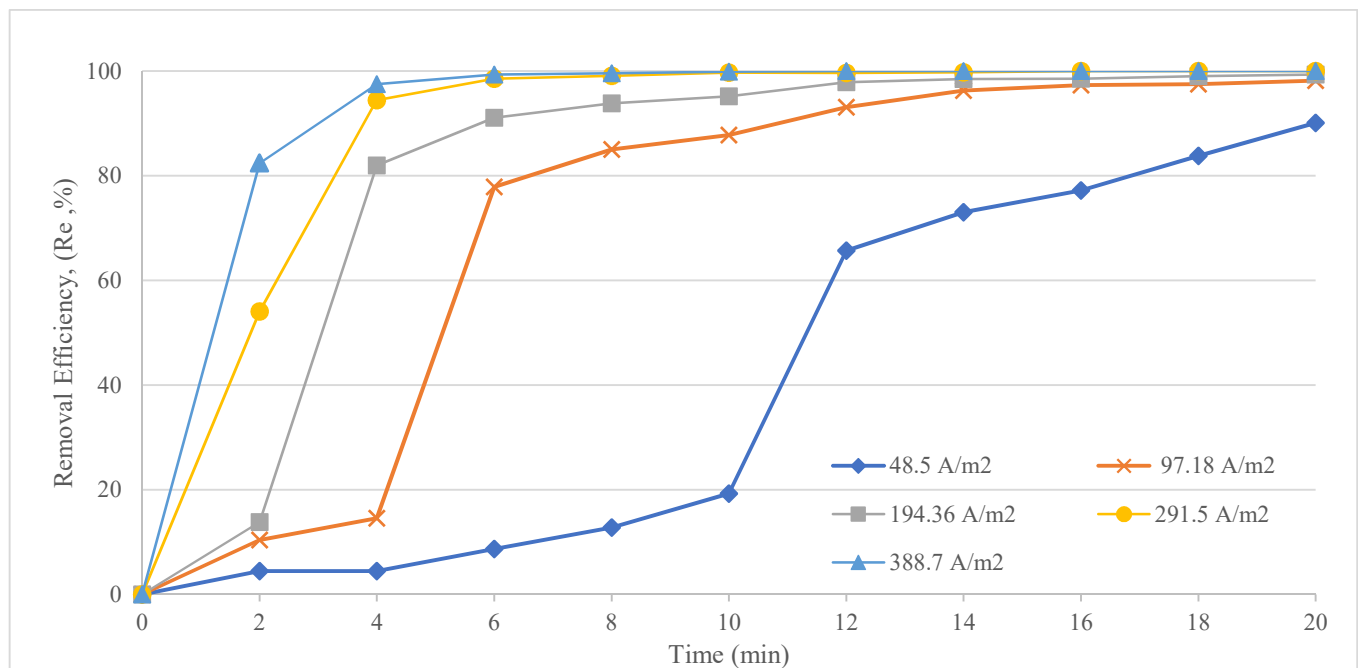


Figure 9. The effect of current density on removal efficiency of color (Re, %)

3.2. ANN modeling result of removal efficiency of RB5 dye

A total of 7 ANNs model was derived from a combination of 6 different topologies (LR: linear regression; MLP: multilayer perceptron; PNN: probabilistic neural network; RBF: radial basis

function; GFF: generalized feed forward; MLPPCA: MLP with principal component analysis), up to two hidden layers (1 vs. 2), two learning algorithms (M: Momentum vs. L: Levenberg-Marquardt) and two learning modes (B: batch vs. O: online), and (N: None). In ANN modeling, experimental dataset was randomly

partitioned into training, cross validation and testing datasets with the allocation order of 75%, 10% and 15%, respectively. Datasets were fed into a total of 7 ANNs to predict removal efficiency of color (Re, %) which included as inputs time (T, min) and current density (CD, A). Performance statistics used for comparing ANNs were mean square error (MSE), coefficient of correlation (R²) and mean absolute error (MAE) as follow[60,61]:

$$MSE = \frac{1}{N} \sum_{i=1}^N (y_i - \hat{y})^2 \dots\dots\dots(5)$$

$$MAE = \frac{1}{N} \sum_{i=1}^N |y_i - \hat{y}| \dots\dots\dots(6)$$

$$R^2 = 1 - \frac{\sum (y_i - \hat{y})^2}{\sum (y_i - \bar{y})^2} \dots\dots\dots(7)$$

Where,

\hat{Y} : Predicted value of y

\bar{Y} : Mean value of y

Table 1. A comparison of 7 artificial neural networks (ANNs) developed as a function of time (T, min), Current Density (C, A), by Removal Efficiency (Re, %) of Reactive Black 5 based on performance metrics of training, cross-validation and independent validation.

Artificial Neural Networks Model	Training (n=26)			Cross Validation (n=5)			Testing (n=4)		
	MSE	R ²	MAE	MSE	R ²	MAE	MSE	R ²	MAE
LR-0-B-L (Linear Regression)	0.04452	0.89928	0.18346	0.07320	0.85752	0.22640	0.02877	0.97111	0.12602
MLP-1-B-L (Multilayer Perceptron)	0.00044	0.99931	0.01407	0.00931	0.98353	0.06069	0.00544	0.99674	0.05332
PNN-0-N-N (Probabilistic Neural Network)	0.05441	0.93956	0.18459	0.11850	0.87575	0.27209	0.0432	0.99803	0.14108
RBF-1-B-L (Radial Basis Function)	0.01072	0.97962	0.068955	0.08618	0.96882	0.20784	0.00212	0.99985	0.03287
GFF-1-B-L (Generalized Feedforward)	0.00103	0.99830	0.02412	0.00774	0.98780	0.07783	0.00433	0.99791	0.04745
MLPPCA-1-B-L (MLP with PCA)	0.02966	0.95300	0.13308	0.07704	0.81495	0.17828	0.01095	0.98969	0.07554
<i>MLP-2-B-L (Multilayer Perceptron)*</i>	<i>0.00028</i>	<i>0.99963</i>	<i>0.00937</i>	<i>0.00135</i>	<i>0.99734</i>	<i>0.03112</i>	<i>0.00352</i>	<i>0.99969</i>	<i>0.03609</i>

*The best performing ANN is designated in italic.

Explanations of ANN Model Topologies: LR: linear regression; MLP: multilayer perceptron; PNN: probabilistic neural network; RBF: radial basis function; GFF: generalized feed forward; MLPPCA: MLP with principal component analysis), up to two hidden layers (1 vs. 2), two learning algorithms (M: Momentum vs. L: Levenberg-Marquardt) and two learning modes (B: batch vs. O: online), and (N:None)

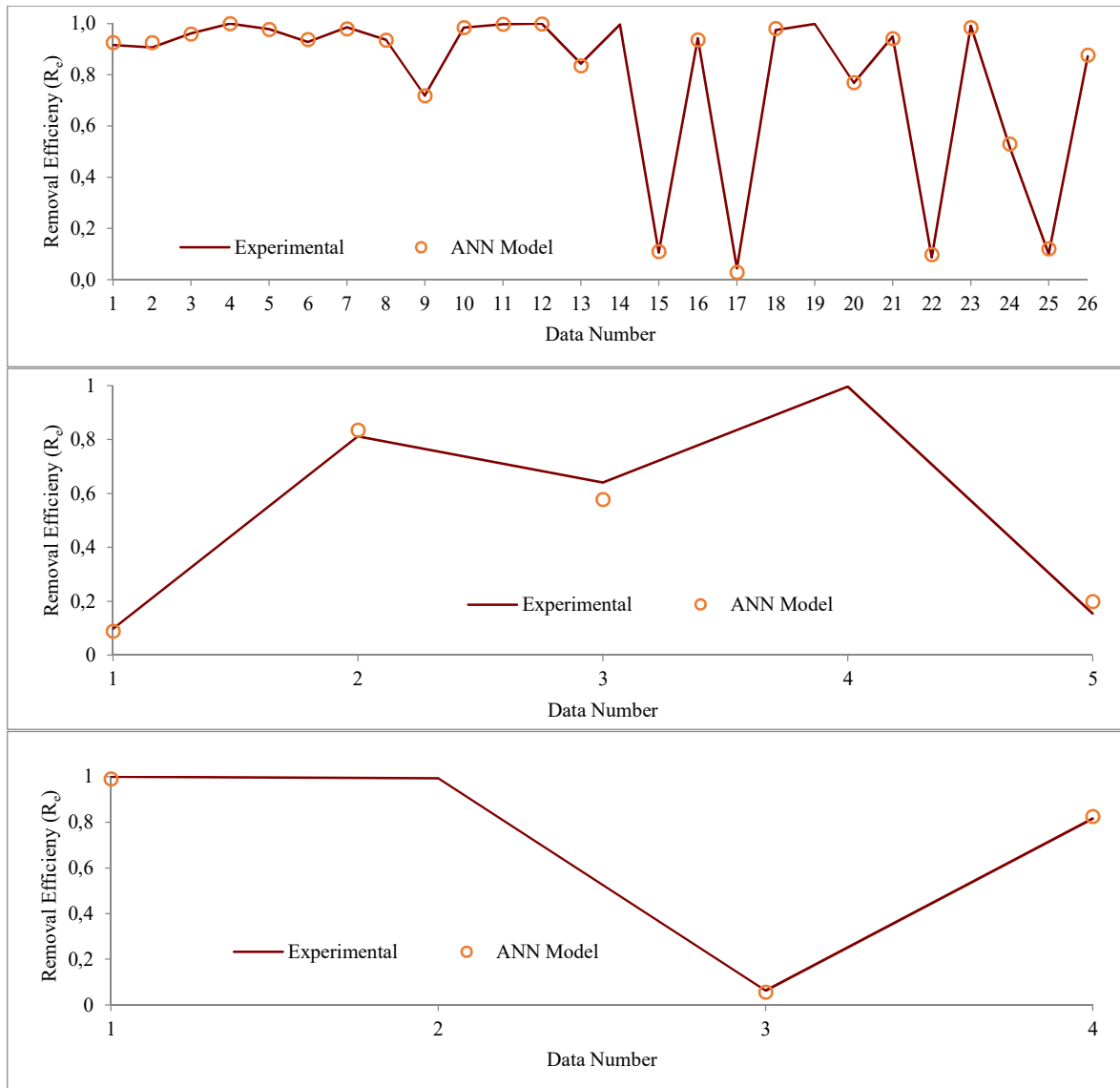


Figure 10. The comparison of Experimental and ANN model results by the best performing neural networks (MLP-2-B-L) for training, cross validation and testing. Solid and discrete lines refer to experimental and ANN model predicted values of Removal Efficiency (R_e), respectively.

The performance statistics derived from training, cross validation and testing of the 7 ANNs indicated that MLP-2-B-L performed best among the other ANNs as a function of Time and Current Density (Table 1). The predictive performance of the ANNs in terms of R^2 values based on testing ranged from 0.97111 by LR-0-B-L to 0.99969 by MLP-2-B-L. Testing-derived values of R^2 , RMSE and MAE pointed to MLP topologies (MLP-2-B-L) as the best ANNs.

4. CONCLUSION

In this study, the removal efficiency of color due to Reactive Black 5 dye at different current densities using electrocoagulation method from aqueous solutions was investigated. The aqueous solution prepared in the laboratory scale was analyzed using a UV spectrophotometer and the color removal efficiencies were calculated. It has been observed that color removal increases with increasing current density. It has been calculated that the removal efficiency after 20 min experimental period, 90,09 %, 98,21%, 99,38%,

100%, 100% is achieved for 48.5 A/m², 97.18 A/m², 194.36 A/m², 291.5 A/m² and 388.7 A/m² current densities, respectively. It is seen that the color removal efficiency increases with increasing current density. However, the electricity cost increases with the increasing of current density. Therefore, the optimum current density was determined as 97.18 A/m².

Also, the experimental results are modeled by artificial neural networks (ANNs). According to ANN modelling, it is seen that R² between experimental and predicted output variable varies in the range of 0.89-0.99. The performance statistics derived from training, cross validation and testing of the 7 different ANN models indicated that MLP-2-B-L (as in section 3.2) performed best among the other ANNs as a function of Time and Current Density. These results show that developed ANN model has good predictive ability for the removal efficiency of color.

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Research and Publication Ethics

This paper has been prepared within the scope of international research and publication ethics.

Ethics Committee Approval

This paper does not require any ethics committee permission or special permission.

Conflict of Interests

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this paper.

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