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MODELING OF MALACHITE GREEN ADSORPTION ONTO AMBERLITE IRC-748 AND DIAION CR-11 COMMERCIAL RESINS BY ARTIFICIAL NEURAL NETWORK

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Highlights

- Amberlite IRC-748 and Diaion CR-11 are effective adsorbent materials for malachite green removal.
- The ANN model is developed using a three-layer feed forward back propagation network.
- Adsorbent dosage, initial malachite green concentration and contact time are studied for modeling.
- The effects of the training algorithm, the transfer function in the hidden layer and the number of hidden neurons on model accuracy were investigated.

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ABSTRACT: In this study, the malachite green adsorption process using Amberlite IRC-748 and Diaion CR-11 resins was modelled by artificial neural network method. In the model created for this study, adsorbent dosage, initial malachite green concentration and contact time parameters, which are the independent variables of the adsorption process, were used as input. Adsorption percentage values, which are the dependent variables of the adsorption process, were obtained as output. Mean squared error (MSE) and determination coefficient (R^2) values were obtained from the models created using thirty-one experimental data for adsorption of malachite green with Amberlite IRC-748 and thirty-eight experimental data for adsorption with Diaion CR-11. By evaluating these values together, the most appropriate training algorithm, transfer function in the hidden layer and the number of neurons in the hidden layer were defined. Accordingly, for both Amberlite IRC-748 and Diaion CR-11 resins, the optimum training algorithm was determined as Levenberg-Marquardt back-propagation and the optimum hidden layer transfer function as tan sigmoid. The optimum number of neurons in the hidden layer was identified as 13 for Amberlite IRC-748 and 12 for Diaion CR11. The MSE, R^2_{all} and R^2_{test} values of the models produced with the optimum parameters were obtained as 0.000261, 0.9972, 0.9903 for Amberlite IRC-748 and 0.000482, 0.9932, 0.9931 for Diaion CR11, respectively.

Keywords: *ANN, Dye, Simulation, Optimization, Wastewater, Algorithm*

1. INTRODUCTION

Artificial intelligence is frequently encountered in many fields such as intelligent search, autonomous driving, robotics, and data processing due to its usefulness in tasks such as learning, understanding, prediction, problem solving, suggestion and decision making in various disciplines [1], [2]. Artificial neural network (ANN) is a computational artificial intelligence and machine learning model that simulates the structure and functions of biological neural networks [1], [3], [4]. ANN learns linear, non-linear or complex relations between inputs and outputs from the presented data like the human brain, and the models created in this way can produce new results for different and unknown processes [5], [6].

In ANN, processing units called "neurons" process the input signals according to a specific algorithm and obtain an output signal. The operation of the neurons mentioned here is similar to the operation of biological neurons in the human brain. Accordingly, as schematised in Fig. 1, in the neuron receiving one or more input signals I_j, these inputs are weighted according to a $w_{i,j}$ value. The term weight here is similar to the synaptic strength between interconnected neurons in the human brain. The weighted signals sent to the neuron are summed to form a signal called activation, denoted by h, and sent to a transfer function "g". The resulting O_j output signal can then be sent to one or more neurons or utilized as the output of the ANN model [7], [8].

The human brain is filled with many interconnected neurons. Each neuron executes a simple task such as responding to an input signal. When these neurons are connected to each other like a network, they can execute complex functions such as speech or image recognition very quickly [9]. Similarly, ANN is formed by connecting artificial neurons to within a particular topology, interacting with each other, as

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schematised in Fig. 1. Typically, an ANN contains of an input layer, a hidden layer and an output layer. There may also be more than one hidden layer in the ANN. Artificial neurons are contained within these layers. The input signals supplied to the neurons in the input layer are processed through the neurons across the input, hidden, and output layers to generate an output signal (Fig. 2) [7], [10]. This structure can be used in chemical processes, as in many other fields, to explain the effects of parameters on the outcome of the process and to model the process [1], [3], [6], [11].

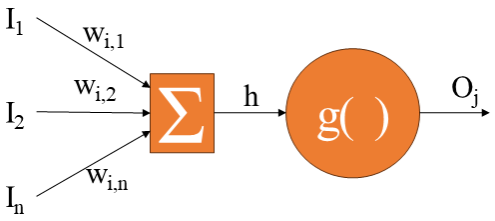

Figure. 1. Structure of a single processing neuron

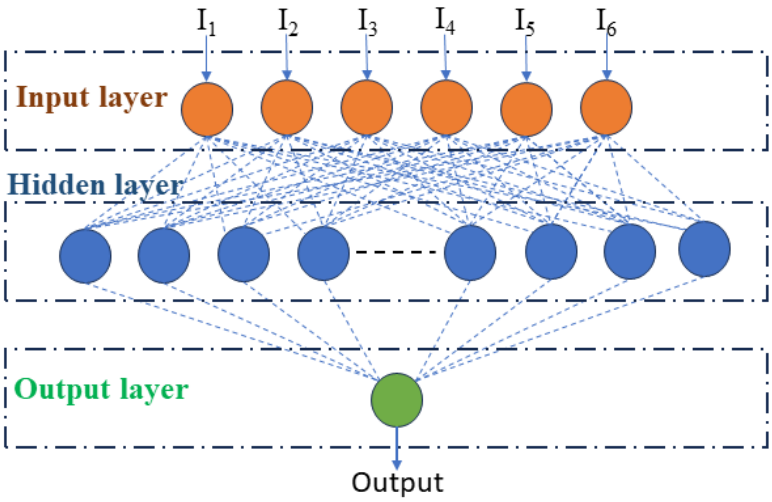

Figure 2. A typical ANN architecture with one hidden layer

In ANN, input signals are converted into output signals in accordance with its structure (Fig. 2). The dataset utilized for the model is divided into three components: training, validation, and test (for example 70% training, 15% validation and 15% test). With the data allocated for training, the network is trained through a training mechanism using the backpropagation algorithm depending on the output obtained. For this purpose, the ANN calculates the error difference between the predicted output and the experimental output (target) and feeds back via backpropagation so that the model readjusts the weights to decrease the error. This process is repeated until the weight changes no longer reduce the error, thus optimizing the model and training the network. Finally, validation and testing are performed with the optimized network [12].

Dyes are widely used in paper, rubber, textile, medical, cosmetic, and plastic production sectors, and are largely discharged into the environment [13], [14]. Since dyes cause colour formation even at low concentrations, they prevent the penetration of sunlight into water and reduce photosynthetic activity [15]. In addition, since their toxicity threatens living life, it is necessary to remove dyes from wastewater [16]. Numerous studies have investigated the removal of dyes using adsorption [17], [18], flocculation [19], chemical precipitation [20], membrane filtration [21], advanced oxidation processes [22], [23], reductive degradation processes [24] and electrochemical methods [25]. Of these methods, adsorption stands out as one of the most effective techniques in wastewater treatment [16].

Batch experiments and adsorption isotherm analyses are commonly employed to examine the removal behavior of different pollutants through adsorption. However, these methods are time-consuming and somewhat inefficient [26]. Since many chemical processes, such as adsorption are affected by many parameters, modelling or optimization of these processes using various statistical or machine learning methods, like ANN, instead of conventional experiments, is very useful [6], [26], [27], [28].

In this study, the adsorption process of malachite green (MG) dye using Diaion CR-11 and Amberlite IRC-748 resins, which were investigated with batch experiments in our previous publication [29] and the results were reported, was modelled by ANN method. The most appropriate ANN algorithm and parameters were determined. As a result, this process was modelled successfully.

2. MATERIALS AND METHODS

2.1. Chemicals

Amberlite IRC-748 and Diaion CR-11 used in the removal studies of MG dye are commercially available materials. Amberlite IRC-748 was purchased from Lenntech. Diaion CR-11 was supplied from Mitsubishi company. Malachite green dye was supplied from Acros Organics. Deionised water was used for preparing dye solution and washing resins before used.

Amberlite IRC-748 and Diaion CR-11 are iminodiacetic acid chelating ion exchange resins with a porous styrene-divinylbenzene matrix. Amberlite IRC-748 features opaque and beige spherical beads, while Diaion CR-11 has highly porous spherical beads. The bulk densities of Amberlite IRC-748 and Diaion CR-11 are 750 g/L and 730 g/L, respectively. The maximum operating temperatures for Amberlite IRC-748 and Diaion CR-11 are 90°C and 120°C, respectively. The suggested pH range for Amberlite IRC-748 is between 1.5 and 14, whereas for Diaion CR-11, it is between 2 and 6.

2.2. Adsorption Studies

MG adsorption by Amberlite IRC-748 and Diaion CR-11 was executed for batch adsorption studies, and experiments were carried out under different parameters. To assess the effect of adsorbent amount, different ratios of resins ranging from 0.1 to 2 g/L were used. Additionally, to investigate the effect of initial dye concentration, dye solutions with concentrations varying from 10 to 240 mg/L were prepared and studies were carried out accordingly. To investigate effect of contact time, experiments were carried out at times ranging from 0-400 min. All experiments were studied at room temperature and with 200 rpm mixer speed.

2.3. Artificial Neural Network

In this study, a neural network model consisting of an input, a hidden and an output layer was created using MATLAB 2022b software. Thirty-one experimental data were used for adsorption of MG with Amberlite IRC-748 resin and thirty-eight experimental data were used for adsorption with Diaion CR-11 resin. The parameters affecting adsorption are input and the removal efficiency obtained by adsorption constitutes the target/output values. Accordingly, three inputs are fed to the model as adsorbent dosage (g/L), MG concentration (mg/L) and contact time (min) and one output value is obtained as adsorption percentage (%). The model has a feed-forward back propagation algorithm. The adsorption percentage (%) value is calculated as in Equation 1.

$$\%\ \text{adsorption} = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

C_0 is the initial MG concentration and C_t is the MG concentration after a certain contact time.

Before creating the model, all input and target values in the experimental data were normalised according to Equation 2 to prevent numerical overflow [6]. Then, these data were divided into 70% for training, 15% for validation and 15% for testing.

$$X_{normal} = \frac{X_i - X_{min}}{X_{max} - X_{min}} \quad (2)$$

X_i is the original data, X_{min} and X_{max} are the minimum and maximum values of the relevant input or target data series.

Subsequently, the learning algorithm of the model, the transfer function in the hidden layer and the number of neurons to be used in the hidden layer were optimized. All training trials for optimization were repeated 10 times, and the result with the lowest mean squared error (MSE) and the highest determination coefficient (R^2) values is presented in this paper. MSE and R^2 values were calculated as in Equations 3 and 4 [6], [30].

$$MSE = \frac{1}{N}\sum_1^n \left(Y_{exp,i} - Y_{prd,i}\right)^2 \quad (3)$$

$$R^2 = 1 - \frac{\sum_1^n \left(Y_{prd,i} - Y_{exp,i}\right)^2}{\sum_{i=1}^n \left(Y_{prd,i} - Y_m\right)^2} \quad (4)$$

$Y_{prd,i}$ is the predicted data of ANN model, $Y_{exp,i}$ is the experimental data, Y_m is the average of the experimental data, and n is the number of data.

In order to determine the training algorithm and the optimum transfer function in the hidden layer, firstly 10 neurons are defined in the hidden layer. Then scaled conjugate gradient backpropagation (trainscg), Levenberg-Marquardt backpropagation (trainlm), gradient descent with momentum backpropagation (traingdm), conjugate gradient backpropagation (traincgp), resilient backpropagation (trainrp) algorithms were used as training algorithms. In the hidden layer, the network was trained using purelin, log sigmoid (logsig) and tan sigmoid (tansig) transfer functions. Purelin, logsig and tansig transfer functions are presented in Equations 5, 6 and 7, respectively [31]. Then, the optimum number of neurons was determined by changing the number of neurons used in the hidden layer from 1 to 20 by using the transfer function with the optimum result. Here, MSE values of the model and R^2 values (R^2_{all} and R^2_{test}) for the whole data set and test data were evaluated together in the selection of the optimum model parameters. Finally, the output and target values obtained in the test using the optimum number of neurons and transfer function were compared, and the accuracy of the model was demonstrated.

$$f(x) = x \quad (5)$$

$$f(x) = \frac{1}{1+e^{-x}} \quad (6)$$

$$f(x) = \frac{e^x - e^{-x}}{e^x + e^{-x}} \quad (7)$$

3. RESULTS AND DISCUSSION

The ranges of the independent variables sent to the model as input in the creation of the ANN model are given in Table 1. The batch adsorption results presented in our previous study [29] were used to determine the ranges. According to these results, the experimental results in the range up to the parameter value where adsorption reaches equilibrium, and 100% adsorption efficiency is obtained were used in the modelling study. Therefore, for Amberlite IRC-748, the ranges of these parameters were set lower since adsorption reached equilibrium at lower adsorbent dosage and contact time. In addition to the

experiments presented in the publication [26], some new experiments were carried out in the ranges shown in Table 1 in order to provide more accurate modelling results.

Table 1. The ranges of data set

Parameters	Ranges	
	Amberlite IRC-748	**Diaion CR-11**
Input layer		
Adsorbent dosage (g/L)	0.03 – 0.42	0.05 – 3.00
MG initial concentration (mg/L)	10 – 240	10 – 240
Contact time (min)	5 – 180	5 – 300
Output layer		
Adsorption (%)	12.8 – 100.0	18.4 – 100.0

3.1. Optimization on ANNs

One of the primary steps to create a suitable ANN for a process is the optimization of model parameters. Model parameters significantly affect the accuracy of a model. For this purpose, the optimal training algorithm and transfer function for the hidden layer were identified by utilizing 10 neurons within the hidden layer. Then, the optimum number of neurons in the hidden layer was determined by using the optimum training algorithm and transfer function.

3.1.1. ANN for MG adsorption with Amberlite IRC-748

The effect of training algorithm and hidden layer transfer functions on model accuracy with regard to MSE and R^2 for modelling MG adsorption data with Amberlite IRC-748 is given in Fig 3. Based on the results obtained, the algorithm and function with optimum accuracy were determined as trainlm and tansig. The MSE, R^2_{all} and R^2_{test} values of this algorithm and function are 0.000405, 0.9930 and 0.9849, respectively.

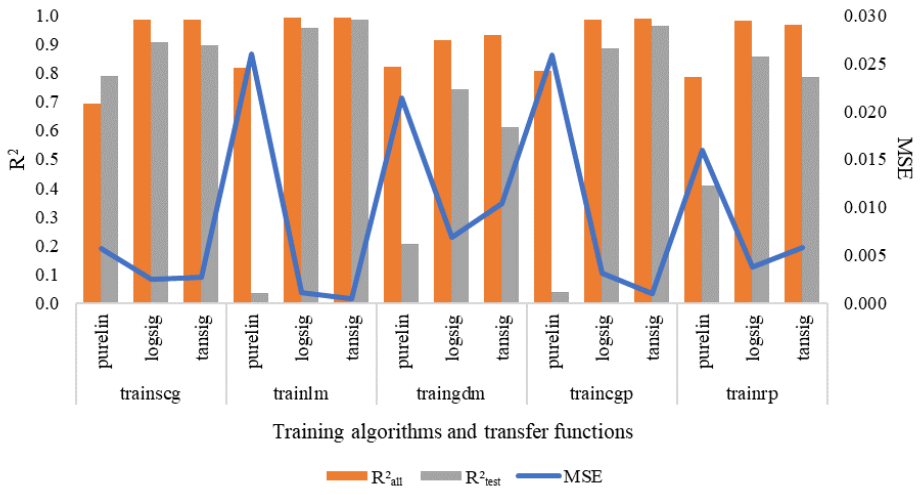

Figure 3. MSE, R^2_{all} and R^2_{test} values for modelling of MG adsorption with Amberlite IRC-748 using different training algorithms and transfer functions

The optimum number of hidden neurons for the ANN using the trainlm algorithm and the tansig transfer function is also analysed (Fig. 4). The figure reveals that the lowest MSE and the highest R^2 values are obtained for 13 neurons. When 13 neurons are used in the hidden layer, MSE, R^2_{all} and R^2_{test} values are 0.000261, 0.9972, 0.9903, respectively. For this reason, 13 was determined as the optimum number of hidden neurons.

The ANN model was constructed using trainlm as the training algorithm, tansig and 13 neurons as the transfer function and number of neurons in the hidden layer. Accordingly, the simulated (output) and experimental (target) adsorption percentage values in the test step are given in Fig. 5. It is understood from the fact that the simulated results and experimental results curves are very close to one to other in the graph that the model can predict the process results with a very high accuracy.

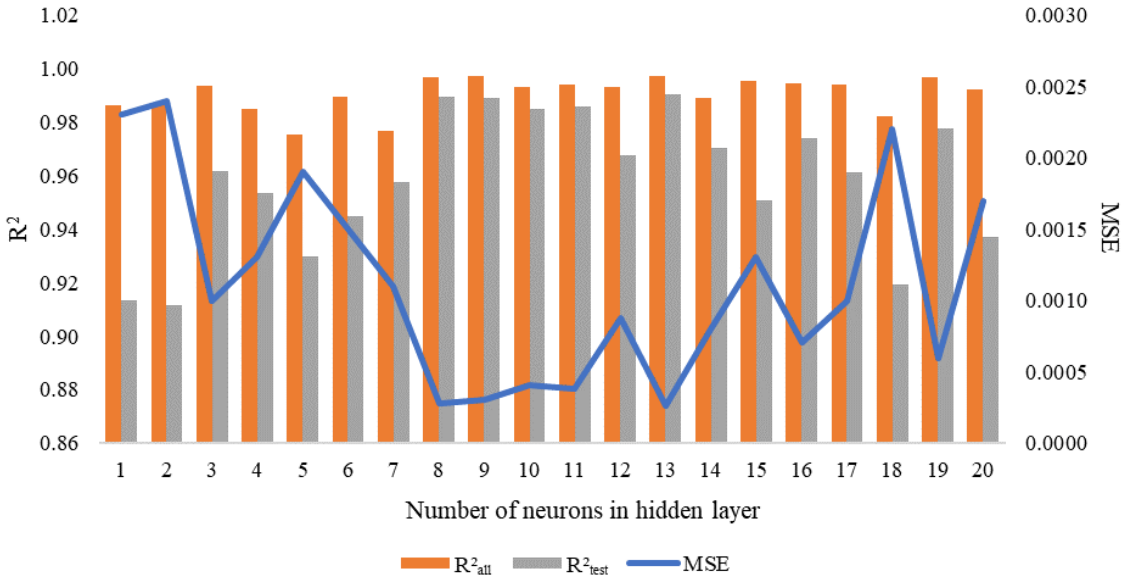

Figure 4. Comparison of ANN models of MG adsorption process with Amberlite IRC-748 according to the number of neurons in the hidden layer

3.1.2. ANN for MG adsorption with Diaion CR-11

The effect of training algorithm and hidden layer transfer functions on model accuracy in terms of MSE and R^2 for modelling MG adsorption data with Diaion CR-11 is given in Fig 6. Based on the results obtained, the algorithm and function with optimum accuracy were determined as trainlm and tansig. The MSE, R^2_{all} and R^2_{test} values of this algorithm and function are 0.001100, 0.9949, 0.9845 respectively.

The optimum number of hidden neurons for the ANN using the trainlm algorithm and the tansig transfer function is also analysed and the results are presented in Fig. 7. The graph illustrates that the lowest MSE and the highest R^2 values are obtained for 12 neurons. When 12 neurons are used in the hidden layer, MSE, R^2_{all} and R^2_{test} values are obtained as 0.000482, 0.9932, 0.9931 respectively. For this reason, 12 was determined as the optimum number of hidden neurons.

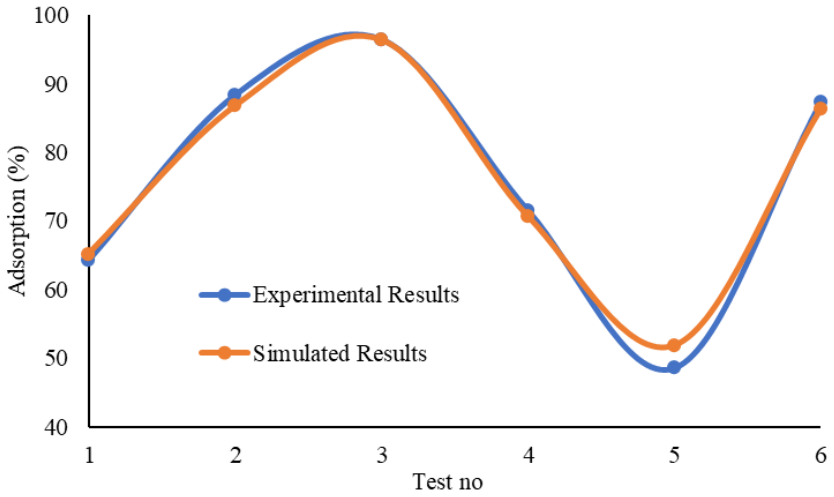

Figure 5. Experimental and simulated results for MG adsorption with Amberlite IRC-748

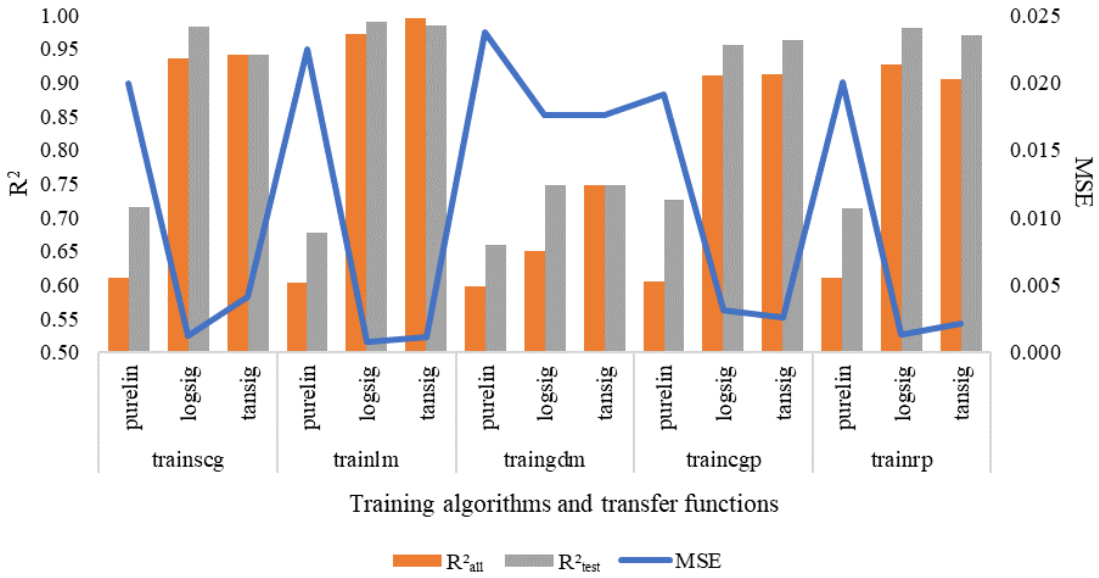

Figure 6. MSE, R^2_{all} and R^2_{test} values for the modelling of MG adsorption by Diaion CR-11 with different training algorithms and transfer functions

The ANN model was created using trainlm as the training algorithm, tansig as the transfer function in the hidden layer and tansig and 12 neurons as the number of neurons. accordingly, the output and target adsorption percentage values in the test step are shown in Fig. 8. This graph was similar to the graph generated for MG adsorption with Amberlite IRC-748. The fact that the simulated results and experimental results curves are very close to each other in Fig. 8 indicates that the model can predict the process results with a very high accuracy.

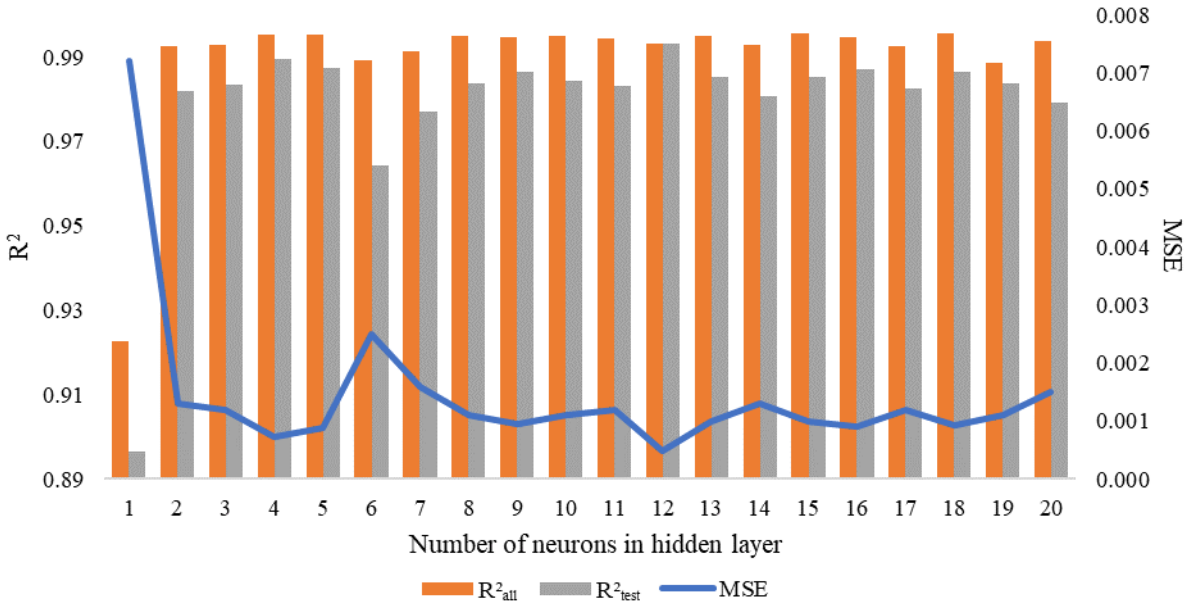

Figure 7. Comparison of ANN models of MG adsorption process with Diaion CR-11 according to the number of neurons in the hidden layer

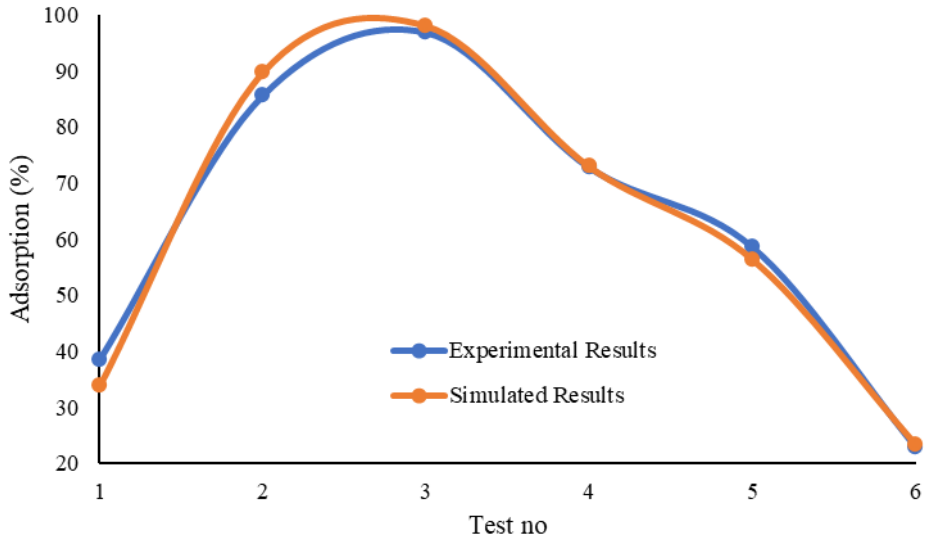

Figure 8. Experimental and simulated results for MG adsorption with Diaion CR-11

According to the studies, MG adsorption processes with Amberlite IRC-748 and Diaion CR-11 resins were modeled successfully and with high accuracy by ANN method. In addition, there is no study in the literature in which these processes are modelled with ANN. For this reason, this study brings an important innovation to the literature.

4. CONCLUSIONS

In this study, the adsorption of malachite green dye by Amberlite IRC-748 and Diaion CR-11 resins, previously studied in batch experiments with published results, has been modelled using the artificial neural network method. Both resins exhibit high adsorption capacities and show significant potential for application in this field. Furthermore, these processes have been successfully modelled with high accuracy in the present study.

Models were created using Thirty-one experimental data for adsorption of malachite green with Amberlite IRC-748 and thirty-eight experimental data for adsorption with Diaion CR-11. In modelling of processes with artificial neural networks, model parameters significantly affect the accuracy of the model. For this reason, the parameters of the training algorithm, the transfer function in the hidden layer and the number of neurons in the hidden layer were optimized through network training experiments. Trainscg, trainlm, traingdm, traincgp and trainrp algorithms were used as training algorithms. Purelin, log sigmoid and tan sigmoid transfer functions were used as transfer functions in the hidden layer. After determining the optimum of these algorithms and functions, the number of neurons in the hidden layer was changed between 1 and 20 to determine the optimum number of neurons. Accordingly, for both Amberlite IRC-748 and Diaion CR-11 resins, the optimum training algorithm was determined as Levenberg-Marquardt backpropagation and the optimum hidden layer transfer function as tan sigmoid. The optimum number of neurons in the hidden layer was determined as 13 for Amberlite IRC-748 and 12 for Diaion CR-11. Modelling results were similar for both resins. MSE, R^2_{all} and R^2_{test} values of the models produced with optimum parameters were obtained as 0.000261, 0.9972, 0.9903 for Amberlite IRC-748 and 0.000482, 0.9932, 0.9931 for Diaion CR-11, respectively. According to these results, malachite green adsorption processes with both resins were successfully modelled using artificial neural network method.

Declaration of Ethical Standards

Authors declare to comply with all ethical guidelines including authorship, citation, data reporting, and publishing original research.

Credit Authorship Contribution Statement

H. ECEVİT: Conceptualization, Methodology, Software, Validation, Formal analysis, Writing - Original Draft, Visualization

D. YANARDAĞ KOLA: Conceptualization, Methodology, Formal analysis, Investigation, Resources, Writing - Original Draft

S. EDEBALİ: Investigation, Resources, Writing - Review & Editing, Supervision, Project administration, Funding acquisition

T. ALTUN: Conceptualization, Writing - Review & Editing, Supervision

Declaration of Competing Interest

The authors declared that they have no conflict of interest.

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Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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