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Application of Artificial Neural Network, Fuzzy Inference System and Adaptive Neuro-Fuzzy Inference System to Predict the Removal of Pb(II) Ions from the Aqueous Solution by Using Magnetic Graphene/Nylon 6

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Abstract

In this study, three modeling techniques based on artificial intelligence were used to predict the removal percent of lead(II) ions from the aqueous solution. These models include Artificial Neural Network (ANN), Fuzzy Inference System (FIS) and Adaptive Neuro-Fuzzy Inference System (ANFIS). Magnetic graphene adsorbent supported on nylon 6 was used for removing lead(II) ions. Optimal conditions for the experimental parameters were performed using the Taguchi methodology. The analysis of variance (ANOVA) test at the 95% confidence level was applied to the results of these models which suggested there were no significant differences among these models.

Keywords: Artificial intelligence models; Magnetic graphene; Nylon6; Lead(II) ions

Introduction

Modeling of chemical processes can lead to lower costs during testing. By using the ability of these models, we can predict the optimal conditions for a process. Intelligent computer systems are the suitable tool that can be employed for this purpose. Artificial Neural Network (ANN), Fuzzy Inference System (FIS) and Adaptive Neuro-Fuzzy Inference System (ANFIS) are part of the intelligent models. ANN is a non-linear statistical data modeling that is inspired by biological neurons. Each neuron is related to a mathematical function with determined inputs, a scientific computation method, and outputs [1,2]. Fuzzy set theory was introduced by Lotfi A. Zadeh in 1965. Fuzzy inference is a process of mapping from a given input to an output data set using the theory of fuzzy sets [3]. On the other hand, ANFIS is a fuzzy inference system combined with the computational power of ANN and acts as an adaptive multilayer feed-forward network. ANFIS is a potent approach to modeling the input and output relationship in non-linear and complex systems [4].

Heavy metals can enter a water supply by industrial and consumer waste by means of various human activity such as mining, fertilizer industries, refining ores, tanneries, battery manufacturing, paper industries, metal-based pesticides and fuels, or even from acidic rain breaking down soils and releasing heavy metals into streams, lakes, rivers, and groundwater [5,6]. Among heavy metals, lead is considered as longstanding environmental contaminant. Lead in large dosage can seriously harm human life and aquatic ecosystems. Where exceeding the permissible concentration limit of lead in the human body can end up with acute or chronic problems such as mental retardation, seizures, anemia, kidney and liver disorder, cancer and hepatitis [7-9]. Therefore, removal of Pb(II) ions from the aqueous system is very important. There are various methods that have been explored to decrease lead(II) ions, namely membrane filtration, chemical precipitation, solvent extraction, ion-exchange, oxidation/reduction, electrode deposition, and bio-adsorption [10-12]. However, these methods have some limitations. Among these methods, adsorption process has been widely used because it is a simple and relatively economical process [13,14]. Graphene (G) as an adsorbent is a two-dimensional form and honeycomb with sp2-bound carbon atoms [15]. It has many unique properties such as singular high room temperature, very high thermal conductivity, flexibility and tensile resistance. This feature has made it a potential adsorbent for the removal of heavy metal ions like lead from the aqueous solution [16-18]. Graphene is produced from graphite based on Hummers and offeman's procedure [19]. In this study, Artificial Neural Network with three algorithms, Fuzzy Inference System and Adaptive Neuro-Fuzzy Inference System have been used for predicting the removal percent of lead ions from the aqueous solution using magnetic graphene oxide supported on nylon 6. The effects of various experimental parameters such as pH of the solution, initial Pb(II) ions concentration, and adsorbent dosage were investigated based on Taguchi experimental design to optimize the absorption performance. To evaluate the significant differences between the reported variances of recovery for removing lead(II) ions the results of ANN, FIS and ANFIS models were compared using ANOVA analysis.

Theory

Artificial neural networks model

Artificial Neural Networks include a contiguous network of nodes, which are separated into many layers. The input and output layers in the neural networks make the main structure of artificial neural networks. In addition, there are a series of hidden layers between input and output layers. The main duty of hidden layers is to evaluate the relationship between unknown and complex by iterative training from many input-output pairs. The hidden layer has some nodes that they have activation functions and numeric weights that are controlled by

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appropriate learning algorithms to obtain the best possible correlation between an input and output set [20]. Figure 1 shows the architecture of the artificial neural network pattern. There is also a weight level between two adjacent levels of input, hidden and output [21,22]. In this research, neural fitting with three algorithms: the Levenberg-Marquardt (LM), Conjugate and Bayesian algorithm were used to evaluate the best algorithm based on prediction accuracy.

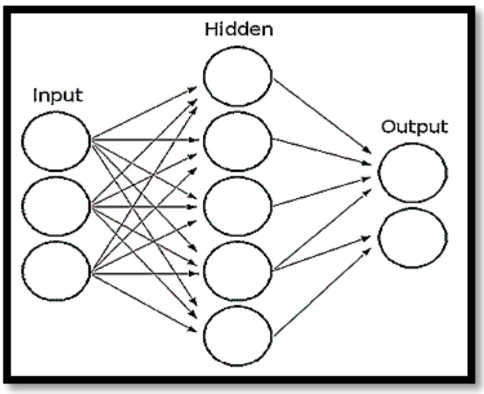

Figure 1: Architecture of artificial neural network model.

Fuzzy inference system model

Fuzzy Inference System applies a nonlinear mapping from a given input to the output using the theory of fuzzy sets. Several fuzzy if-then rules used in this mapping. The parameters of the if-then rules specify a fuzzy area of the input space, and the output parameters [23]. FIS consist of three important parts: rule base, which contains a selection of fuzzy rules, database, which specifies the membership functions (MF) were applied in the fuzzy rules and a reasoning mechanism, evaluates which rules are suitable at the current time [24]. This structure is shown in Figure 2.

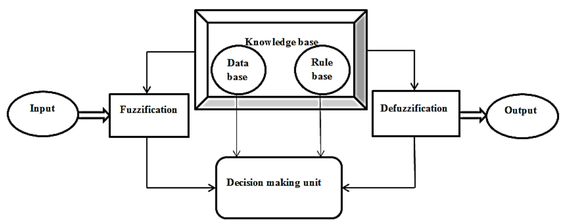

Figure 2: The Fuzzy Inference System Structure.

Membership functions use the fuzzification and defuzzification steps of a fuzzy logic system. Gaussian, triangular and trapezoidal are most usual membership functions [25]. The Mamdani and Sugeno methods are two types of fuzzy inference systems that can be performed in the fuzzy logic. In the Mamdani method subsequences are fuzzy sets and the final output is based on defuzzification of the overall fuzzy output. In the Sugeno method, the results are real numbers, which they are linear or constant [26]. In this study the Sugeno fuzzy model was used.

ANFIS architecture

For a Sugeno fuzzy model a typical rule set with two fuzzy if-then rules can be demonstrated as:

Rule 1: If X is V_1 and Y is D_1, then $F_1=p_1x\ q_1y+r_1$.

Rule 2: If X is V_2 and Y is D_2, then $F_2=p_2x + q_2y+r_2$.

ANFIS structure based on Sugeno model is shown in Figure 3 [27-30].

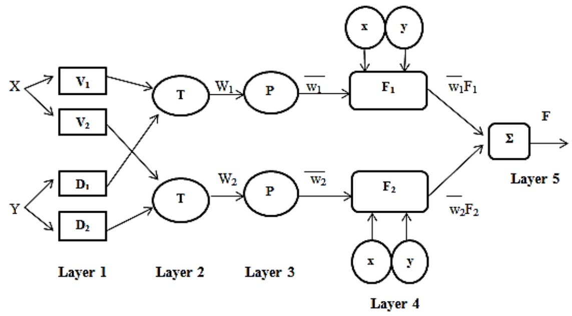

Figure 3: Basic structure of ANFIS.

It has five layers as follows:

Layer 1: this layer is input nodes, which every node is an adaptive node.

Layer 2: rule layer, which multiplies the incoming signals and outputs the product.

Layer 3: average nodes, which they have a normalization rule.

Layer 4: consequent nodes. This layer provides consequent parameters.

Layer 5: output nodes. This layer calculates the overall output of all incoming signals.

In this model, all output membership functions are the same type and they must be either linear or constant. Also, the number of output membership functions and the number of rules is equal. The ANFIS structure is adjusted automatically by the hybrid algorithm that contains a combination of the back propagation and least-squares methods [31,32]. This algorithm optimizes the premise parameters which define the shape of the membership functions [33].

Experimental

Instruments and software

Scanning electron microscopy (SEM, philips XL 30 S-FG) for examining the morphologies of the synthesized particles and flame atomic absorption spectrophotometer (Varan AA 240 Model) were used for measuring the amount of Pb(II) ions removal from the aqueous solution. The results obtained using Microsoft Excel 2010, ANN, FIS and ANFIS Toolbox of MATLAB 8.6.

Chemicals and reagents

Commercial nylon 6 was obtained from the DMS Chemical Company, with averaged number molecular weight 67 Kg/mol. In

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addition, natural graphite powder was supplied from Sigma Aldrich. Ferric chloride ($FeCl_3{\cdot}6H_2O$), ferrous chloride ($FeCl_2.7H_2O$), lead nitrate Pb $(NO_3)_2$, sodium borohydride ($NaBH_4$), sulfuric acid 98% (H_2SO_4), hydrogen peroxide 25% (H_2O_2), potassium permanganate ($KMnO_4$), sodium nitrate ($NaNO_3$), formic acid and other reagents such as HCl, NaOH and Ethanol were obtained from Merck Germany. Stock solutions of Pb(II) ions were prepared from lead nitrate in deionized water.

Procedure

Synthesis of nano-adsorbent

Graphene oxide was synthesized from natural graphite by Hummers and offeman's method [19]. Then, the magnetic graphene oxide (MGO) was synthesized by co-precipitation of $FeCl_3$ and $FeCl_2$. Finally, the magnetic graphene of nylon 6 (MGN_6) was prepared by reduction reaction using sodium borohydride. The pH of the solution, initial lead(II) ions concentration and adsorbent dosage were determined by using optimization based on Taguchi method are given in Table 1. The amount of lead(II) ions removal from the aqueous solution was evaluated using a flame atomic absorption spectrophotometer (Varan AA 240 Model).

Run	pH	Dose (g)	Concentration (mg/L)	Recovery (%)
1	5	0.01	2	20
2	5	0.02	4	55
3	5	0.05	6	60
4	7	0.01	2	88
5	7	0.02	4	92
6	7	0.05	6	93
7	9	0.01	2	96
8	9	0.02	4	89
9	9	0.05	6	91

Table 1: Experimental data obtained using Taguchi method.

Results and Discussion

Scanning electron microscopy (SEM)

The SEM images of the nylon 6 (N_6) and nanocomposite (MGN_6) are shown in Figure 4a and 4b respectively. Figure 4a represents that nylon-6 exhibits a rough and lumpy surface with some uneven and irregular morphology. The morphology of MGN_6 in Figure 4b shows magnetic nanoparticles with a size ranging from 33 to 74 nm, evenly distributed over the graphene/nylon6 composite.

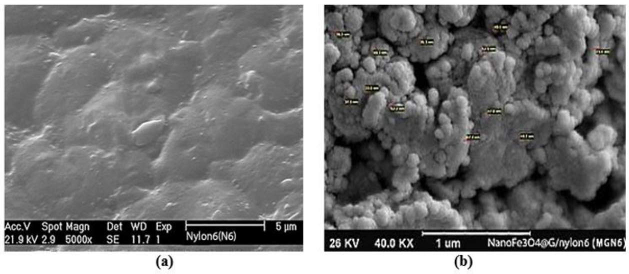

Figure 4: The SEM images of (a) Nylon 6 (N6) and Magnetic Graphene/Nylon6 (MGN6).

Artificial neural network model

The plots of mean squer error (MSE) versus the number of neurons and layers for Levenberg-Marquardt, Conjugate and Bayesian algorithms are shown in Figure 5.

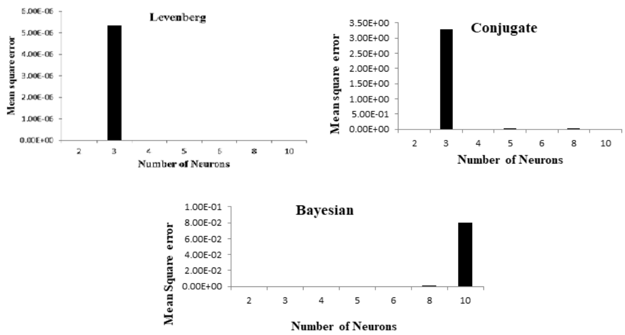

Figure 5: MSE values versus number of neurons for Levenberg-Marquardt, Conjugate and Bayesian algorithms.

The equation of MSE is defined as follows:

$$MSE = \sum_{i=1}^{n} (y_{pred} - y_{obs})^2$$

Where y_{pred} is the predicted value and Type equation here y_{obs} is the actual value.In Levenberg-Marquardt algorithm, the least error is related to hidden layer 2. In Conjugate algorithm, hidden layer 10 has less error. In the Bayesian algorithm, the hidden layer number 3 has the least error. The predicted values against the actual values of validation and training set for the best neurons and layers are shown in Figure 6. As can be seen in Figure 6, correlation coefficients (R^2) clearly show better predictive ability of the algorithms. The statistical results obtained by different training and validation algorithm sets are reported in Table 2.

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(a)

(b)

(c)

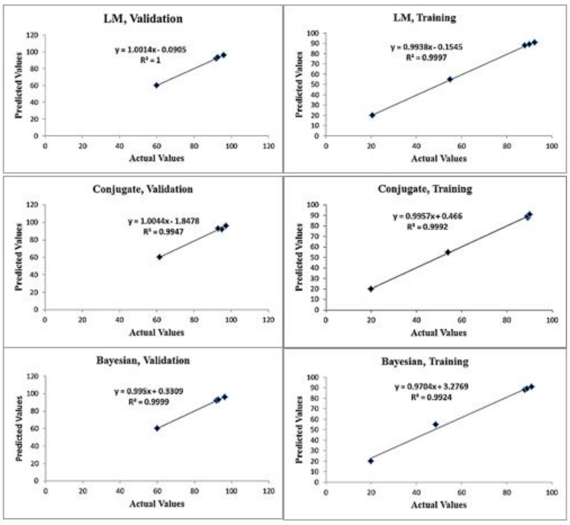

Figure 6: Predicted values versus actual values for validation and training of (a) LM, (b) Conjugate and (c) Bayesian algorithms.

As it can be observed in Table 2, among the three algorithms the Root Mean Square Error (RMSE) of the Levenberg-Marquardt algorithm is lower than the Conjugate and Bayesian algorithms. The RMSE is defined by the following equation [34]:

$$RMSE = \sum_{i=1}^{n} (y_{pred} - y_{obs})^2$$

Where y_{pred} is the predicted value in the sample, y_{obs} is the actual value of the sample, N is the number of samples and e is the mean of the actual value.

Sample Number	Actual	Recoveries (%) of Levenberg-Marquardt	Recoveries (%) of Conjugate	Recoveries (%) of Bayesian
Training Data				
1	20	102.85	100	100
2	55	100	98.18	88.34
3	88	99.99	101.3	100
4	89	101.14	100	100
5	91	101.45	98.9	100
Mean Recovery		101.09	99.67	97.66
RMSE		0.79	0.815	2.86
Validation Data				
18	92	99.99	103.26	100
19	93	99.87	100	100
20	96	99.99	101.25	100.4
21	60	100.01	102.73	100
Mean recovery		99.96	101.81	100.1
RMSE		0.006	1.81	0.195

Table 2: Recovery data obtained by application of the Levenberg-Marquardt, Conjugate and Bayesian algorithms.

Fuzzy inference system model

There are three inputs containing the concentration of the solution (c), amount of absorbent (mg) and pH of the solution (pH).

In the proposed model, 9 rules are established, and these rules can be expressed in the if-then form. For example:

Rule 1: If input 1=4 and input 2=0.02 and input 3=7 then output=89

Rule 2: If input 1=4 and input 2=0.02 and input 3=7 then output=100

Table 3 shows fuzzy inference rules for the multivariate systems. Figure 7 shows the rule viewer of FIS model.

Serial No	C	mg	pH	Recovery (%)
1	2	0.01	5	20
2	4	0.02	5	55
3	6	0.05	5	60
4	2	0.01	7	88
5	4	0.02	7	92
6	6	0.05	7	93
7	2	0.01	9	96
8	4	0.02	9	89
9	6	0.05	9	91

Table 3: Fuzzy rules of FIS and ANFIS models.

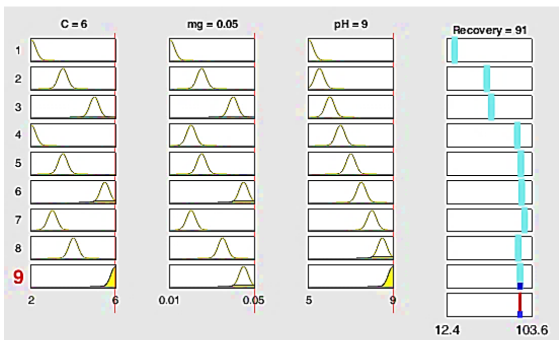

Figure 7: Rule viewer of Fuzzy Inference System model.

In this model, the relationship between the concentration (c), adsorption dosage (mg), and percent recovery is presented in Figure 8.

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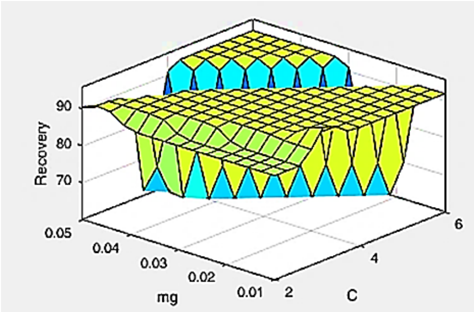

Figure 8: The output surface for Fuzzy Inference System model.

Also predicted values against actual values for FIS model is shown in Figure 9.

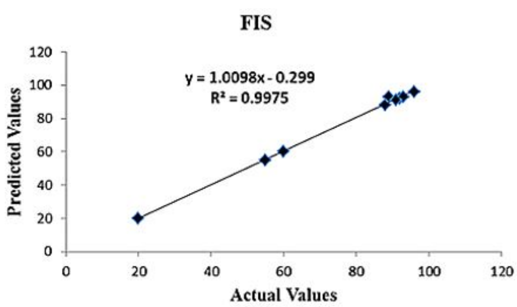

Figure 9: The relationship between predicted values and actual values for FIS model.

In Figure 9 the R^2 value is 0.9975 that indicates better ability of the prediction of this model. Mean recovery (%) and RMSE of the proposed method calculated are summarized in Table 4.

FIS		
Actual	**Predicted**	**Recoveries (%)**
20	20	100
55	55	100
60	60	100
88	88	100
92	92	100
93	93	100
96	96	100
89	96	104.49
91	91	100
Mean		100.49
RMSE		1.33

Table 4: Recovery and RMSE obtained by application of the FIS model.

The results in Table 4 show that this model is fairly good for predicting the mean recovery (%) and RMSE values. As can be observed in Table 4, the value of RMSE was found to be 1.33.

Adaptive neuro-fuzzy inference system model

This model developed using 9 numbers of membership functions of type 'gaussmf ' with 9 If-then rules. The Rules based on Neuro- fuzzy for multivariable are displayed in Figures 10 and 11 represents the structure of ANFIS model which has three inputs and one output.

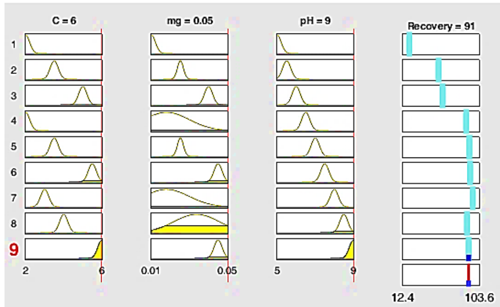

Figure 10: Rule viewer of ANFIS technique.

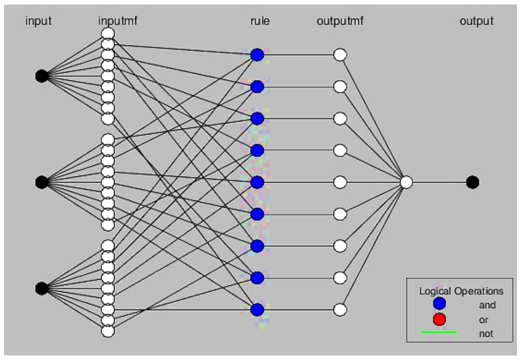

Figure 11: Adaptive Neuro-Fuzzy Structure.

Each input is connected to 9 membership functions. This model has 9 rules. Each rule is connected with one membership function and these membership functions produced the output. Several types of membership functions were used in proposed models and the gaussmf shape selected based on the statistical parameters. Hybrid learning algorithm combining the least-squares method and the gradient descent method are used in ANFIS model. The developed model was applied by using 10 epochs and error of tolerance was found to be zero.

The ANFIS graphs for describing the relationship between inputs and recovery factor are presented in Figure 12.

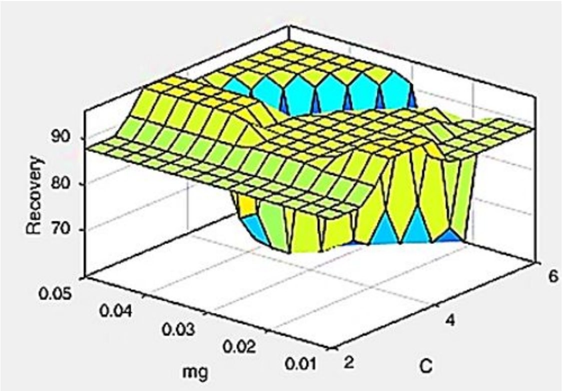

Figure 12: The relationship between predicted values and actual values for ANFIS model.

The plots of the predicted versus reference values are shown in Figure 13.

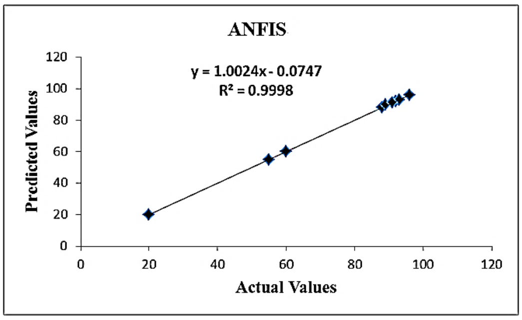

Figure 13: The relationship between predicted values and actual values for ANFIS model.

The correlation coefficient (R^2) was 0.9998 which is close to 1 that shows the suitability of the ANFIS model. The statistical results of Adaptive Neuro-Fuzzy Inference System consist of mean recovery (%) are summarized in Table 5. Table 5 shows that ANFIS model has the higher ability of prediction because of lowest RMSE value.

ANFIS		
Actual	**Predicted**	**Recoveries (%)**
20	20	100
55	55	100
60	60	100
88	88	100
92	92	100
93	93	100
96	96	100
89	90	101.12
91	91	100
Mean		100.12
RMSE		0.333

Table 5: Recovery and RMSE obtained by application of the ANFIS model.

The analysis of variance

To investigate the existence of the significant differences between the reported variances of recovery for removing lead(II) ions the results of ANN, FIS and ANFIS models were compared with each other using ANOVA test. The obtained results are presented in Table 6. Due to the calculated F value which is less than the F critical, there is no significant differences between the variances at the 95% confidence level.

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	0.3675	2	0.18375	0.999637	0.464859	9.552094
Within Groups	0.5514	3	0.18381			
Total	0.9189	5				

Table 6: The ANOVA test results by applying ANN, Fuzzy and ANFIS models.

Conclusion

In this study, the prediction ability of artificial intelligence models for the removal of lead(II) ions from the aqueous solution using the magnetic nano absorbent were carried out. The artificial neural network with three different algorithms (Levenberg-Marquardt, conjugate, and Bayesian), fuzzy inference system and adaptive neuro fuzzy inference system, were used. The results obtained from the neural network with the Levenberg-Marquardt algorithm showed better performance than the other two algorithms. Moreover, according to the results obtained using FIS and ANFIS models, it can be suggested that the prediction ability of the ANFIS is better than the other model. The results from proposed models compared with the analysis of variance (ANOVA) test at the 95% confidence level showed that there were no significant differences between these models.

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