Application of Adaptive Neural Fuzzy Inference System and Fuzzy C-Means Algorithm in Simulating the 4-Chlorophenol Elimination from Aqueous Solutions by Persulfate/Nano Zero Valent Iron Process

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ABSTRACT
This study investigated the application of adaptive neural fuzzy inference system (ANFIS) and Fuzzy c-means (FCM) algorithm for the simulation and prediction of 4-chlorophenol elimination in aqueous media by the persulfate/Nano zero valent iron process. The structure of developed model which resulted to the minimum value of mean square error was a Gaussian membership function with a total number 10 at input layer, a linear membership function at output layer and a hybrid optimum method, which is a combination of backpropagation algorithm and least squares estimation, for optimization of Gaussian membership function parameters. The prediction of developed model in elimination 4-chlorophenol was significantly close to the observed experimental results with R2 value of 0.9942. The results of sensitivity analysis indicated that all operating variables had a strong effect on the output of model (4-CP elimination). However, the most effective variable was pH followed by persulfate, NZVI dosage, reaction time and 4-CP concentration. The performance of developed model was also compared with a quadratic model generated in a study by Response Surface Methodology (RSM). The results indicated that the ANFIS-FCM model was superior to the quadratic model in terms of prediction accuracy and capturing the behavior of the process.

Keywords: persulfate, nano zero valent iron, ANFIS, fuzzy c-means, RSM

INTRODUCTION
Chlorophenols are toxic and hazardous compounds which can be introduced to the aquatic media via various industrial effluents and chlorination of phenol-containing wastewater [1, 2]. Due to their adverse effects on human beings and aquatic life, these contaminants have been listed among the high priority toxic and dangerous pollutants [3-6]. In the recent years, application of nanoparticles in the treatment processes has been increased [7-11]. Advanced oxidation processes (AOPs) like zero valent iron/persulfate, ferrous iron/ persulfate and Nano zero valent iron/ persulfate are well known and effective techniques for elimination of various organic pollutants from aqueous solutions with generating sulfate free radicals and reactive oxygen species (ROS). Application of these processes in the decomposition of phenolic compounds such as bisphenol [12], 2, 4-dichlorophenol [13] and alkyl phenol polyethoxylate [14] has been reported in the previous works. Wastewater treatment by means of AOPs is really complicated [15], because these processes are affected by several process factors. Owing to the complication of oxidation based processes, simulating and modeling of these processes are not easy by mathematical standard modeling [16]. Hence, computational intelligent methods such as artificial neural network and adaptive neural fuzzy inference system have been introduced to the engineering sciences and researches due to the higher accuracy.
and more simplicity of them in modelling and prediction toward the conventional modeling methods [16, 17]. Furthermore, the intelligent models are considered as non-parametric models (no need to high background of statistic) and can acquire linear and nonlinear relations among the process parameters only from a set of experiment runs [18]. Application of intelligent methods for modeling and simulating the wastewater treatment processes has been well documented in several articles [19-22]. Artificial neural network and adaptive neural fuzzy inference system as most well-known intelligent methods were used in simulating the physicochemical and biological wastewater treatment processes [16]. However, a small number of studies has been conducted on applications of these methods in advanced oxidation processes.

The current work studied the application of adaptive neural fuzzy inference system and fuzzy c-means algorithm for the forecast of 4-chlorophenol degradation by persulfate/Nano zero valent iron process. The ANFIS-FCM modelling results were compared with the actual values of experimental data. In addition, a sensitivity analysis was implemented for determine the most influencing factors in the process.

EXPERIMENTAL

Chemicals

All chemicals were analytical grade and used without further purification. 4-Aminoantipyrine, Sodium persulfate, 4-chlorophenol, Potassium ferricyanide, NH₄OH, KH₂PO₄, H₂SO₄, NaOH and K₂HPO₄ were purchased from Merck Company. Zero valent iron nanoparticles (pore size 35-45 nm) were purchased from Nanosany Corporation (Iran).

Experimental Procedure

All experimental tests were conducted in a six liters laboratory scale stainless steel reactor. Two L of synthetic wastewater with 50-500 mg/L of 4-chlorophenol and a proper amount of NZVI (0-1g/L) and persulfate (0-2 mM) was set to pH values 3-11 using 1 M H₂SO₄/NaOH and added into the reactor at room temperature (20 ± 2 °C). The reactor was placed on the shaker to provide the homogenized mixture of solution throughout a predesignated reaction times (5-90 min) of the process. At the end of a reaction time, 10 mL of sample was taken and evaluated to determine the 4-CP concentration changes. The concentration of 4-CP at different conditions were determined by UV–visible spectrophotometer (colorimetric method, 5030D, APHA-2005). The removal percentage of 4-CP was calculated based on the equation (1):

\[
\text{Removal} \% = \left(\frac{C_i - C_f}{C_i}\right) \times 100
\]

(Eq.1)

where, \(C_i\) and \(C_f\) are initial and final concentration of 4-CP respectively in the process. It is noted that, before measuring the 4-CP concentration the samples were filtrated using syringe filter 0.22 micron to separate the NZVI and diluted with deionized water according to the colorimetric method, 5030D, APHA-2005.

Design of Experiments

In the present work, a central composite design (CCD) of experiments was used for design of experiment runs. CCD is one of the most commonly used approach in Respons Surface Methodology (RSM). In general, RSM is used for mathematical description of the response (% removal) and process parameters relationships. A set of 62 experiment trials (Table 1) was obtained using CCD with applying the five process parameters including: pH, initial persulfate concentration, reaction time, initial 4-CP concentration and initial NZVI dosage.

ANFIS

ANFIS is an adaptive neural fuzzy inference system which was first proposed in 1993 by Jung. The system is an intelligent hybrid method, which uses features of the Takagi-Sugeno fuzzy inference system and adaptive neural network [23]. An ANFIS model is usually defined via five certain layers for the purpose of description the model structure (Figure 1(a)). These 5 layers of ANFIS, according to hierarchy and also their roles, consecutively are: the Fuzzification layer (first layer), in this layer all the nodes are adaptive nodes. The outputs of the first layer are the degree of membership which are computed by applying a Gaussian membership function to the inputs in a form of fuzzy (Eq. (2)), firing strength (weight) of each rule is computed in the second layer according to Eq. (3), it includes the fixed nodes and its nodes are labeled with \(\Pi\), denoting that they apply the process of multiplying, third layer is known as the normalization layer, in this layer similar to the second layer nodes are fixed nodes and labeled by N, each node performs the normalization for firing strength of each rule (Eq. (4)), nodes in the defuzzification layer (fourth layer) are adaptive nodes and the parameters of these nodes (ri, qi, pi) are called
consequent parameters. Eq. (5) can be written to describe the layer 4. In this equation f is a first order function of the Sugeno fuzzy system and finally the fifth layer or summation layer which is a single node labeled by \( \Sigma \), representing the overall output of the ANFIS model (Eq. (6)) [24].

\[
\mu_{A_i} = \exp \left( - \left( \frac{x - c_i}{a_i} \right)^2 \right) \quad \text{(Eq. 2)}
\]

where \( a_i \) and \( c_i \) are the parameters of the membership function and \( x \) is the input to node \( i \).

\[
w_i = \mu_{A_i}(x_1) \times \mu_{B_i}(x_2) \quad \text{(Eq. 3)}
\]

where \( A_i \) and \( B_i \) are the fuzzy set associated with node.

\[
\bar{w}_i = \frac{w_i}{w_1 + w_2} \quad \text{(Eq. 4)}
\]

\[
\bar{w}_{i1}f_i = \bar{w}_i(p_1\alpha + q_1y + r_1) \quad \text{(Eq. 5)}
\]

where \( \bar{w}_i \) is the output of third layer and \( [p_1, q_1, r_1] \) are the parameters set.

\[
\sum_i \bar{w}_{i1}f_i = \frac{\sum_i w_{i1}f_i}{\sum_i w_i} \quad \text{(Eq. 6)}
\]

FCM

Fuzzy C-Means (FCM) is an approach of data clustering in which a given dataset is partitioned or grouped into some clusters according to the principles of the fuzzy C-partition. The introduction of FCM first was by Ruspini then it was improved and generalized by Dunn and Bezdek, respectively [25]. In this approach, each point of a dataset belonging to a cluster, consistent with score of its membership degree. For instance, a data point is near to the center of a cluster takes high score of belonging (membership degree) to that cluster and any other data point which is far away from center of a cluster takes low score of belonging to that cluster. In general, putting each of the data points to one of the clusters is main goal of the FCM algorithm. In This study, the FCM algorithm were applied to divide the total training dataset (input/ output) into the various number of Gaussian membership functions (subsets) with different centers. Every subset trained using ANFIS function to find the best number of membership functions base on the minimum values of MSE for Training, validation and testing datasets.

Data Processing

In this work, before developing the ANFIS-FCM models the experimental datasets were randomly divided into training (80%), validation (10%) and test (10%) subsets. The purpose of splitting the experimental data was to evaluate the robustness of the developed models for the prediction of unseen data. In addition, The ANFIS-FCM model inputs (process parameters) and outputs (% removal) were normalized between -1 and 1 using Eq. (7).

\[
y = \frac{x_i - x_{\text{min}}}{x_{\text{max}} - x_{\text{min}}} \times (b - a) + a \quad \text{(Eq. 7)}
\]

where \( y \) is normalized value of \( x_i \), \( x_{\text{max}} \) and \( x_{\text{min}} \) are maximum and minimum values of experimental data respectively; \( a \) and \( b \) are equal to -1 and 1 respectively.

![Figure 1. (a) The architecture of developed ANFIS model; (b) The fuzzy section of the ANFIS model](image-url)
RESULTS AND DISCUSSION

Central Composite Design

The experimental tests were conducted in accordance with CCD to figure out the operating parameters effects on 4-chlorophenol elimination in persulfate/Nano zero valent iron system. As can be seen in Table 1, a set of sixty two experiments, was used for the study. According to the obtained results from the experimental study, a second order polynomial equation (quadratic model) was developed using multiple regression analysis. Eq. (8) shows the reduced quadratic model which was developed in this study. The developed model contained five main effects, seven interaction effects and three quadratic effects as follows:

\[ Y = 43.926 - 7.664X_1 + 9.866X_2 + 3.917X_3 - 3.833X_4 + 7.95X_5 - 7.009X_1X_2 + 4.576X_1X_3 - 15.71X_1X_4 - 11.718X_2X_3 - 8.038X_2X_4 - 10.237X_3X_5 \]

\[ - 5.764X_4X_5 + 14.585X_1^2 + 10.640X_2^2 - 7.374X_3^2 \]  

(Eq.8)

Equation (8) suggests that 4-chlorophenol removal efficiency increases with increasing the persulfate concentration (X2), reaction time (X3), zero valent iron nanoparticle dosage (X5) and along with decreasing the pH (X1) and initial 4-chlorophenol concentration (X4).

Analysis of variance (ANOVA) was employed to figure out the significance of the developed quadratic model. It is recommended that for a statistically valid model, adjusted determination coefficient (R^2_adjust) should be in the range of determination coefficient (R^2) ± 0.2 [26]. As can be seen in Table 2, the values of R^2 and R^2_adjust for the developed model are 0.913 and 0.8847 respectively. Therefore, difference between them is 0.0283. Furthermore, the value of model lack fit was 0.0595. The model lack fit is another index which is used to describe the adequacy of developed models and it should not be significant toward the pure error [26]. The obtained values of R^2, R^2_adjust

<table>
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<th>Table 1. Central composite design of experiments and responses</th>
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</table>

x1=pH; x2=persulfate; x3=Time; x4=4-CP concentration; x5=NZVI dosage
and model lack fit in this study suggest that the behavior of persulfate/Nano zero valent iron system can be explained by the developed quadratic model. Figure 2(a) shows the scatter plot of observed removal percentages in the experimental study against the predicted removal percentages by the developed model. As seen in this figure, the fitted values of the model are relatively close to the observed values in the experimental study. The developed model can explain 91.3 percent variation in the forecasted and actual values. The distribution of the residual errors is shown in the Figure 2(b). As seen, the residual errors were within the ±6.

**Effect of Process Operating Parameters on 4-Chlorophenol Elimination**

The graphical results of developed quadratic model (Eq. (3)) in the current study are presented in Figure 3(a-f). A set of three dimension (3D) response graphs was employed for visualizing the interaction effects of operating process parameters toward the response (% removal). A 3D response graph can illustrate the optimal points in which the maximum response is occurred [26]. 4-CP degradation as a function of NZVI dosage, pH and persulfate concentration is presented in Figure 3(a, b). As seen, 4-CP degradation increased with decreasing the solution pH along with increasing the persulfate concentration and NZVI dosage. Usually, acid conditions accelerate the generation rate of ferrous ions (Fe²⁺) according to the Eq. (9) in the persulfate/NZVI system. Hence, with increasing the Fe²⁺ in the system, the formation of sulfate free radicals increases (Eq. (10)). This result is in agreement with previously studied works [27, 28].

\[
Fe^0 + H^+ \rightarrow Fe^{2+} + H_2 \quad \text{(Eq.9)}
\]

\[
S_2O_8^{2-} + Fe^{2+} \rightarrow Fe^{3+} + SO_4^{2-} + SO_4^{2-} \quad \text{(Eq.10)}
\]

Figures 3(c) and 3(e) demonstrate that 4-CP removal is a function of the reaction time. As shown in these figures, the elimination of 4-CP was occurred in the all predesignated times. The figures suggest, the removal efficiency increases at the lower reaction times and higher dosage of NZVI (Figure 3(d)) as well as higher concentration of persulfate (Figure 3(c)). This is expected due to the rapid transformation of NZVI to Fe²⁺ by the oxygen molecules.
and persulfate at initial phase of reaction. However, at higher reaction times the removal efficiency was also increased. This is due to the unique feature of NZVI which can produce ferrous ions slowly over the time [27]. Therefore, with consumption of oxygen in the initial phase of reaction it is required more reaction times to produce sufficient quantities of ferrous ions.

Figures 3(d) and 3(f) show that 4-CP elimination is a function of initial 4-CP concentration. It is clear from these figures, when the 4-CP concentration increases from 50 to 500 mg/L in the persulfate/NZVI system, the removal efficiency decreases. This is a consequence of covering the active sites on the surface of NZVI by 4-CP molecules which hinder the release of Fe^{2+} ions to the system [29].

**ANFIS-FCM Modeling**

With introduction of fuzzy inference systems in solving the engineering problems, due to the great features of these methods in the control systems, as well as simple expression of the variables in terms of linguistic variables, use of these methods in many applications have been developed and expanded quickly. But designing a powerful fuzzy inference system, which needs to determine proper membership functions and fuzzy rules, is not easy, and mainly will be possible with a lot of trial and error and with the involvement of experts’ comments in order to achieve the best performance, and in some cases, identification of all the rules is impossible. For this reason, the idea of using learning algorithms of the artificial neural network was introduced for fuzzy systems. There are two learning strategies in neural part of the ANFIS: Hybrid learning and back-propagation learning methodologies. In fuzzy part of the ANFIS, only first or zero order Sugeno inference system may be used [23, 24]. The architecture of fuzzy section of the developed ANFIS for 4-CP removal is shown in Figure 1(b). As can be seen in this figure, Output variable (removal %) is calculated by applying 10 fuzzy rules to fuzzy sets of 5 input variables including: pH, initial persulfate concentration, reaction time, initial 4-CP concentration and initial NZVI dosage. An ANFIS works by applying above mentioned neural network learning approaches to adjust the parameters of a Fuzzy
Inference System (FIS). To develop ANFIS-FCM based models the experimental dataset [input/output] in training step were clustered into the various numbers according to the FCM algorithm as mentioned in section of FCM. In fact, the initial Fuzzy inference system which contains the Gaussian membership function (MF) was determined by FCM to each variable. For this, a MATLAB code was written by help of MATLAB R 2013a software. The performance of ANFIS-FCM models were assessed based on the mean square error index (MSE). The results of different Gaussian membership function numbers (clusters number) along with optimum method types, epoch number and corresponding MSE values are presented in Table 3. It was found that when ANFIS-FCM model, developed with a total membership numbers equal to 10 (for each variable), optimum method hybrid and with 200 epochs resulted to the MSE values of 0.0003, 0.0039 and 0.0011 for training, validation and testing sets respectively. As seen, in the best ANFIS-FCM model there are 10 linguistic fuzzy rules for describing the 4-CP removal (Figure 1(b)).

<table>
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<th>Number of Cluster</th>
<th>Output MF type</th>
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Figure 4. (a) Scatter plot of ANFIS-FCM prediction values and actual experimental data; (b) distribution of the residual errors for training, validation and test dataset

Inference System (FIS). To develop ANFIS-FCM based models the experimental dataset [input/output] in training step were clustered into the various numbers according to the FCM algorithm as mentioned in section of FCM. In fact, the initial Fuzzy inference system which contains the Gaussian membership function (MF) was determined by FCM to each variable. For this, a MATLAB code was written by help of MATLAB R 2013a software. The performance of ANFIS-FCM models were assessed based on the mean square error index (MSE). The results of different Gaussian membership function numbers (clusters number) along with optimum method types, epoch number and corresponding MSE values are presented in Table 3. It was found that when ANFIS-FCM model, developed with a total membership numbers equal to 10 (for each variable), optimum method hybrid and with 200 epochs resulted to the MSE values of 0.0003, 0.0039 and 0.0011 for training, validation and testing sets respectively. As seen, in the best ANFIS-FCM model there are 10 linguistic fuzzy rules for describing the 4-CP removal (Figure 1(b)). Figure 4(a) is the scatter plot of forecast values for 4-CP removal in the training, validation, testing and all prediction sets versus corresponding experimental data. As can be seen in this figure, for all prediction set, determination coefficient (R²) value is 0.9942 and best linear fit equation is, y = 0.985x + 0.6125. The R² value 0.9942 demonstrates that the developed model with 10 linguistic fuzzy rules can explain 99.26 percent variation in the forecasted and actual values. The distribution of the residual errors is shown in the Figure 4(b). As can be seen in this figure, the residual errors were within the ±1.8.

Sensitivity Analysis

To perform the sensitivity analysis and determine the most influencing variables in the ANFIS-FCM model output, five ANFIS-FCM models were constructed using a single variable in the optimum conditions of best developed model [30]. The developed models were evaluated based on the determination coefficient (R²) to determine the most effective variable. Therefore, each of the developed models, which had the highest R² value, it was considered as the most important process variable. The values of R² for the developed models are shown in
Figure 5. Sensitivity analysis for ANFIS-FCM model

Figure 5. As can be seen in this figure, all input variables (pH, persulfate concentration, reaction time, initial 4-CP concentration and NZVI dosage) had a strong effect on the output of ANFIS-FCM models (4-CP removal). However, the most effective variable was pH with $R^2$ value equals to 0.3409 followed by persulfate ($R^2=0.3212$), NZVI dosage ($R^2=0.2335$), reaction time ($R^2=0.1118$) and 4-CP concentration ($R^2=0.0749$).

CONCLUSION

In the present study, we investigated NZVI as a source of ferrous ions to convert the persulfate to sulfate free radicals and decompose the 4-CP in aqueous solutions. In the developed system the conversion of persulfate to sulfate free radical was fast as a result of rapid corrosion of NZVI surface and release of ferrous ions. Higher dosages of NZVI and persulfate led to the higher 4-CP elimination owing to enhanced transformation of persulfate to sulfate radical via NZVI. In addition, we investigated the application of adaptive neural fuzzy inference system and Fuzzy c-means algorithm for the simulation and prediction of 4-chlorophenol elimination in the process. The best ANFIS-
FCM model which resulted to the minimum value of MSE was a Gaussian membership function with a total number 10 at input layer, a linear membership function at output layer. A hybrid optimum method was used to find the best values of Gaussian membership function parameters in training step of ANFIS-FCM model. The prediction of developed ANFIS-FCM model in elimination 4-chlorophenol was significantly close to the observed experimental results with R² value of 0.9942. The results of sensitivity analysis indicated that all operating variables (pH, persulfate concentration, reaction time, initial 4-CP concentration and nZVI dosage) had a strong effect on 4-CP elimination. However, the most effective variable was pH with R² value of 0.3409 followed by persulfate, nZVI dosage, reaction time and 4-CP concentration. The performance of the developed ANFIS-FCM model was also compared with a quadratic model generated in a study by the Response surface methodology (RSM). The results indicated that the ANFIS-FCM model was superior to the quadratic model of RSM in terms of prediction accuracy (capturing the behavior of the process) and residual errors.

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