Chapter – IV

PROCESS OPTIMIZATION TECHNIQUES

The jar-test experiments and Fenton oxidation for removal of chlorpyrifos from aqueous solution were optimized using response surface methodology and adaptive neural-fuzzy inference system for describing the correlation between experimental factors (independent variable) and observed results (responses). Here the response surface methodology and adaptive neural-fuzzy inference system are briefly described.

4.1 Response Surface Methodology

The overall objective of design of experiment (DOE) is to assure that the dependencies between experimental conditions and the results of the experiments (the responses) can be optimized at minimal cost, i.e. with a minimum number of experiments and is well suited for fitting a quadratic model. Response surface methodology (RSM) was used to optimize the process. The central composite design (CCD) was designated in order to analyze the effect of process parameters.

In the field of water and wastewater treatment, the RSM is commonly used for process optimization. The RSM is a statistical technique for empirical model building and it was documented by Box and co-workes in the 50s. An essential aspect of RSM is the design of experiments (Bezerra et al., 2008; Kushwaha et al., 2010). An experiment is a series of tests, called runs, in which input variables are changed in order to identify the changes in the output response. This is appropriate for fitting a second order polynomial with a minimum number of runs. Therefore, an attempt has been made by using Software Design Expert 9.0.5 (State-Ease Inc., USA) for the design of experiment, statistical analysis of data, development of regression models and optimization of coagulation-flocculation and Fenton process conditions with the factorial experimental design of central composite design.

4.1.1 Central composite design

Numerous process parameters in Fenton oxidation have been premeditated with a standard RSM design called a central composite design (CCD) (Sinha et al., 2012; Ismail et al., 2013). The central composite design (CCD) was first used by Box and Wilson in 1951 (Bezerra et al., 2008). Normally, the CCD consists of *2n* factorial runs with *2n* axial runs and *nc* center runs (six replicates).

$$N = 2n + 2n + nc \quad (4.1)$$

where, N represents the total number of experiments required and n is the number of process variables. Figures 4.1 (a) and (b) represent CCD for optimization of two and three variables, respectively (Bezerra et al., 2008).

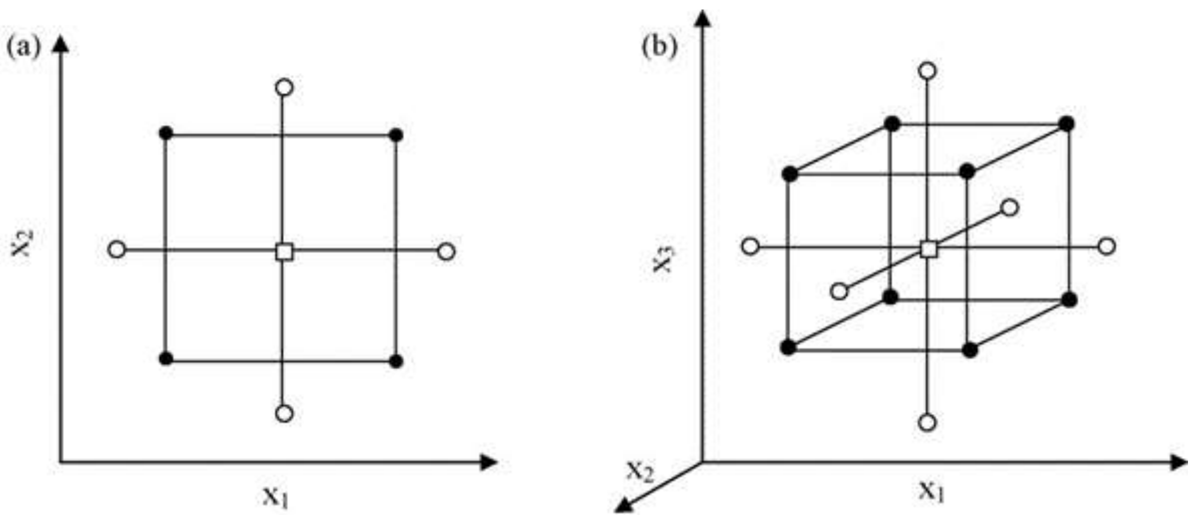

Figure 4.1. Central composite designs for the optimization of (a) two variables (α = 1.41) and (b) three variables (α = 1.68). (●) Points of factorial design, (○) axial points and (□) central point.

A quadratic polynomial regression model was used to evaluate and quantify the influence of the variables on the responses obtained after the experiments:

$$Y = b_0 + \sum_{i=1}^{k} b_i X_i + \sum_{i=1}^{k} b_{ii} X_i^2 + \sum_{i=1}^{k} \sum_{j=1}^{k} b_{ij} X_i X_j + \mathcal{E} \quad (4.2)$$

where, Y is the response (predicted); k is the number of patterns; X_i and X_j are the coded levels of input factors; b_0 is an intercept; b_i is the linear coefficient; b_{ii} is the regression coefficient for squared effects; b_{ij} is the interaction coefficient; X_i X_j represent interactions of input factors, while X_i^2 represent quadratic terms, i and j are the index numbers for factor and $\mathcal{E}$ is the residual error associated with the experiments (Seres et al., 2016). Equation (4.3) was used to convert the actual value of an independent variable (X_i) to its coded form (χ_i) for calculating statistical value:

$$\chi_i = \frac{(X_i - X_0)}{\delta X} \quad (4.3)$$

where, χ_i, X_i, X_0 and $\boldsymbol{\delta} X$ are the coded values of i^{th} independent variable, actual value, actual value at the center point and the step change of the i^{th} variable, respectively.

4.1.2 Analysis of variance

Data obtained from CCD was analysed using analysis of variance. Analysis of variance (ANOVA) is similar to regression in that it is used to investigate and model. The relationship between a response variable and one or more independent variables produce the analysis of variance for the selected model, and test the significance and the adequacy of the model (Rodrigues et al., 2009). The ability of the quadratic polynomial model was expressed on the basis of coefficients of determination (regression coefficients) R^2, adjusted R^2 and predicted R^2. The R^2 coefficient represents the fitness of polynomial models. The statistical significance of the quadratic model was verified with Fisher variation ratio (F-value), adequate precision and the probability value (Prob > F) with 95% confidence level using Equations (4.4) and (4.5):

$$Adequate\ Precision = \frac{\max(Y)-\min(y)}{\sqrt{\bar{V}(Y)}} \quad (4.4)$$

$$\bar{V}(Y) = \frac{1}{n}\sum_{i=1}^{n}\bar{V}\,(Y) = \frac{p\sigma^2}{n} \quad (4.5)$$

where, p and n are number of experiments and quadratic model parameters, respectively and σ^2 is the residual mean square (Pouran et al., 2015).

4.2 Modeling Uses Adaptive Neuro-Fuzzy Inference System

The adaptive neuro-fuzzy inference system (ANFIS) is a type of artificial neural network that is based on Takagi–Sugeno fuzzy inference system. It is a branch of artificial intelligence (AI) and is a blend of artificial neural networks (ANN) and fuzzy systems. Therefore, offers the advantages of both. Fuzzy technology has been used to reduce the complexity of data to an acceptable degree usually either via linguistic variables or via fuzzy data analysis and can be used as a tool for modeling, problem solving and data mining (Khoshnevisan et al., 2014). The theory of fuzzy sets has already been applied successfully to a number of operations research problems.

Recently, Mandal et al. (2015) used the neuro fuzzy approach for arsenic(III) and chromium(VI) removal from water. Other researchers have used the same model for prediction of effluent quality of a paper mill wastewater treatment unit (Wan et al., 2011). In another study Pai et al. (2011) used three types of ANFIS and ANN to predict the effluent from a wastewater treatment plant in the industrial park. Elmolla et al. (2010) used ANN for modeling of COD removal from antibiotic aqueous solution by the Fenton process. Pai et al. (2009) employed three types of ANFIS and ANN to predict the effluent from a hospital wastewater treatment plant. Mingzhi et al. (2009) presented a fuzzy neural network predictive control scheme for studying the coagulation process of wastewater treatment in a paper mill.

4.2.1 ANFIS Architecture

The results of the RSM were used as the training data in the ANFIS network. ANFIS is a type of artificial neural network that is based on Takagi–Sugeno fuzzy inference system. The MATLAB software (R2013a (8.1.0.604)) with neuro-fuzzy design was used for chlorpyrifos reduction prediction in Fenton process. Fuzzy technology involves fuzzy logic operations on the concept of membership function. The “degree of membership” can be invented via membership function in the real continuous interval [0,1]. The fuzzy system is directed by fuzzy *if-then* rules and a set of fuzzy rules converts the input set to the output set (Zimmermann, 2012). For convenience, it is assumed that a simplified framework of ANFIS under consideration has two inputs, x, y, two rules and one output, z. The rule base contains two fuzzy *if-then* rules for Takagi–Sugeno fuzzy model is as follows (Mingzhi et al., 2009):

If x is A and y is B, then z is $f(x,y)$ (4.6)

where, A and B are the fuzzy sets and $f(x,y)$ is usually a polynomial for the input variables x and y. When the value of $f(x,y)$ becomes constant, a zero order Sugeno fuzzy model is formed.

If $f(x,y)$ is taken to be a first order polynomial, a first-order Sugeno fuzzy model can be formed with two fuzzy if–then rules and can be expressed as (Mandal et al., 2015):

Rule 1: If x is A_1 and y is B_1, then $f_1 = m_1x + n_1y + r_1$ (4.7)

Rule 2: If x is A_2 and y is B_2, then $f_2 = m_2x + n_2y + r_2$ (4.8)

where, A_1, A_2, B_1 and B_2 are the fuzzy sets for the inputs x and y, respectively, and $\{m_i, n_i, r_i\}$and $\{m_j, n_j, r_j\}$ (i, j = 1, 2) are the design constraints that are determined during the training process so-called consequent parameters. As can be seen the frameworks of ANFIS as revealed in Figure 4.2 consists of five layers, which perform different actions, but the nodes of the same layer have similar functions and are detailed below. Each input corresponds to two associated membership functions (MFs). The functions of individual layers of this ANFIS structure are detailed below in Equations (4.9) through (4.16):

Layer 1: Input nodes

Every node in this layer is an adaptive node, which represents a membership grade of a linguistic label. The outputs of this layer in terms of membership functions can be described as:

$$O_{Ai}^{1} = \mu_{Ai}\,(x), \quad i = 1, 2 \tag{4.9}$$

$$O_{Bj}^{1} = \mu_{Bj}\,(x), \quad j = 1, 2 \tag{4.10}$$

where, x is the input to node i and y is the input to node j, and Ai and Bj are linguistic labels (low, medium, high) characterized by appropriate MFs, μ_{Ai} is the membership functions of A_1 and μ_{Bj} is the membership functions of B_1. In this study, the generalized bell-shaped fuzzy MFs are chosen as ($\mu_{Ai}(x)$ and $\mu_{Bj}(y)$) with maximum equal to1 and minimum equal to 0, which are defined below as:

$$\mu_{Ai}(x) = \frac{1}{1+\left(\frac{x-c_i}{a_i}\right)^{2bi}}, \quad i = 1, 2 \tag{4.11}$$

$$\mu_{Bj}(x) = \frac{1}{1+\left(\frac{x-c_j}{a_j}\right)^{2bj}}, \quad j = 1, 2 \tag{4.12}$$

where, {ai, bi, ci} and {aj, bj, cj} are the adaptive parameters related to the input MFs, so-called premise parameters.

Layer 2: Rule nodes

The node in this layer is a fixed node which provides the firing strength w_i of the rule via multiplication. Every single node represents the antecedent part of the rule (the *if* part of the rule is called the antecedent). The output of this layer is the multiplication of all the incoming signals and is represented as:

$$O_{ij}^{2} = w_{ij} = \mu_{Ai}(x) \times \mu_{Bj}(y), \qquad i,j = 1,2 \tag{4.13}$$

Layer 3: Normalized layer

Each node in this layer is a fixed node and they all play a normalization role in the network, which is labelled as N. The i^{th} node calculates the ratio of the i^{th} rules firing strength to the sum of all rules firing strengths:

$$O_{ij}^{3} = \overline{w_{ij}} = \frac{w_{ij}}{w_{11}+w_{12}+w_{21}+w_{22}}, \qquad i,j = 1,2 \tag{4.14}$$

The output from the i^{th} node is called normalized firing strength.

Layer 4: Consequent nodes

The role of this layer is the defuzzyfication and each node in this layer is an adaptive node. The output of this is simply the multiplication of the normalized firing strength with the function of Sugeno fuzzy rule is given by Equation (4.15):

$$O_{ij}^{4} = \overline{w_{ij}} f_{ij} = \overline{w_{ij}}\left(m_{ij}x + n_{ij} + r_{ij}\right), \quad i,j = 1,2 \tag{4.15}$$

where, w_{ij} is the output of Layer 3 and $\{m_{ij}, n_{ij}, r_{ij}\}$ is the parameter set are referred to as consequent parameters.

Layer 5: Output nodes

There is only one fixed node labelled Σ in this layer, which computes the sum of all outputs of each rule incoming from the previous layer, thus we have:

$$z = O_{1}^{5} = \sum_{i=1}^{2}\sum_{j=1}^{2} \overline{w_{ij}}\, f_{ij} = \sum_{i=1}^{2}\sum_{i=2}^{2} \overline{w_{ij}}\left(m_{ij}x + n_{ij}y + r_{ij}\right)$$

$$= \sum\sum[(\overline{w_{ij}}\, x)m_{ij} + (\overline{w_{ij}}\, y)n_{ij} + (\overline{w_{ij}})r_{ij}] \tag{4.16}$$

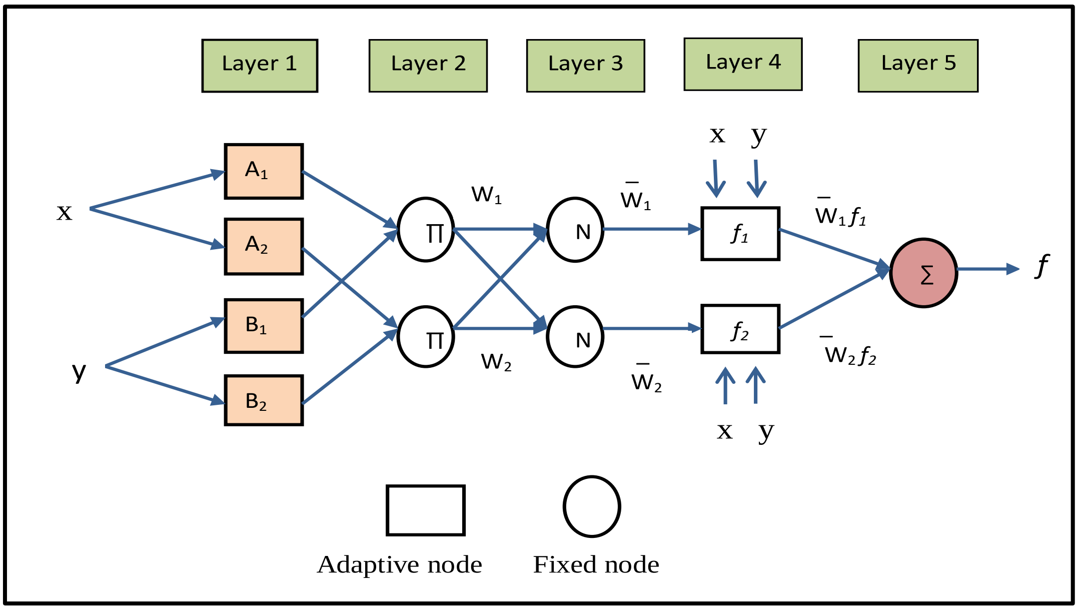

Figure 4.2: Basic architecture of ANFIS for two inputs with four rules.

4.2.2. The index

The performance of the ANFIS model was appraised by the correlation coefficient (R), mean square error (MSE) and root mean square error (RMSE). The average value of the squares of the errors is determined by MSE (Pai et al., 2009; Wan et al., 2011). The MSE is defined as:

$$MSE = \frac{1}{n}\sum_{i=1}^{n}(q_e - q_p)^2 \qquad (4.17)$$

where, q_e and q_p is the measured and predicted values and n is the number of experiments.

RMSE to regulate the inaccuracy between the simulated and training data was defined as:

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(q_e - q_p)^2}{n}} \qquad (4.18)$$

The CCD (Trinh and Kang, 2011; Torrades and Garcia-Montano, 2014; Gatsios et al., 2015;) and ANFIS (Mingzhi et al., 2009; Elmolla et al., 2010; Pai et al., 2011) have been applied for the optimization of several treatment technology.